Table III. Integral Molar Quantities for Solid Alloys at 1173 K[a]

x_{Pt}	ΔG^E/(J mol^{-1})	ΔH^M/(J mol^{-1})	ΔS^E/ (J K^{-1} mol^{-1})
0.0	0 ± 0	0 ± 0	0 ± 0
0.1	−4 230 ± 100	−3300 ± 800	0.8 ± 0.6
0.2	−7 500 ± 200	−5800 ± 1500	1.4 ± 1.1
0.3	−10 000 ± 290	−7800 ± 2000	1.9 ± 1.5
0.4	−11 690 ± 370	−9200 ± 2300	2.2 ± 1.7
0.5	−12 470 ± 430	−9800 ± 2500	2.3 ± 1.8
0.6	−12 160 ± 430	−9600 ± 2400	2.2 ± 1.7
0.7	−10 430 ± 380	−8200 ± 2100	1.9 ± 1.5
0.8	−7 510 ± 300	−5800 ± 1600	1.4 ± 1.1
0.9	−3 930 ± 170	−3000 ± 900	0.8 ± 0.6
1.0	0 ± 0	0 ± 0	0 ± 0

[a] $(1-x)\mathrm{Co}_{(s)} + x\mathrm{Pt}_{(s)} = \mathrm{Co}_{(1-x)}\mathrm{Pt}_{x(s)}$.

and obtain the values of the integral function $\Delta G^E(1173) = (1 - x_{Co})I(x_{Co})$ and the partial function of platinum

$$\Delta G_{Pt}{}^E(1173) = [\Delta G^E(1173) - x_{Co}\Delta G_{Co}{}^E(1173)]/(1 - x_{Co})$$

for various values of the atomic fraction x_{Co}. From this, we obtain γ_{Co}, a_{Co}, γ_{Pt}, and a_{Pt} at 1173 K. The results of these calculations are shown in Tables I and II.

To calculate the mixture properties of cobalt-platinum alloys at any given temperature T between 1000 and 1400 K, we take values of $\Delta G_{Co}{}^E(1173)$ and $\Delta G_{Pt}{}^E(1173)$ from Table I and calculate

$$\Delta G_{Co}{}^E(T,x_{Co}) = \Delta G_{Co}{}^E(1173,x_{Co}) - (T - 1173)\Delta S_{Co}{}^E(x_{Co})$$

with

$$\Delta S_{Co}{}^E(x_{Co}) = (9 \pm 7)(1 - x_{Co})^2 \text{ J K}^{-1} \text{ mol}^{-1}$$

By replacing the subscript Co by Pt, we obtain an analogous expression for platinum.

Finally we may note that the results shown on Tables I and II allow the partial molar enthalpy of mixing of cobalt and platinum between 1000 and 1400 K to be calculated since

$$\Delta H_{Co}{}^M = \Delta H_{Co}{}^E = \Delta G_{Co}{}^E(1173) + 1173\Delta S_{Co}{}^E$$

and a similar expression exists for platinum. The molar integral enthalpy of mixing is very easily obtained since

$$\Delta H^M = x_{Co}\Delta H_{Co}{}^M + x_{Pt}\Delta H_{Pt}{}^M$$

Table III shows the results of these calculations.

We can see that the integral heat of mixing is not very far from the results (*3*, *4*) mentioned in section 2 of this work.

Uncertainties were given for Gibbs energies and for entropies from the experimental dispersion of Miner's results, as explained in section 4. Uncertainties of enthalpies are issued from the relation $H = G + TS$.

6. Conclusion

The results of this critical evaluation are summarized in three tables. Tables I–III give respectively partial quantities of cobalt, partial quantities of platinum, and integral quantities: activities, activity coefficients, excess Gibbs energies, enthalpies of mixing, and excess entropies. These quantities are given at 1173 K but lead to activity and Gibbs energies between 1000 and 1400 K with enthalpies and entropy independent of temperature and Gibbs energy given by $\Delta G = \Delta H - T\Delta S$, for integral and partial quantities.

No new information was obtained from experiments in low-temperature data or in liquid alloys, so that Hultgren's compilation does not need modification in these domains.

Glossary

a_i	activity with respect to solids elements i
emf	electromotive force
ΔG^E	excess molar Gibbs energy
ΔH^M	enthalpy of mixing
R	perfect gas constant (8.31441 J K^{-1} mol^{-1})
ΔS^E	excess molar entropy
T	thermodynamic temperature in Kelvin
x_i	mole fraction of element i

Greek Letters

γ_i	activity coefficient of element i

Subscripts

Co	partial quantities of cobalt
Pt	partial quantities of platinum

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Received for review February 19, 1980. Accepted October 24, 1980.

Excess Thermodynamic Functions for Ternary Systems. 7. Total Pressure Data and G^E for Acetone/1,4-Dioxane/Water at 50 °C

Joseph R. Loehe, Hendrick C. Van Ness,* and Michael M. Abbott*

Chemical and Environmental Engineering Department, Rensselaer Polytechnic Institute, Troy, New York 12181

Isothermal P–x data for the ternary system acetone/1,4-dioxane/water at 50 °C are reported, together with data for the constituent binaries. Data reduction by Barker's method provides a correlation for G^E.

Reported here are VLE measurements for the acetone (1)/1,4-dioxane (2)/water (3) system at 50 °C. Experimental values of total vapor pressure are presented for the full composition range of the three constituent binaries and for runs with ternary mixtures formed by addition of each pure species to equimolar, 2:1, and 1:2 mixtures of the other species. The

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Table I. Vapor Pressures of Pure Species at 50 °C in kPa

	acetone (1)	1,4-dioxane (2)	water (3)
present work, av value	81.830	15.684	12.321
lit. values	81.989 (*2*)	15.706 (*5*)	12.345 (*8, 9*)
	81.835 (*3*)	15.908 (*6*)	12.350 (*10*)
	82.018 (*4*)	15.785 (*7*)	12.347 (*5*)

Table II. *P*-*x* Data for Acetone (1)/Water (3) at 50 °C

x_1	x_3	P, kPa	x_1	x_3	P, kPa
0.0	1.0000	12.313	0.5017	0.4983	70.932
0.0347	0.9653	32.624	0.5169	0.4831	71.216
0.0598	0.9402	41.780	0.5706	0.4294	72.298
0.1024	0.8976	51.632	0.6248	0.3752	73.408
0.1518	0.8482	58.041	0.6796	0.3204	74.575
0.2025	0.7975	61.897	0.7371	0.2629	75.862
0.2531	0.7469	64.422	0.7930	0.2070	77.178
0.3028	0.6972	66.184	0.8497	0.1503	78.576
0.3522	0.6478	67.606	0.9003	0.0997	79.798
0.4018	0.5982	68.803	0.9505	0.0495	80.919
0.4519	0.5481	69.900	1.0000	0.0	81.822
0.4637	0.5363	70.147			

Table III. *P*-*x* Data for 1,4-Dioxane (2)/Water (3) at 50 °C

x_2	x_3	P, kPa	x_2	x_3	P, kPa
0.0000	1.0000	12.337	0.4761	0.5239	22.010
0.0508	0.9492	16.266	0.4995	0.5005	22.035
0.1013	0.8987	18.543	0.5015	0.4985	21.977
0.1518	0.8482	19.906	0.5022	0.4978	22.001
0.1519	0.8481	19.911	0.5344	0.4656	22.045
0.2014	0.7986	20.721	0.5488	0.4512	22.054
0.2021	0.7979	20.719	0.5918	0.4082	22.049
0.2517	0.7483	21.229	0.5984	0.4016	22.049
0.2522	0.7478	21.223	0.6483	0.3517	22.005
0.3019	0.6981	21.540	0.6501	0.3499	22.007
0.3020	0.6980	21.547	0.6984	0.3016	21.908
0.3519	0.6481	21.735	0.7006	0.2994	21.895
0.3522	0.6478	21.747	0.7312	0.2688	21.781
0.4021	0.5979	21.864	0.7789	0.2211	21.509
0.4022	0.5978	21.866	0.8590	0.1410	20.663
0.4519	0.5481	21.954	0.9495	0.0505	18.304
0.4523	0.5477	21.968	1.0000	0.0	15.678

Table IV. *P*-*x* Data for Acetone (1)/1,4-Dioxane (2) at 50 °C

x_1	x_2	P, kPa	x_1	x_2	P, kPa
0.0	1.0000	15.682	0.5030	0.4970	51.677
0.0293	0.9707	18.451	0.5051	0.4949	51.708
0.0515	0.9485	20.543	0.5460	0.4540	54.294
0.0552	0.9448	20.762	0.5504	0.4496	54.528
0.1010	0.8990	24.634	0.5556	0.4444	54.765
0.1043	0.8957	24.907	0.6009	0.3991	57.536
0.1452	0.8548	28.087	0.6062	0.3938	57.799
0.1547	0.8453	28.768	0.6507	0.3493	60.517
0.1917	0.8083	31.530	0.6564	0.3436	60.795
0.2074	0.7926	32.607	0.7007	0.2993	63.499
0.2399	0.7601	34.916	0.7065	0.2935	63.809
0.2566	0.7434	36.000	0.7487	0.2513	66.358
0.2878	0.7122	38.137	0.7502	0.2498	66.472
0.3060	0.6940	39.328	0.7985	0.2015	69.374
0.3365	0.6635	41.310	0.8000	0.2000	69.466
0.3550	0.6450	42.485	0.8481	0.1519	72.405
0.3852	0.6148	44.419	0.8496	0.1504	72.498
0.4046	0.5954	45.615	0.8984	0.1016	75.495
0.4343	0.5657	47.480	0.8998	0.1002	75.558
0.4506	0.5494	48.467	0.9488	0.0512	78.658
0.4546	0.5454	48.637	0.9493	0.0507	78.631
0.4533	0.5467	48.650	0.9750	0.0250	80.254
0.4844	0.5156	50.546	1.0000	0.0	81.827
0.5005	0.4995	51.509			

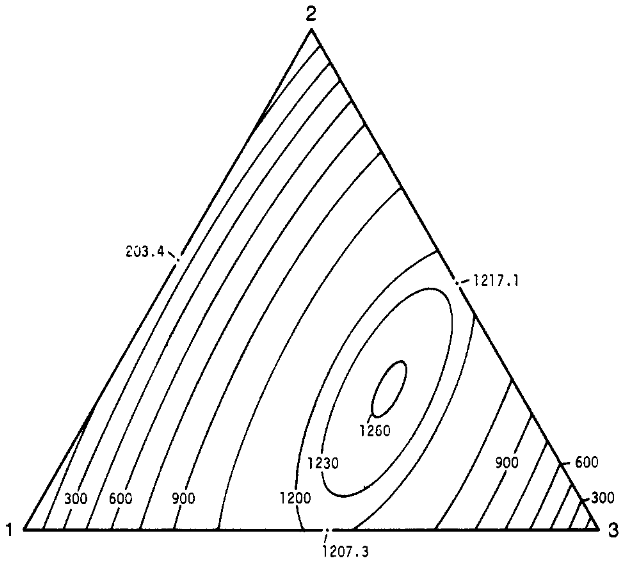

Figure 1. Lines of constant G^E (J/mol) for the acetone (1)/1,4-dioxane (2)/water (3) system at 50 °C.

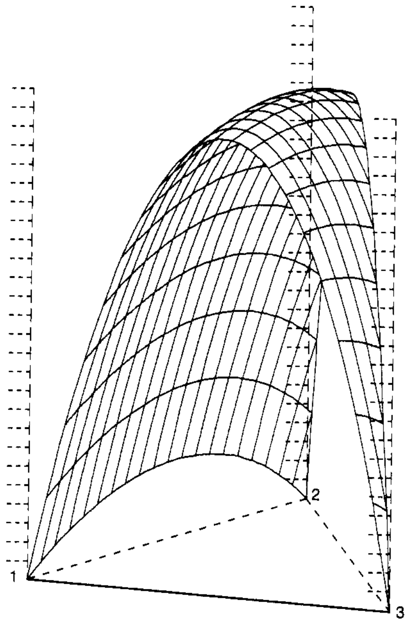

Figure 2. Pictorial view of the G^E–x surface for the acetone (1)/1,4-dioxane (2)/water (3) system at 50 °C.

apparatus is that of Gibbs and Van Ness (*1*) as modified by DiElsi et al. (*2*).

Acetone and dioxane were obtained as chromatoquality reagents from Matheson Coleman and Bell with indicated purities of at least 99.96 mol %. The water was doubly deionized. Except for degassing, all reagents were used as received. Six or more measurements of the vapor pressure of each pure species were made during the course of this work, and average values are compared with some literature values in Table I.

Results and Correlations

Tables II–IV give experimental values of total pressure for the three constituent binary systems, and Table V displays all data for runs made with ternary mixtures. Data reduction is by Barker's method according to procedures described earlier (*11*, *12*). For the binary systems, the Margules equation with up to five parameters provides suitable expression of G^E:

$$g_{ij} \equiv G_{ij}^E/(RT) =$$
$$[A_{ji}x_i + A_{ij}x_j - (\lambda_{ji}x_i + \lambda_{ij}x_j)x_ix_j + \eta(x_ix_j)^2]x_ix_j \quad (1)$$

For the acetone (1)/water (3) system, correlation requires the full five parameters, but for acetone (1)/1,4-dioxane (2) and 1,4-dioxane (2)/water (3)

$$\eta = 0 \qquad \lambda_{ji} = \lambda_{ij}$$

Data for the ternary system are well correlated by the

Table V. P-x Data for Acetone (1)/1,4-Dioxane (2)/Water (3) at 50 °C

x_1	x_2	x_3	P, kPa	x_1	x_2	x_3	P, kPa
1.0000	0.0	0.0	81.837	1.0000	0.0	0.0	81.827
0.4992	0.5008	0.0	51.396	0.3335	0.0	0.6665	67.035
0.4750	0.4765	0.0484	51.854	0.3168	0.0506	0.6326	62.345
0.4507	0.4521	0.0972	51.860	0.3001	0.1010	0.5989	58.150
0.4260	0.4273	0.1467	51.588	0.2833	0.1516	0.5650	54.415
0.4014	0.4026	0.1959	51.238	0.2667	0.2016	0.5316	51.107
0.3764	0.3775	0.2461	50.805	0.2501	0.2515	0.4983	48.162
0.3512	0.3522	0.2966	50.323	0.2334	0.3017	0.4649	45.476
0.3260	0.3270	0.3470	49.805	0.2168	0.3516	0.4316	43.047
0.3009	0.3018	0.3973	49.298	0.2002	0.4015	0.3983	40.805
0.0	1.0000	0.0	15.705	1.0000	0.0	0.0	81.807
0.4997	0.5003	0.0	51.383	0.6663	0.0	0.3337	74.248
0.4748	0.4754	0.0498	51.844	0.6326	0.0506	0.3168	71.005
0.4495	0.4501	0.1003	51.827	0.5990	0.1011	0.2999	67.901
0.4246	0.4251	0.1503	51.557	0.5655	0.1515	0.2830	64.835
0.3997	0.4002	0.2000	51.156	0.5320	0.2018	0.2662	61.927
0.3748	0.3753	0.2499	50.704	0.4943	0.2584	0.2473	58.735
0.3499	0.3503	0.2998	50.203	0.4570	0.3144	0.2286	55.666
0.3248	0.3252	0.3501	49.700	0.4332	0.3502	0.2166	53.731
0.2999	0.3002	0.3999	49.208	0.3999	0.4002	0.1999	51.052
0.0	1.0000	0.0	15.696	1.0000	0.0	0.0	81.839
0.0	0.5001	0.4999	21.937	0.6663	0.3337	0.0	61.419
0.0510	0.4746	0.4744	27.746	0.5900	0.2944	0.1155	60.976
0.1008	0.4498	0.4495	32.660	0.5526	0.2765	0.1709	60.305
0.1505	0.4249	0.4246	36.985	0.5156	0.2581	0.2263	59.598
0.2002	0.4000	0.3998	40.919	0.4793	0.2400	0.2806	58.860
0.2515	0.3744	0.3741	44.625	0.4433	0.2220	0.3347	58.155
0.3016	0.3493	0.3491	47.973	0.4081	0.2043	0.3876	57.458
0.3519	0.3242	0.3239	51.117	0.3734	0.1869	0.4397	56.798
0.4016	0.2993	0.2991	54.011	1.0000	0.0	0.0	81.835
0.0	1.0000	0.0	15.692	0.3329	0.6671	0.0	41.073
0.0	0.6663	0.3337	21.931	0.3166	0.6344	0.0489	42.005
0.0500	0.6330	0.3170	26.693	0.3002	0.6015	0.0982	42.364
0.1006	0.5993	0.3001	30.991	0.2838	0.5686	0.1476	42.381
0.1568	0.5619	0.2814	35.384	0.2672	0.5352	0.1976	42.196
0.2073	0.5282	0.2645	39.029	0.2504	0.5016	0.2479	41.947
0.2581	0.4943	0.2476	42.480	0.2400	0.4810	0.2790	41.749
0.3073	0.4615	0.2311	45.644	0.2165	0.4337	0.3498	41.275
0.3570	0.4284	0.2145	48.715	0.1991	0.3989	0.4020	40.860
0.4059	0.3959	0.1983	51.588	0.0	0.0	1.0000	12.309
1.0000	0.0	0.0	81.837	0.0	0.3341	0.6659	21.653
0.4981	0.0	0.5019	70.765	0.0497	0.3175	0.6328	29.028
0.4732	0.0503	0.4766	67.063	0.1032	0.2997	0.5972	35.431
0.4484	0.1003	0.4514	63.583	0.1516	0.2835	0.5649	40.304
0.4235	0.1503	0.4262	60.265	0.2004	0.2672	0.5324	44.520
0.3987	0.2003	0.4011	57.221	0.2495	0.2508	0.4997	48.228
0.3739	0.2500	0.3760	54.337	0.2988	0.2343	0.4669	51.510
0.3490	0.3001	0.3509	51.582	0.3487	0.2177	0.4337	54.522
0.3242	0.3500	0.3258	48.988	0.3983	0.2011	0.4006	57.254
0.2993	0.4000	0.3007	46.513				

Table VI. Summary of Results for Binary Systems at 50 °C[a]

	acetone (1)/ 1,4-dioxane (2)	acetone (1)/ water (3)	1,4-dioxane (2)/ water (3)
P_i^{sat}, kPa	81.830	81.830	15.684
P_j^{sat}, kPa	15.684	12.321	12.321
V_i^L, cm³/mol	76.83	76.83	88.12
V_j^L, cm³/mol	88.12	18.23	18.23
B_{ii}, cm³/mol	−1450	−1450	−1380
B_{jj}, cm³/mol	−1380	−1120	−1120
B_{ij}, cm³/mol	−730	−870	−450
A_{ij}	0.3627 ± 0.0026	2.2767 ± 0.0022	1.9702 ± 0.0038
A_{ji}	0.2841 ± 0.0015	1.7251 ± 0.0044	1.8976 ± 0.0037
λ_{ij}	0.08756 ± 0.0067	1.3226 ± 0.0210	0.4876 ± 0.0139
λ_{ji}	0.08756 ± 0.0067	0.6172 ± 0.0334	0.4876 ± 0.0139
η		0.5489 ± 0.0635	
rms δP, kPa	0.035	0.033	0.019
max $\lvert\delta P\rvert$, kPa	0.089	0.058	0.051

[a] Pairs of components are listed in the order i, j.

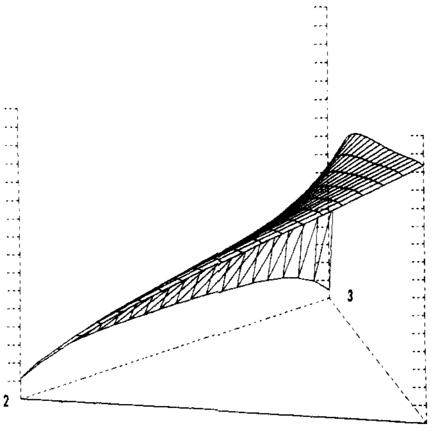

Figure 3. Pictorial view of the P–x surface for the acetone (1)/1,4-dioxane (2)/water (3) system at 50 °C.

three-parameter Wohl equation:

$$g_{123} = g_{12} + g_{13} + g_{23} + (C_0 + C_1x_1 + C_2x_2)x_1x_2x_3 \quad (2)$$

Correlations for the g_{ij} are given by eq 1; parameters C_0, C_1, and C_2 result from regression of just the ternary data.

Second virial coefficients B_{ij} required for estimation of vapor-phase nonidealities come from the correlation of Hayden and O'Connell (*13*).

Results of correlation for the binary systems, together with all ancillary information, are summarized in Table VI. With parameters for the binary systems fixed at the values of Table VI, the ternary-mixture data are correlated when the parameters of eq 2 have the values $C_0 = 2.460 \pm 0.032$, $C_1 = 1.396 \pm 0.035$, and $C_2 = 1.377 \pm 0.061$. The root-mean-square (rms) value of δP for the ternary data is 0.053 kPa; the maximum $|\delta P|$ is 0.181 kPa.

Discussion

No previous work comparable with ours appears in the literature for the ternary system or for the acetone/1,4-dioxane binary. The data of Chaudhry et al. (*4*) for acetone/water at 50 °C and the data of Balcázar-Ortiz et al. (*5*) for 1,4-dioxane/water at 50 °C are both nearly as well correlated by our equations for G^E as are our own data sets. The rms δP in each case is ~0.03 kPa.

The results of this study appear pictorially in Figures 1–3. Figure 1 is a contour diagram showing lines of constant G^E on a triangular mole-fraction grid. Figure 2 is an oblique view of the same surface, and Figure 3 is a similar representation of the $P-x$ surface. There is no ternary azeotrope and only one binary azeotrope: for 1,4-dioxane/water at $x_{\text{dioxane}} = 0.5664$ and $P^{az} = 22.040$ kPa.

Glossary

A_{ij}, A_{ji}	parameters in eq 1
B_{ij}	second virial coefficient
C_0, C_1, C_2	parameters in eq 2
G^E	excess Gibbs function, liquid phase
g	$G^E/(RT)$
P	total pressure
P_i^{sat}	vapor pressure of pure i
R	universal gas constant
T	absolute temperature
V_i^L	molar volume of pure liquid i
x	mole fraction, liquid phase

Greek Letters

λ_{ij}, λ_{ji}, η	parameters in eq 1
δ	denotes the difference, calculated – experimental

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Received for review August 18, 1980. Accepted November 17, 1980. Support for this work came from National Science Foundation Grant No. CPE78-10048.

Solubility of Benzoic Acid in Aqueous Polymeric Solutions

Haribansh Sahay, Surendra Kumar, Siddh N. Upadhyay,*[†] and Yogendra D. Upadhya

Department of Chemical Engineering and Technology, Institute of Technology, Banaras Hindu University,
Varanasi-221005, India

Solubility of benzoic acid in distilled water and aqueous solutions of poly(vinyl alcohol) (PVA) and poly(vinylpyrrolidone) (PVP) containing 0.5, 1.0, 2.0, 3.0, 4.0, 5.0, and 6.0% polymer by weight has been determined in the temperature range of 30–55 °C. It has been observed that, at any given temperature, the solubility in polymer solution is more than that in water, increases with increasing polymer concentration, and approaches a constant value at higher concentrations.

Introduction

Solid–liquid mass-transfer investigations with both Newtonian and non-Newtonian fluids are frequently made by following the rate of dissolution of a low-solubility solute such as benzoic acid because of many inherent advantages (*1*) In all such studies, accurate solubility data are required. This communication reports the solubility of benzoic acid in distilled water and a number of aqueous solutions of poly(vinyl alcohol) (PVA) and poly(vinylpyrrolidone) (PVP) over a polymer concentration range of 0.5–6.0% polymer by weight. Besides applications in the above area, these data will also be useful to the pharmaceutical industry where benzoic acid, PVA, and PVP are used in many pharmacuetical preparations.

Experimental Section

Materials. Conductivity-grade distilled water was prepared in the laboratory. Poly(vinyl alcohol) (mol wt = 14000; natural granules; viscosity of 4% aqueous solution at 20 °C, 4–6 cP; degree of hydrolysis, 98.5–100%; residual monomer content, 0–3%; product number, 30573) was obtained from BDH Chemicals Ltd., (Poole, England) and poly(vinylpyrrolidone) (PVP) (mol wt = 25000; natural powder; residual monomer content, 12.6 ± 0.3%; product number, 39579A; batch number,

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[†] Present address: Department of Energy Engineering, University of Illinois at Chicago Circle, Chicago, IL 60680.

0021-9568/81/1726-0181$01.25/0