Densities, Viscosities, and Surface Tensions for the Two Ternary Systems H_2O + LiBr + LiI and H_2O + LiCl + $LiNO_3$

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Densities, viscosities, and surface tensions for the H_2O + LiBr + LiI (salt mole ratio 4:1) and the H_2O + LiCl + $LiNO_3$ (salt mole ratio 2.8:1) systems were measured at various temperatures and absorbent concentrations. Polynomial equations for densities, viscosities, and surface tensions of these two ternary systems were obtained as a function of absolute temperature by a least-squares method from the individual experimental data. The average absolute deviations between the measured density, viscosity, and surface tension data and the individual calculated values from these polynomial equations were 0.19, 1.66, and 0.57% for the H_2O + LiBr + LiI system and 0.14, 0.54, and 0.24% for the H_2O + LiCl + $LiNO_3$ system, respectively.

Introduction

Accurate and extensive thermodynamic (heat capacity, heat of mixing, vapor pressure, solubility, density, and surface tension) and transport (viscosity) properties of working fluids are required for the research and the reliable design of absorption refrigeration and heat pump systems. The density (*1–15*), viscosity (*4–17*), and surface tension (*11–15, 18, 19*) data are important basic properties to analyze heat and mass transfers of working fluids in the systems. The viscosity data are essential to examine fluidity for working fluids in the systems. The surface tension affects the transfer rates of vapor absorption where a vapor–liquid interface exists. The surface tension data are also indispensable for clarification of the absorption mechanism where the working medium vapor is absorbed absorbent solution in the absorber.

The H_2O + LiBr + LiI system was proposed in order to improve the performance characteristics of the basic H_2O + LiBr system (*11*). And the H_2O + LiCl + $LiNO_3$ system was also proposed in order to improve the performance characteristics and to reduce the corrosion caused by the basic H_2O + LiBr system (*11, 20*) at the same time. In our previous papers, the optimum mixing ratios (*21, 22*) of absorbents used for these two ternary systems and vapor pressure (*22, 23*), heat capacity (*21, 24*), heat of mixing (*25*), and solubility (*26*) data for the individual absorbent solutions at the optimum mixing ratios were reported.

The densities, viscosities, and surface tensions for these two ternary systems were measured over wide ranges of temperatures and absorbent concentrations. Polynomial equations for these two ternary systems were obtained as a function of absolute temperature by a least-squares method from the individual measured density, viscosity, and surface tension data.

Experimental Section

Materials. The LiBr, LiI, and LiCl used in this work were from the Honjo Chemical Co., Ltd. (Japan), analytical reagent grade with a minimum purity of 99.9 mass %. The analytical results of the aqueous LiBr, LiI, and LiCl solutions have been described in our previous papers (*21–23*). The $LiNO_3$ used in this work was from Wako Pure Chemical Industries Ltd. (Japan), analytical reagent grade with a minimum purity of 99.8 mass %. All the reagents were used without further purification.

The optimum mixing ratios of absorbents for these two ternary systems were determined by measuring the crystallization temperatures of the individual absorbent solutions at constant absorbent concentration. Consequently, the optimum mixing ratios of LiBr and LiI and of LiCl and $LiNO_3$ were found to be 4 and 1 mol and 2.8 and 1 mol, respectively (*21, 22*).

Apparatus and Procedure. In this work, the temperature was measured using a calibrated mercury-in-glass thermometer. The temperature of a constant-temperature water bath used for the density, viscosity, and surface tension measurements was controlled within ±0.01 K. In all cases, double-distilled and degassed water and absorbent solution were kept in the bath for about 30 min to establish the equilibrium condition of temperature. The absorbent concentrations in absorbent solution were determined by Fajans' method (*27*) with use of standardized aqueous $AgNO_3$ solution and dichlorofluorescein as an adsorption indicator. The absorbent solution was titrated by using a microburet of 10-mL total delivery, with divisions of 0.02 mL. All weighings were made on a direct-reading balance (weighing capacity 200 g, reciprocal sensibility 1 mg). The apparatus and the experimental procedures for density, viscosity, and surface tension measurements adopted were checked by using the H_2O + LiBr system (*2, 9, 11, 16, 18, 19*) at various temperatures, and the relative errors between the measured and the published values were less than 1%.

The density measurements were carried out by means of a commercial 50 cm^3 capillary pycnometer of a Gay-Lussac type having a capillary diameter of about 1 mm. The pycnometer was carefully dried, weighed, and then filled with double-distilled and degassed water or absorbent solution and placed in the bath. After thermal equilibrium was reached, the pycnometer was then removed from the bath and weighed again. The internal volumes of the pycnometer were carefully calibrated with water at the eight temperatures of this study using the accurately known densities (*28*). Experiments on internal volumes of the pycnometer were performed in 10 replicates for each temperature, and the results were averaged. Experiments on densities of absorbent solutions were performed in five replicates for each absorbent concentration and at each temperature, and the results were averaged. The differences of the density in repeat runs were smaller than 0.01%.

The viscosity measurements were made by using a commercial capillary viscometer of an Ostwald type having a fine capillary with flared ends in order to minimize kinetic energy corrections. The viscometer was 25 cm in height and had a capillary part of 10 cm in length and an internal diameter of

0021-9568/93/1738-0302$04.00/0

Table I. Densities, Viscosities, and Surface Tensions for the H_2O + LiBr + LiI (LiBr:LiI = 4:1 mol) System at Mass Fraction *w*

$100w$	283.15 K	293.15 K	303.15 K	313.15 K	323.15 K	333.15 K	343.15 K	353.15 K
				$\rho/(\text{kg m}^{-3})$				
10.2	1078	1076	1073	1069	1065	1060	1054	1048
19.5	1162	1158	1155	1151	1146	1141	1135	1129
29.4	1262	1258	1254	1249	1244	1239	1233	1227
40.4	1396	1391	1386	1381	1375	1370	1364	1358
50.6	1556	1550	1544	1539	1533	1526	1520	1514
60.3	1761	1754	1747	1739	1732	1725	1718	1711
65.5		1865	1858	1850	1841	1834	1825	1818
				$\eta/(\text{mPa s})$				
10.2	1.44	1.13	0.90	0.75	0.63	0.54	0.46	0.41
19.5	1.62	1.27	1.02	0.85	0.72	0.62	0.54	0.48
29.4	1.87	1.49	1.21	1.00	0.85	0.72	0.63	0.55
40.4	2.41	1.92	1.57	1.30	1.10	0.95	0.82	0.72
50.6	3.76	2.99	2.43	2.01	1.70	1.45	1.24	1.08
60.3	9.20	7.02	5.51	4.39	3.57	2.96	2.46	2.08
65.5		12.1	9.13	7.03	5.52	4.48	3.64	3.02
				$\sigma/(\text{mN m}^{-1})$				
10.2	75.1	73.7	72.1	70.6	69.1	67.5	65.7	63.9
19.5	77.0	75.7	74.4	72.5	71.6	69.8	67.8	66.3
29.4	78.7	77.8	76.8	75.5	73.9	72.9	71.5	69.9
40.4	82.1	81.3	80.4	79.4	78.3	76.8	75.7	74.3
50.6	85.4	84.7	84.1	83.2	82.3	81.0	79.7	79.0
60.3	88.6	87.8	87.2	86.5	85.8	85.1	84.3	83.7
65.5		91.0	89.9	89.3	88.1	87.0	86.0	85.3

Table II. Densities, Viscosities, and Surface Tensions for the H_2O + LiCl + $LiNO_3$ (LiCl:$LiNO_3$ = 2.8:1 mol) System at Mass Fraction *w*

$100w$	283.15 K	293.15 K	303.15 K	313.15 K	323.15 K	333.15 K	343.15 K	353.15 K
				$\rho/(\text{kg m}^{-3})$				
10.6	1063	1060	1055	1053	1049	1044	1039	1034
21.3	1131	1129	1125	1120	1116	1111	1106	1101
31.1	1197	1193	1190	1185	1181	1176	1171	1166
41.7	1284	1279	1276	1270	1265	1260	1257	1251
48.1	1341	1336	1331	1326	1322	1315	1310	1305
50.1	1361	1356	1351	1346	1340	1336	1330	1325
				$\eta/(\text{mPa s})$				
10.1	1.71	1.32	1.06	0.87	0.73	0.63	0.54	0.48
20.1	2.40	1.85	1.49	1.23	1.04	0.89	0.77	0.68
30.2	3.94	3.02	2.42	1.99	1.67	1.42	1.23	1.08
40.3	8.63	6.44	4.96	3.92	3.20	2.66	2.27	1.95
50.1	30.9	20.1	13.8	9.93	7.44	5.75	4.54	3.66
				$\sigma/(\text{mN m}^{-1})$				
10.6	79.2	78.0	76.5	75.0	73.5	71.9	70.2	68.5
21.3	84.2	83.1	81.8	80.6	79.2	77.7	76.1	74.5
31.1	88.8	87.8	86.8	85.7	84.4	83.0	81.5	79.9
41.7	93.8	92.9	92.1	91.2	90.0	88.8	87.4	85.8
48.1	96.9	95.9	95.2	94.6	93.0	92.6	91.0	89.4

about 1 mm. The viscometer was rinsed with distilled water and dried with acetone at room temperature. The interior glass surface was kept clean during all measurements. The viscometer was clamped vertically in the bath, and an exactly specified amount (10 cm^3) of double-distilled and degassed water or absorbent solution at various absorbent concentrations was added from a pipet. After thermal equilibrium was reached, the liquid was then allowed to flow up through the capillary, and a stopwatch was started when the upper meniscus of the liquid passed the top mark and stopped when it passed the bottom mark. They were determined by observing the top and bottom marks in the viscometer by a cathetometer fitted with a vernier scale, capable of readings to 0.01 mm. The flow time of a constant volume of the liquid through the capillary was measured with an electric digital stopwatch with an accuracy of ±0.01 s. The flow times of absorbent solutions, t, and of water, t_w, were kept relatively long to minimize kinetic energy corrections. Five repetitions of each data set were made to ensure reproducibility, and the results were averaged. The experimental data on water are used in conjunction with those on the absorbent solutions to calculate the viscosity, η, of the absorbent solutions by

$$\eta/\eta_w = \rho t/\rho_w t_w \qquad (1)$$

The viscometer was calibrated by using water of accurately known densities (*28*), ρ_w, and viscosities (*28*), η_w, at the eight temperatures of this study. The errors of the viscosity in repeat runs were smaller than 0.1%.

The surface tension measurements were made by using a capillary-rise method (*29*). The capillary-rise apparatus consisted primarily of a fine capillary tube 30 cm in length and with radius of about 0.5 mm, a glass vessel, a rubber stopper, and a rubber tube. The capillary tube was first carefully cleaned with surfactant, rinsed with distilled water and acetone, and dried under reduced pressure by using an aspirator at room temperature. The assembled apparatus containing the sample solution (60 cm^3) was always kept in a vertical position in the bath. After thermal equilibrium was reached, the liquid in the capillary tube was raised slightly by gently blowing into the rubber tube and was then allowed to fall back to its equilibrium level. It was then depressed by slight suction and again allowed to come to the equilibrium

Table III. Values of Coefficients for H_2O + LiBr + LiI (LiBr:LiI = 4:1 mol) and H_2O + LiCl + $LiNO_3$ (LiCl:$LiNO_3$ = 2.8:1 mol) Systems in Equation 2

	H_2O + LiBr + LiI	H_2O + LiCl + $LiNO_3$
a_0	$7.538\,62 \times 10^{2}$	$7.431\,44 \times 10^{2}$
a_1	$1.184\,88 \times 10$	$1.755\,78 \times 10$
a_2	$1.334\,41 \times 10^{-1}$	$-5.465\,21 \times 10^{-2}$
a_3	$2.221\,76 \times 10^{-6}$	
b_0	$1.883\,11$	$1.952\,52$
b_1	$-2.186\,70 \times 10^{-2}$	$-7.793\,27 \times 10^{-2}$
b_2	$-9.833\,95 \times 10^{-4}$	$6.194\,41 \times 10^{-4}$
b_3	$1.057\,48 \times 10^{-5}$	
c_0	$-3.589\,34 \times 10^{-3}$	$-3.687\,99 \times 10^{-3}$
c_1	$2.743\,89 \times 10^{-5}$	$1.214\,42 \times 10^{-4}$
c_2	$1.710\,70 \times 10^{-6}$	$-1.026\,12 \times 10^{-6}$
c_3	$-1.975\,95 \times 10^{-8}$	

level. The height of the liquid rise in the capillary tube was measured by means of a cathetometer fitted with a vernier scale, capable of readings to 0.01 mm. The radius of the capillary tube was obtained by measuring the height of the capillary rise with double-distilled and degassed water, using the accurately known values of surface tensions (28) at the eight temperatures of this study. Experiments on the height of water or absorbent solution in the capillary tube were performed in five replicates to ensure reproducibility, and the results were averaged. The surface tension values were reproducible within 0.6%.

Results and Discussion

The densities for the H_2O + LiBr + LiI system (salt mole ratio 4:1) were measured in the range of temperatures from 283.15 to 353.15 K and in the range of absorbent concentrations, x, from 10.2 to 65.5 mass % and from 283.15 to 353.15 K and from 10.6 to 50.1 mass % for the H_2O + LiCl + $LiNO_3$ system (salt mole ratio 2.8:1). Only the average values of the densities from five independent measurements for these two ternary systems are presented in Tables I and II. The individual experimental data for these two ternary systems were fitted to polynomial equations as a function of absolute temperature, T, by a least-squares method:

$$\rho/(\mathrm{kg\ m^{-3}}) = \sum_{i=0}^{n} x^i(a_i + b_iT + c_iT^2) \qquad (2)$$

The values of coefficients a_i, b_i, and c_i of the polynomial equations describing densities, ρ, for these two ternary systems are listed in Table III. Some of the experimental data for these two ternary systems are plotted in Figure 1. The solid lines shown in Figure 1 are the calculated values from eq 2 with the coefficients from Table III. As can be seen from Figure 1 and Tables I and II, the densities of the H_2O + LiBr + LiI system are about 10% larger than those of the H_2O + LiCl + $LiNO_3$ system in the range of concentrated absorbent concentrations. The average absolute deviations between the measured density data and the individual calculated values from these polynomial equations were 0.19% for the H_2O + LiBr + LiI system and 0.14% for the H_2O + LiCl + $LiNO_3$ system.

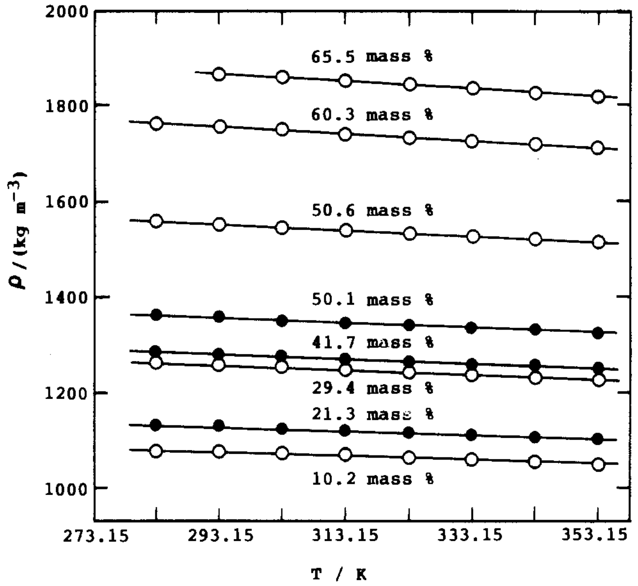

Figure 1. Densities for two ternary systems: ○, H_2O + LiBr + LiI (LiBr:LiI = 4:1 mol); ●, H_2O + LiCl + $LiNO_3$ (LiCl:$LiNO_3$ = 2.8:1 mol); —, calculated values from eq 2.

Table IV. Values of Coefficients for H_2O + LiBr + LiI (LiBr:LiI = 4:1 mol) and H_2O + LiCl + $LiNO_3$ (LiCl:$LiNO_3$ = 2.8:1 mol) Systems in Equation 3

	H_2O + LiBr + LiI	H_2O + LiCl + $LiNO_3$
a_0	$-2.423\,71$	$-1.839\,98$
a_1	$7.310\,65 \times 10^{-4}$	$5.193\,14 \times 10^{-2}$
a_2	$1.961\,51 \times 10^{-5}$	$-8.667\,53 \times 10^{-4}$
a_3	$-8.554\,11 \times 10^{-10}$	$3.585\,41 \times 10^{-5}$
a_4	$8.692\,68 \times 10^{-12}$	$-3.881\,43 \times 10^{-7}$
b_0	$-6.147\,52 \times 10^{2}$	$-9.555\,39 \times 10^{2}$
b_1	$2.232\,30 \times 10$	$-4.882\,68$
b_2	$-9.872\,18 \times 10^{-1}$	$-5.205\,43 \times 10^{-1}$
b_3	$2.860\,81 \times 10^{-2}$	$3.034\,77 \times 10^{-2}$
b_4	$-2.404\,64 \times 10^{-4}$	$-5.211\,96 \times 10^{-4}$
c_0	$3.887\,32 \times 10^{5}$	$4.446\,51 \times 10^{5}$
c_1	$-5.494\,25 \times 10^{3}$	$-1.875\,06 \times 10^{3}$
c_2	$2.843\,76 \times 10^{2}$	$2.980\,75 \times 10^{2}$
c_3	$-8.639\,27$	$-1.310\,60 \times 10$
c_4	$8.291\,13 \times 10^{-2}$	$2.109\,28 \times 10^{-1}$

The viscosity data for these two ternary systems were obtained at the same temperature used for density measurements and at the same absorbent concentration used for density measurements except for that of the H_2O + LiCl + $LiNO_3$ system. The viscosities for the H_2O + LiCl + $LiNO_3$ system were measured in the range of absorbent concentrations, x, from 10.1 to 50.1 mass %. Only the average values of the viscosities from five independent measurements for these two ternary systems are presented in Tables I and II. The individual experimental data for these two ternary systems were fitted to polynomial equations as a function of absolute temperature, T, by a least-squares method:

$$\ln(\eta/(\mathrm{mPa\ s})) = \sum_{i=0}^{4} x^i(a_i + b_i/T + c_i/T^2) \qquad (3)$$

The values of coefficients a_i, b_i, and c_i of the polynomial equations describing viscosities, η, for these two ternary systems are listed in Table IV. Some of the experimental data for these two ternary systems are plotted in Figure 2. The solid lines shown in Figure 2 are the calculated values from eq 3 with the coefficients from Table IV. As Figure 2 shows, the viscosity of the H_2O + LiCl + $LiNO_3$ system is larger than that of the H_2O + LiBr + LiI system at the same temperature and absorbent concentration. The average absolute deviations between the measured viscosity data and the individual calculated values from these polynomial equations were 1.66% for the H_2O + LiBr + LiI system and 0.54% for the H_2O + LiCl + $LiNO_3$ system.

The surface tension data for these two ternary systems were obtained at the same temperature used for density and viscosity measurements and at the same absorbent concentration used for density and viscosity measurements except for that of the H_2O + LiCl + $LiNO_3$ system. The surface tensions for the H_2O + LiCl + $LiNO_3$ system were measured in the range of absorbent concentrations, x, from 10.6 to 48.1 mass %. Only the average values of the surface tensions

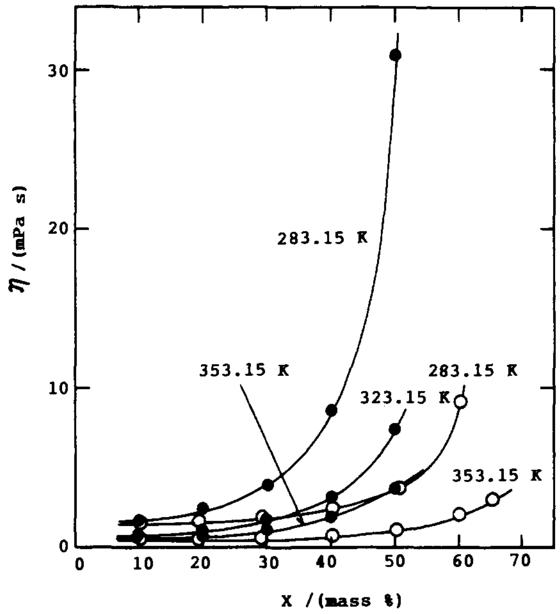

Figure 2. Viscosities for two ternary systems: ○, H_2O + LiBr + LiI (LiBr:LiI = 4:1 mol); ●, H_2O + LiCl + $LiNO_3$ (LiCl:$LiNO_3$ = 2.8:1 mol); —, calculated values from eq 3.

Table V. Values of Coefficients for H_2O + LiBr + LiI (LiBr:LiI = 4:1 mol) and H_2O + LiCl + $LiNO_3$ (LiCl:$LiNO_3$ = 2.8:1 mol) Systems in Equation 4

	H_2O + LiBr + LiI	H_2O + LiCl + $LiNO_3$
a_0	$2.757\,20 \times 10^{2}$	$2.757\,09 \times 10^{2}$
a_1	$1.394\,87 \times 10^{-1}$	$6.521\,36 \times 10^{-1}$
a_2	$-1.781\,54 \times 10^{-3}$	$-2.036\,50 \times 10^{-2}$
a_3	$9.536\,09 \times 10^{-6}$	$6.703\,39 \times 10^{-4}$
a_4	$-1.326\,28 \times 10^{-9}$	$-7.573\,20 \times 10^{-6}$
b_0	$-4.117\,68 \times 10^{-1}$	$-4.118\,02 \times 10^{-1}$
b_1	$-2.702\,67 \times 10^{-4}$	$2.027\,90 \times 10^{-3}$
b_2	$8.021\,48 \times 10^{-5}$	$6.383\,92 \times 10^{-5}$
b_3	$-1.077\,73 \times 10^{-6}$	$-2.013\,88 \times 10^{-6}$
b_4	$6.640\,39 \times 10^{-9}$	$2.269\,11 \times 10^{-8}$

from five independent measurements for these two ternary systems are presented in Tables I and II. The individual experimental data for these two ternary systems were fitted to polynomial equations (*30*) as a function of absolute temperature, T, by a least-squares method:

$$\sigma/(\mathrm{mN\,m^{-1}}) = [\sum_{i=0}^{4} x^i(a_i + b_iT)]^{0.85} \qquad (4)$$

The values of coefficients a_i and b_i of the polynomial equations describing surface tensions, σ, for these two ternary systems are listed in Table V. Some of the experimental data for these two ternary systems are plotted in Figure 3. The solid lines shown in Figure 3 are the calculated values from eq 4 with the coefficients from Table V. As can be seen from Figure 3 and Tables I and II, the surface tensions of the H_2O + LiCl + $LiNO_3$ system are about 10% larger than those of the H_2O + LiBr + LiI system in the range of absorbent concentrations. The average absolute deviations between the measured surface tension data and the individual calculated values from these polynomial equations were 0.57% for the H_2O + LiBr + LiI system and 0.24% for the H_2O + LiCl + $LiNO_3$ system.

The density, viscosity, and surface tension data for these two ternary systems are very useful for the analyses of heat and mass transfers of working fluids in these absorption refrigeration and heat pump systems. In particular, these viscosity and surface tension data are also useful to examine the fluidity for working fluids in the systems and to clarify the absorption mechanism in the absorber, respectively.

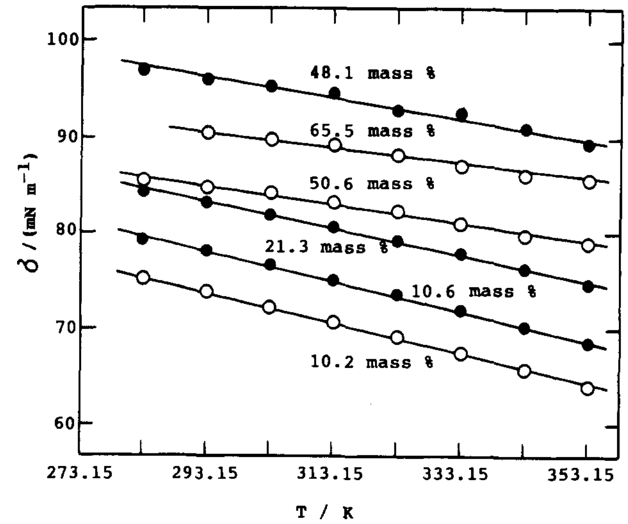

Figure 3. Surface tensions for two ternary systems: ○, H_2O + LiBr + LiI (LiBr:LiI = 4:1 mol); ●, H_2O + LiCl + $LiNO_3$ (LiCl:$LiNO_3$ = 2.8:1 mol); —, calculated values from eq 4.

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Received for review September 1, 1992. Accepted December 15, 1992. We gratefully acknowledge the partial support for this research by the Yazaki Memorial Foundation for Science and Technology.