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Solubilities, Vapor Pressures, Densities, and Viscosities of the LiBr + LiI + $HO(CH_2)_3OH$ + H_2O System

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The lithium bromide + lithium iodide + 1,3-propanediol + water (LiBr/LiI mole ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ mass ratio = 4) solution was selected as the potential working fluid for an air-cooled absorption chiller, and its basic four thermophysical properties were measured. The solubilities were measured in the range of absorbent (LiBr + LiI + $HO(CH_2)_3OH$) concentration from 70.0 mass % to 77.8 mass %. The vapor pressures were measured in the ranges of temperature and absorbent concentration from 339.75 K to 444.35 K and from 55.0 mass % to 80.0 mass %. Both the densities and viscosities were measured in the temperature and concentration ranges of 283.15 K to 343.15 K and 25.0 mass % to 75.0 mass %, respectively. The individual data set was correlated with a regression equation with high accuracy. The absorption chiller using the new solution as a working fluid is expected to have a wide operation range without the danger of crystallization, which is the critical problem in constructing an air-cooled absorption chiller.

Introduction

The absorption chiller is the cooling cycle of a working fluid that consists of an absorbent and refrigerant pair using combustion heat of fuel or waste heat as energy sources.[1] The absorption cooling cycle is one of the best alternatives to the vapor compression cooling cycle. The absorption cycle is similar to the vapor compression cycle, but the major advantage of the cycle is the use of heat as the energy source to obtain cooling power instead of expensive electric energy in the form of shaft work. For that reason, the vapor-compression cycle is described as a work-operated cycle and the absorption cycle as a heat-operated cycle. The increase of electricity cost and environmental problems has made this heat-operated cycle more attractive for both residential and industrial applications. Recently, much effort has been expended to develop a small-size absorption chiller which is closely related to the air-cooled cycle operation using a new working fluid. A conventional absorption chiller consisting of a generator, absorber, condenser, and evaporator requires an additional water cooling tower in order to remove heats and maintain desirable temperatures in absorber and condenser units. A small-scale absorption chiller has been constructed by replacing the cooling tower with the direct air-cooling system. The lithium bromide + water solution cannot be recommended as a potential working fluid due to an operational limit caused by salt crystallization. The high absorber temperature required by air-cooling needs a high absorbent concentration in order to generate a proper level of vapor pressure in the absorber. Thus, the development of a new working fluid with low crystallization temperature is most important in order to make a successful highly efficient air-cooled absorption chiller. In general, the crystallization temperature can be considerably lowered either by adding other salt(s) to the lithium bromide + water solution by using a specific mixing ratio among salts to give the highest solubility[2−5] or by adding anticrystallization organic chemicals with high boiling points and hygroscopic properties into the absorbent solution.[2,6−8] To check the validity of a potential working fluid to a specific chiller cycle, the physical and thermal properties should be accurately measured over the wide operation range expected. In our previous studies[9−13] three new working fluid mixtures of lithium bromide + ethanolamine + water, lithium bromide + 1,3-propanediol + water, and lithium bromide + diethanolamine + water were selected as possible candidates after examining their adaptability to an absorption chiller. Accordingly, their basic thermodynamic properties were measured at the fixed mass ratio (LiBr/$H_2N(CH_2)_2OH$, LiBr/$HO(CH_2)_3OH$, and LiBr/$(HOCH_2CH_2)_2NH$) of 3.5. The theoretical performance characteristics of these three solutions were also investigated in our previous work.[14] It should be noted that the addition of ethanolamine, 1,3-propanediol, and diethanolamine as anticrystallization additives into the lithium bromide + water solution results in increasing salt solubility and lowering of the crystallization temperature. On the other hand, the use of these organic-containing working fluids introduced undesired operational difficulties because of the high viscosity of the solution and the requirement of a rectifier to ensure the purity of regenerated refrigerant. For the lithium bromide + ethylene glycol + water solution suggested by Biermann and Relmann,[7] Inoue[2] mentioned the requirement of a rectifier in order to guarantee the purity of refrigerant because a noticeable amount of ethylene glycol can exist in the vaporized refrigerant in the generator. Therefore, viscosity and boiling point as well as solubility should be simultaneously considered when considering the use of additives into the working fluid. However, a solution containing 1,3-propanediol showed the smallest viscosity increment at the same condition. The boiling temperature of 1,3-propanediol is 17 K higher than

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that of ethylene glycol, and therefore, the lithium bromide + lithium iodide + 1,3-propanediol + water system is expected to contain less 1,3-propanediol in the vaporized mixture when compared with the amount of the lithium bromide + ethylene glycol + water solution. The relative amount of organics in the salt solutions can be reduced by using more than a single salt because the mixed-salt solution possesses a higher solubility power than the single-salt solution.

In this study, the lithium bromide + lithium iodide + 1,3-propanediol + water system (LiBr/LiI mole ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ mass ratio = 4) was selected as a possible new working fluid for an air-cooled absorption chiller. The mixing ratio of LiBr/LiI used was that suggested by Iyoki et al.[15] to give the highest solubility. The 1,3-propanediol was added to enhance salt solubilities. The four thermophysical properties of solubility, vapor pressure, density, and viscosity needed for the design of the air-cooled absorption chiller were measured for this newly proposed working fluid.

Experimental Section

The experimental apparatuses and procedures were the same as those described previously.[9−13] The solubility and viscosity measurements have an uncertainty accuracy of ±1% relative errors, while vapor pressure and the density have uncertainties of ±1.5% and ±0.1%, respectively.

Materials. The lithium bromide (mass fraction 0.99+), lithium iodide (mass fraction 0.99), and 1,3-propanediol (mass fraction 0.98) were supplied by Aldrich Chemical Co. and used without further purification. All solutions were prepared with deionized water. The mixing mole ratio of LiBr/LiI was fixed at 4 and the mixing mass ratio (LiBr + LiI)/$HO(CH_2)_3OH$ at 4, respectively.

Solubility and Vapor Pressure. The salt solubilities were measured by a visual polythermal method. The apparatus consisted of a glass vessel with an internal volume of 50 cm^3, a thermistor thermometer capable of reading to 0.01 K, a constant-temperature bath, a circulator for both cooling and heating, and two stirrers. Each sample solution of a desired absorbent concentration at a given mixing ratio was accurately prepared by weighing each component with a precise balance which has a resolution to 0.0001 g. The sample solution with an approximate volume of 30 cm^3 was placed in the vessel and stirred well. The solution was first incrementally heated above the crystallization temperature to dissolve all the crystals. The solution temperature was lowered and then raised at a very slow rate less than 0.1 K min^{-1} using the constant-temperature bath and the circulator. Through a series of dissolving and nucleation procedures, the temperature at which the last crystal disappeared was taken as the crystallization temperature for a given solution in this polythermal run. The vapor pressures were measured by a boiling point method. The apparatus primarily consisted of an equilibrium vessel with an internal volume of 500 cm^3, a constant-temperature bath, a condenser, a U-tube mercury manometer capable of reading to 0.05 mm, a calibrated K-type thermocouple with the accuracy of ±0.4% of reading, and two stirrers. A sample solution was initially prepared to the desired absorbent concentration and placed in the vessel with an approximate volume of 250 cm^3. The entire apparatus was then evacuated to a proper degree of pressure which is a little higher than the approximate vapor pressure of each sample. This sample solution was then heated and stirred well with a magnetic stirrer to prevent superheating. After thermal equilibrium was reached, the temperature of the sample solution and the corresponding pressure were measured.

Table 1. Solubilities of the LiBr + LiI + $HO(CH_2)_3OH$ + H_2O (LiBr/LiI Mole Ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ Mass Ratio = 4) System at Various Concentrations

w/mass frac	*T*/K	*w*/mass frac	*T*/K
0.700	276.15	0.750	302.35
0.708	280.45	0.754	302.45
0.719	287.85	0.757	303.65
0.727	292.45	0.760	304.35
0.733	295.35	0.763	308.35
0.737	297.55	0.767	315.75
0.743	299.95	0.773	324.35
0.747	301.65	0.778	332.15

Table 2. Vapor Pressures of LiBr + LiI + $HO(CH_2)_3OH$ + H_2O (LiBr/LiI Mole Ratio = 4, (LiBr + LiI)/$HO(CH_2)_3OH$ Mass Ratio = 4) System at Various Concentrations and Temperatures

T/K	*P*/kPa	*T*/K	*P*/kPa
w = 0.800		*w* = 0.750	
399.95	11.79	388.95	13.79
417.25	24.37	394.25	16.87
424.35	29.66	403.05	23.80
430.65	36.81	411.45	33.23
438.15	46.67	417.95	41.10
444.35	56.68		
w = 0.700		*w* = 0.650	
373.85	12.22	362.55	13.37
380.35	16.58	370.85	18.51
388.75	23.23	376.25	22.94
404.15	42.95	384.95	31.95
413.25	55.32	392.25	42.24
		399.45	54.10
w = 0.600		*w* = 0.550	
342.85	8.648	339.75	10.65
353.75	13.79	353.25	18.94
365.25	22.08	361.95	26.94
372.95	30.09	369.75	36.24
382.05	41.95	377.95	48.67
388.85	52.53		

Density and Viscosity. The temperature was controlled within ±0.05 K with a constant-temperature bath and was measured by a thermistor thermometer for both density and viscosity measurements. Densities were measured using a set of hydrometers capable of reading up to 1 $kg\cdot m^{-3}$. The sample solution of a desired concentration was first placed in the cylinder-type vessel clamped in the constant-temperature bath. A suitable hydrometer was then selected and immersed in the sample solution. After the desired solution temperature was attained, the corresponding density value was read from the meniscus of the solution with a hydrometer. Viscosities were measured by using five well-calibrated Ubbelohde-type viscometers which have different capillary diameters. A suitable viscometer was clamped in the constant-temperature bath, and the temperature was adjusted at a desired point. The efflux time of a sample solution through the capillary of the viscometer was accurately measured using a stopwatch. The reproducibility of the efflux time measurement was within ±0.1% of each value. The viscosity value was calculated by the following equation:

$$\eta = tK\rho \tag{1}$$

where η is the absolute viscosity, t the efflux time, K the given constant for the calculation of kinematic viscosity (tK), and ρ the density at the same condition.

Table 3. Densities and Viscosities of LiBr + LiI + $HO(CH_2)_3OH$ + H_2O (LiBr/LiI Mole Ratio = 4, (LiBr + LiI)/$HO(CH_2)_3OH$ Mass Ratio = 4) System at Various Concentrations and Temperatures

	ρ/kg·m^{-3}					
T/K	w = 0.750	w = 0.650	w = 0.550	w = 0.450	w =0.350	w =0.250
283.15		1586	1458	1350	1253	1169
293.15		1581	1453	1346	1250	1166
303.15	1735	1576	1448	1341	1246	1163
313.15	1730	1571	1443	1336	1242	1159
323.15	1724	1565	1437	1331	1237	1155
333.15	1717	1559	1432	1326	1233	1150
343.15	1711	1553	1427	1321	1228	1145

	η/m·Pa·s					
T/K	w = 0.750	w = 0.650	w = 0.550	w = 0.450	w =0.350	w =0.250
283.15		16.89	6.39	3.65	2.61	1.99
293.15		12.15	4.95	2.82	2.06	1.53
303.15	40.50	9.29	3.85	2.23	1.58	1.23
313.15	27.93	7.20	3.04	1.81	1.33	1.01
323.15	20.28	5.77	2.57	1.52	1.15	0.847
333.15	15.32	4.76	2.12	1.29	0.991	0.723
343.15	11.94	3.90	1.83	1.12	0.891	0.629

Table 4. Measured Properties and Regression Equations

property	regression equation
solubility	$T/\mathrm{K} = \sum_{i=0}^{2} A_i(100w)^i$ (2)
vapor pressure	$\log P/\mathrm{kPa} = \sum_{i=0}^{3}[A_i + 1000B_i/(T/\mathrm{K} - 43.15)](100w)^i$ (3)
density	$\rho/\mathrm{kg{\cdot}m}^3 = \sum_{i=0}^{3}[A_i + B_iT/\mathrm{K} + C_i(T/\mathrm{K})^2](100w)^i$ (4)
viscosity	$\log(\eta/\mathrm{mPa{\cdot}s}) = \sum_{i=0}^{4}[A_i + B_i/(T/\mathrm{K}) + C_i/(T/\mathrm{K})^2](100w)^i$ (5)

Table 5. Values of Coefficients for Each Least-Squares Representation (Each Parameter Set Is Only Valid for the Corresponding Experimental Range)

eq 2	A_0	A_1	A_2	AAD %[a]
		$w \le 0.760$		0.13
	-2.4955×10^3	7.1583×10^1	-4.5711×10^{-1}	
		$0.760 \le w$		
	-2.7605×10^3	6.4525×10^1	-3.1843×10^{-1}	

eq 3	i	A	B	AAD %
	0	9.537 943	−2.181 261	1.47
	1	$-1.303\,482 \times 10^{-1}$	$2.793\,675 \times 10^{-2}$	
	2	$1.919\,265 \times 10^{-3}$	$-3.596\,105 \times 10^{-4}$	
	3	$-7.926\,247 \times 10^{-6}$	$2.400\,981 \times 10^{-8}$	

eq 4	i	A	B	C	AAD %
	0	$1.428\,00 \times 10^3$	−2.447 15	$2.814\,95 \times 10^{-3}$	0.05
	1	$-5.186\,70 \times 10^1$	$3.698\,49 \times 10^{-1}$	$-5.485\,58 \times 10^{-4}$	
	2	1.875 45	$-1.176\,33 \times 10^{-2}$	$1.760\,69 \times 10^{-5}$	
	3	$-1.597\,73 \times 10^{-2}$	$1.038\,36 \times 10^{-4}$	$-1.568\,33 \times 10^{-7}$	

eq 5	i	A	B	C	AAD %
	0	$-5.324\,32 \times 10^1$	$2.880\,81 \times 10^4$	$-3.870\,10 \times 10^6$	0.75
	1	4.687 26	$-2.591\,05 \times 10^3$	$3.568\,23 \times 10^5$	
	2	$-1.425\,11 \times 10^{-1}$	$7.797\,14 \times 10^1$	$-1.056\,55 \times 10^4$	
	3	$1.804\,84 \times 10^{-3}$	$-9.702\,02 \times 10^{-1}$	$1.281\,87 \times 10^2$	
	4	$-8.118\,53 \times 10^{-6}$	$4.264\,44 \times 10^{-3}$	$-5.407\,07 \times 10^{-1}$	

[a] AAD % = $(100/\mathrm{NP})\sum_{i=1}^{\mathrm{NP}}|(\mathrm{value}_i^{\mathrm{exp}} - \mathrm{value}_i^{\mathrm{calc}})/\mathrm{value}_i^{\mathrm{exp}}|$; NP = number of data points.

Results and Discussion

All the measured properties for the lithium bromide + lithium iodide + 1,3-propanediol + water (LiBr/LiI mole ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ mass ratio = 4) system were listed in Tables 1−3. All the data were correlated by the fitting equations suggested in Table 4. For these equations T is the absolute temperature in K, X the absorbent (LiBr + LiI + $HO(CH_2)_3OH$) concentration in mass %, and A_i, B_i, and C_i the regression parameters. The resulting parameters determined by a least-squares method are listed in Table 5. The deviations of calculated data sets from the measured sets were expressed by the average absolute deviation (AAD). It should be noted that the fitting equations and corresponding parameters are valid only within the specified temperature and concentration ranges.

The saturated solubilities at crystallization temperatures were measured in the range of absorbent concentration from 70.0 mass % to 77.8 mass %. Due to the change of crystal structure of the solid equilibrated with the saturated solution, the invarient point at which two different types of hydrates coexist appears in the solubility curve at the specified temperature and concentration. For the lithium bromide + lithium iodide + 1,3-propanediol +

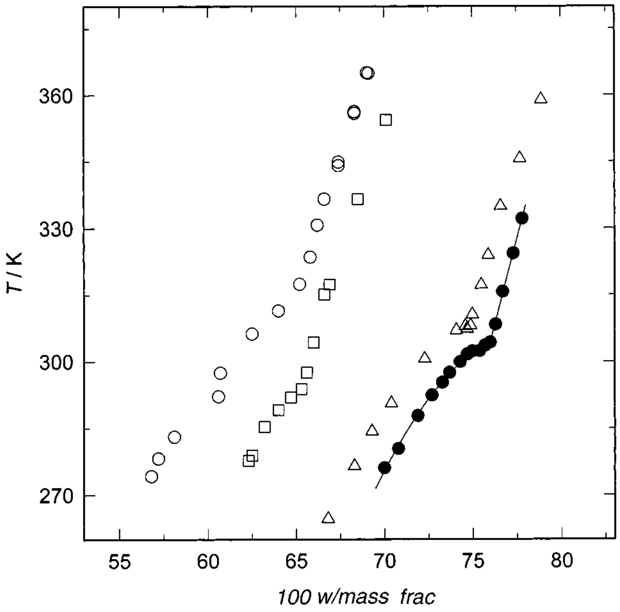

Figure 1. Solubilities of the LiBr + LiI + $HO(CH_2)_3OH$ + H_2O (LiBr/LiI mole ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ mass ratio = 4) system: ○, LiBr + LiI + $HO(CH_2)_3OH$ + H_2O system; □, H_2O + LiBr system;[16] △, LiBr + LiI + H_2O (LiBr/LiI mole ratio = 4) system;[4] ●, LiBr + $HO(CH_2)_3OH$ + H_2O (LiBr/$HO(CH_2)_3$ OH mass ratio = 3.5) system;[12] —, calculated.

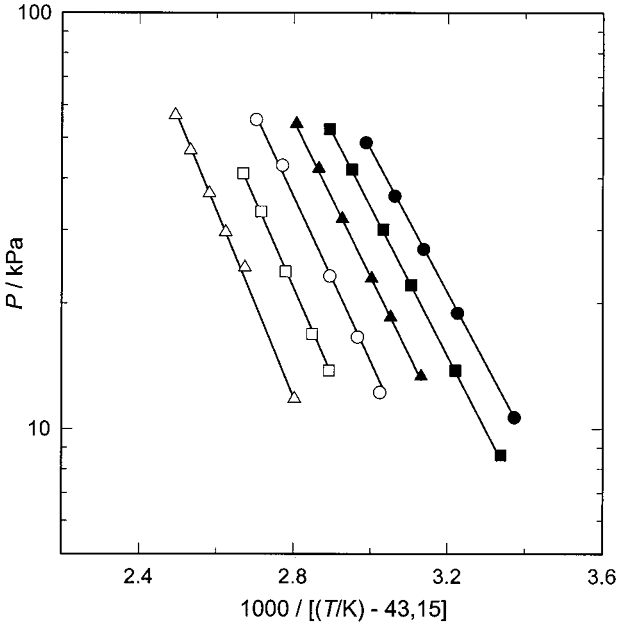

Figure 2. Vapor pressures of the LiBr + LiI + $HO(CH_2)_3OH$ + H_2O (LiBr/LiI mole ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ mass ratio = 4) system: △, 55.0 mass %; □, 60.0 mass %; ○, 65.0 mass %; ▲, 70.0 mass %; ■, 75.0 mass %; ●, 80 mass %; —, calculated.

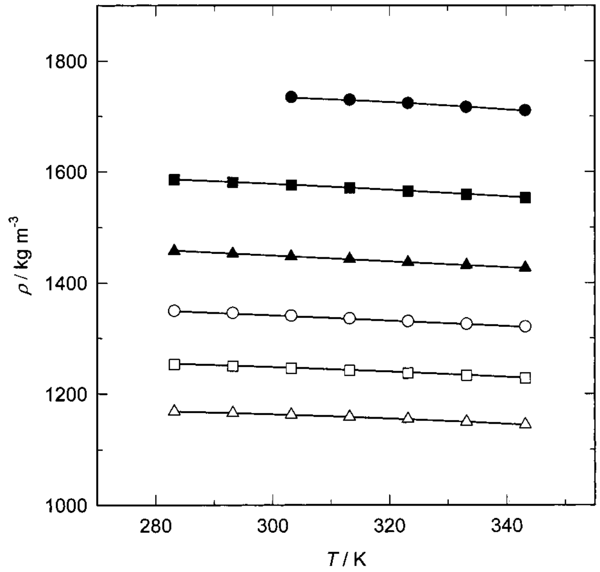

Figure 3. Densities of the LiBr + LiI + $HO(CH_2)_3OH$ + H_2O (LiBr/LiI mole ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ mass ratio = 4) system: △, 25.0 mass %; □, 35.0%; ○, 45.0 mass %; ▲, 55.0 mass %; ■, 65.0 mass %; ●, 75.0 mass %; —, calculated.

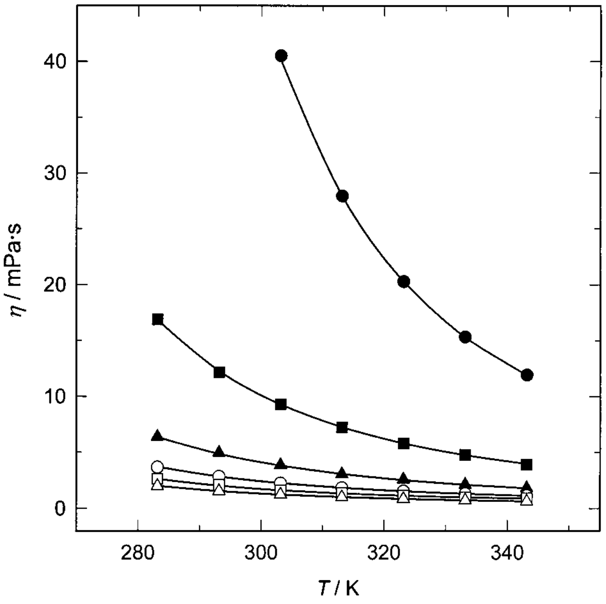

Figure 4. Viscosities of the LiBr + LiI + $HO(CH_2)_3OH$ + H_2O (LiBr/LiI mole ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ mass ratio = 4) system: △, 25.0 mass %; □, 35.0%; ○, 45.0 mass %; ▲, 55.0 mass %; ■, 65.0 mass %; ●, 75.0 mass %; —, calculated.

water solution the invariant point was found at 304.35 K and 76.0 mass % of absorbent. Two different solubility data sets divided at this point were fitted with eq 2 in Table 4. The calculated results are plotted in Figure 1 together with experimental values. The figure also includes the solubilities of the lithium bromide + water, lithium bromide + lithium iodide + water (LiBr/LiI mole ratio = 4), and lithium bromide + 1,3-propanediol + water (LiBr/HO-$(CH_2)_3OH$ mass ratio =3.5) systems. As shown in this figure, the lithium bromide + lithium iodide + 1,3-propanediol + water system has the highest solubility among four systems. This fact reveals that the lithium bromide + lithium iodide + 1,3-propanediol + water system can be a potential candidate for a new working fluid with high solubility. Vapor pressures were measured in the temperature and concentration ranges of 339.75 K to 444.35 K and 55.0 mass % to 80.0 mass %, respectively. The measured results are plotted in Figure 2 along with the results calculated by eq 3. As expected from this figure, a common linear relationship between log P/kPa and 1000/(T/K − 43.15) at a given concentration is found over the entire temperature range.

Densities and viscosities were measured at seven different temperatures between 283.15 K and 343.15 K and for the absorbent concentration range of 25.0 mass % to 75.0 mass %. However, the densities and viscosities of the 75 mass % solution at the temperatures 283.15 K and 293.15 K were not measured due to the crystallization of the solution. The experimental results are plotted in

Figures 3 and 4 along with the calculated results by polynomial eqs 4 and 5. The solution density increases with increasing concentration of absorbent and is almost linear with temperature. The solution viscosity increases to a large extent with an increase in the absorbent concentration and slowly decreases with increasing temperature.

Conclusions

A new working fluid for the air-cooled absorption chiller has been developed by using the optimally mixed salt solution and by adding an anticrystallization agent. As a potential solution mixture, the lithium bromide + lithium iodide + 1,3-propanediol + water (LiBr/LiI mole ratio = 4 and (LiBr + LiI)/$HO(CH_2)_3OH$ mass ratio = 4) system in which the 1,3-propanediol was used as an additive has been first attempted. The solubilities, vapor pressures, densities, and viscosities were measured over a wide concentration and temperature range. The data for each property were correlated with a proper regression equation, and the calculated result shows good agreement with the measured result. The new working fluid is expected to be used for the air-cooled absorption chiller as a working fluid. The measured data sets and the correlation results can be used to give the required thermodynamic data for the detailed system design. However, to investigate the heat and mass transfer characteristics of the working fluid, more properties such as heat capacity, enthalpy of mixing, thermal conductivity, mass diffusivity, and surface tension are required.

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