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Articles

Vapor−Liquid Equilibria for the HI + H_2O System and the HI + H_2O + I_2 System

Masatoshi Hodotsuka,[†] Xiaoyong Yang,[‡] Hiroyuki Okuda, and Kaoru Onuki*

Nuclear Science and Engineering Directorate, Japan Atomic Energy Agency, 4002, Narita-cho, Oarai-machi, Higashiibaraki-gun, Ibaraki, 311-1393, Japan

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Two equilibrium stills have been designed for measuring vapor−liquid equilibria of hydriodic acid and polyhydriodic acid. The acids are so corrosive that the devices must be made of corrosion resistant materials. For the HI + H_2O binary system, a still has been made of tantalum and Hastelloy and operated under pressure. For the HI + H_2O + I_2 ternary system, another still has been made of glass and operated at atmospheric pressure. With these devices, VLE data of hydriodic acid have been acquired. The experimental conditions for the binary system were as follows: the pressure range was from (0.11 to 0.58) MPa; the HI mole fraction range in the liquid was from 0 to 0.157; and the temperature range was from (375 to 456) K. For the ternary system, the pressure was atmospheric pressure; the HI concentration in the liquid was close to the azeotrope; the HI to I_2 mole ratio in the liquid range was from 1/0 to 1/4; and the temperature range was from (396 to 408) K.

Introduction

The sulfur−iodine cycle (SI) was invented by General Atomics in the 1970s and has been researched as the most promising thermochemical water-splitting cycle for hydrogen production.[1,2] In the SI cycle, there is a distillation column which distills polyhydriodic acid (HI (1) + H_2O (2) + I_2 (3)). It is essential to have vapor−liquid equilibrium data of hydriodic acid (HI (1) + H_2O (2)) and polyhydriodic acid to design a distillation column.

However, only few VLE data of hydriodic acid have been published so far. The first report concerning the hydriodic acid was published by Hasse et al.[3] They reported the total vapor pressure and the vapor composition at 298 K in a range of x_1 from 0 to 0.346. In 1964, Lutugina et al.[4] reported isobaric equilibrium of hydriodic acid at atmospheric pressure for x_1 from 0 to 0.155. In the 70s and 80s, total vapor pressure was examined by Neuman,[5] Engels and Knoche,[6] and Berndhäuser and Knoche[7] on polyhydriodic acid with x_1 up to 0.193 in a wide temperature range of (350 to 553) K, although the vapor composition was not measured. They calculated bubbling point curves of the azeotropes whose HI mole fraction was ranging from about 0.130 to 0.155.

Recently Doizi et al.[8] reported data using a static cell which featured optical windows for the measurement of vapor composition. They published experimental VLE data of hydriodic acid and polyhydriodic acid.

Table 1. Component, Supplier, and Component Purity (wt %)

component	supplier	purity
hydriodic acid	Kanto Chemical Co., INC. Tokyo, Japan	55.0 to 58.0
hydriodic acid	Merck Ltd., Japan	67
Iodine	Kanto Chemical Co., INC. Tokyo, Japan	>99.8

To obtain VLE data of hydriodic acid and polyhydriodic acid, two stills were designed and used. With these apparatuses, both the liquid composition and the vapor composition could be measured directly.

The VLE data of hydriodic acid was measured with a metal-made Gillespie-type still, while that of polyhydriodic acid was made with a glass-made Othmer-type still.

Experimental Section

Materials. Purities and suppliers of used experimental materials are presented in Table 1. Deionized water (Millipore Milli-RX12) was used for preparing the test solutions. 0.1 $mol \cdot dm^{-3}$ sodium hydroxide solution (Kanto Chemical Co., INC., Japan), 0.1 $mol \cdot dm^{-3}$ KIO_3 solution (Kanto Chemical Co., INC.), and 0.1 $mol \cdot dm^{-3}$ $Na_2S_2O_3$ solution (Kanto Chemical Co., INC.) were used for titration. Helium gas (99.995 %, Tomoe Shokai Co., Ltd., Japan) was used as a cover gas.

Equipment and Procedure. Two ebulliometers were designed for measuring vapor−liquid equilibrium. Device 1 was for the HI + H_2O binary system at elevated pressure, and device 2 was for the HI + H_2O + I_2 ternary system at atmospheric pressure.

High Pressure Apparatus. The schematic of the VLE still (device 1) is shown in Figure 1. This apparatus is based on a Gillespie still, in which both the vapor and the liquid circulate. Hiaki et al. modified a still by using stainless steel to measure VLE data of many substances and mixtures at high pressure.[9]

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* Corresponding author. E-mail: onuki.kaoru@jaea.go.jp.

[†] Present address: TOSHIBA corporation, 4-1, Ukishima-cyo, Kawasaki-ku, Kawasaki-shi, Kanagawa, 211-0862, Japan. Telephone: +81-44-288-8161. Fax: +81-44-270-16. masatoshi.hodotsuka@toshiba.co.jp.

[‡] Present address: Institute of Nuclear and New Energy Technology Tsinghua Univ., Nengke Lou, INET, Beijing, China, 100084. Telephone: +86-10-6277-1746. Fax: +86-10-6277-1150. xy-yang@mail.tsinghua.edu.cn.

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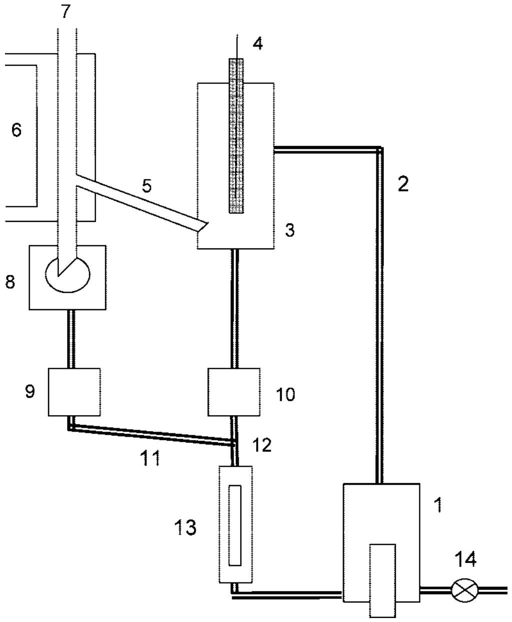

Figure 1. Schematic of the VLE still (device 1). 1, Evaporator; 2, Cotterell Pump; 3, Equilibrium Chamber; 4, Temperature Monitor; 5, Vapor Stream with Heater; 6, Condenser; 7, To Pressure Control System; 8, Drop Counter; 9, Sampling Pot (vapor); 10, Sampling Pot (liquid); 11, Condensate Stream with Cooling jacket; 12, Merging Point; 13, Level Meter; 14, Drain Valve.

However, the reagents employed in this report were so corrosive that the material of the still was changed from stainless steel to MA22 (equivalent of Hastelloy C-22, Mitsubishi Materials), MA276 (equivalent of Hastelloy C-276, Mitsubishi Materials), and tantalum.[10] A heat exchanger (cooling jacket) was installed just before the merging section, where the liquid flow from the equilibrium chamber and the condensate from the condenser meet, to prevent the condensate from evaporating as a result of heat conduction from the evaporator. The difference of boiling points between hydrogen iodide and the azeotrope of the hydriodic acid + water is estimated to be about 435 K at atmospheric pressure.

The apparatus can be operated at a constant pressure up to 0.6 MPa with the aid of helium cover gas and a pressure regulating system (Druck DPI520 and RUI101) which enables control of the system pressure with an uncertainty of ± 0.025 % full scale. The uncertainty should be 15 Pa. The equilibrium temperature is measured by a Pt100 resistance thermometer, whose signal is monitored by an ASL F200 system with an uncertainty of ± 0.01 K.

First, the still was evacuated. Then about 100 cm^3 of hydriodic acid was placed in the evaporator and heated. The vapor and liquid went through the Cottrell pump to the equilibrium chamber. A sheath for the Pt resistance thermometer was inserted to measure the temperature in the chamber. Just after passing through the equilibrium chamber, the vapor and the liquid were separated. The liquid returned to the evaporator by gravity. The vapor went through the pipe which was heated to avoid condensation of the vapor and condensed in the condenser which was chilled by ethanol down to about 243 K. The condensate was observed through the drop counter window and built up in the sampling pot to about 10 cm^3. In the pot, the solution was always stirred during operation. Overflow solution merged with the liquid at the merging point and went back together to the evaporator.

After the pressure and the temperature in the equilibrium chamber were stable for about 1 h and the number of drops per minute kept in the range of 30 to 90, the system was considered to be in equilibrium, and the sampling from both the liquid phase and the vapor phase took place. The drop rate was largely dependent on the surface tension of the condensate. When the condensate was almost water, its surface tension was quite large, and the volume of each drop was larger. Finally the drop rate ends up decreasing.

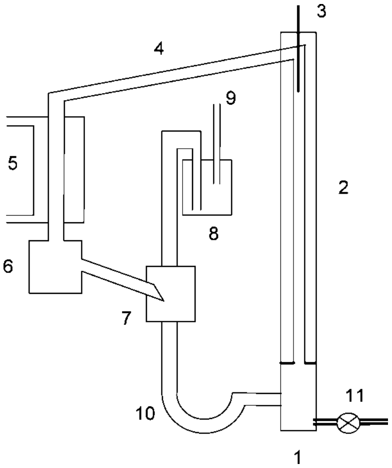

Figure 2. Schematic of the VLE still (device 2). 1, Evaporator; 2, Vapor Stream (vacuum insulation); 3, Temperature Monitor; 4, Vapor Stream with Heater; 5, Condenser; 6, Sampling Pot (vapor); 7, Drop Counter; 8, Water Seal Pot; 9, Vent; 10, Condensate Stream with Cooling Jacket; 11, Drain Valve.

Atmospheric Pressure Apparatus. The schematic of the VLE still (device 2) is shown in Figure 2. This apparatus was an Othmer still, in which only the vapor circulates. It was made of glass and operated at atmospheric pressure. The temperature in the upper part of the column was measured by a thermocouple, with an uncertainty of ± 1.5 K (K-type, class 1, Sukegawa-Denki Co., Ltd., Japan). The pressure was measured by a Fortin barometer, with an uncertainty of ± 1 mmHg (G1-16, Isuzu Seisakusyo Co., Ltd., Japan).

About 300 cm^3 of polyhydriodic acid was added to the evaporator and heated. The vapor started to go up through the column to the top, which was covered with insulation. A sheath for a thermocouple was inserted to measure the temperature at the top. Just after passing through the top, the vapor condensed in the condenser which was chilled by ethanol to about 238 K, because the boiling point of hydriodic acid at the atmospheric pressure is about 238 K.

The condensate was observed through the drop counter window and built up in the sampling pot about 10 cm^3. In the pot, the solution was always stirred during operation. The overflow solution went back to the evaporator. A water seal pot was installed to absorb the HI vapor, and the height of the water was set about 10 cm.

After the temperature at the top of the column and the drop rate of the condensate were stable for more than 3 h, the system was considered to be in equilibrium. Then the sampling from both the condensate and the boiling solution took place.

Titration. Samples of both vapor and liquid phases were analyzed using an automatic potentiometric titrator (COM-2500, Hiranuma Sangyo Co., Ltd., Japan). The uncertainty in the analysis was ± 0.1 %.

Sodium hydroxide solution (0.1 mol·dm^{-3}), KIO_3 solution (0.1 mol·dm^{-3}), and $Na_2S_2O_3$ solution (0.1 mol·dm^{-3}) were used for measuring the concentration of H^+, I^-, and I_2, respectively.

Table 2. Experimental Vapor−Liquid Equilibrium Data for HI (1) + H_2O (2)[a,b]

P/MPa	T/K	x_1	x_2	y_1	y_2
0.1115	375.05	0	1	0	1
0.1115	387.01	0.090	0.910	0.003	0.997
0.1114	400.85	0.134	0.866	0.086	0.914
0.1113	401.20	0.147	0.853	0.127	0.873
0.1500	383.73	0	1	0	1
0.1500	386.72	0.036	0.964	0.000	1.000
0.1500	390.53	0.063	0.937	0.000	1.000
0.1500	397.00	0.094	0.906	0.001	0.999
0.1500	407.77	0.134	0.865	0.039	0.961
0.1500	409.96	0.157	0.842	0.211	0.789
0.1500	410.22	0.150	0.850	0.129	0.871
0.1500	410.26	0.156	0.843	0.168	0.832
0.1500	410.27	0.154	0.845	0.170	0.830
0.1500	410.29	0.152	0.847	0.156	0.844
0.1500	410.30	0.154	0.845	0.151	0.849
0.2027	392.91	0	1	0	1
0.2025	419.46	0.149	0.850	0.159	0.841
0.3040	406.24	0	1	0	1
0.3040	431.07	0.119	0.881	0.050	0.950
0.3039	432.52	0.146	0.853	0.174	0.826
0.3041	432.75	0.138	0.861	0.157	0.843
0.3039	432.83	0.136	0.863	0.123	0.877
0.3040	432.85	0.144	0.856	0.147	0.853
0.5799	429.62	0	1	0	1
0.5800	429.78	0	1	0	1
0.5800	433.98	0.039	0.960	0.000	1.000
0.5799	444.91	0.086	0.913	0.003	0.997
0.5801	452.41	0.114	0.886	0.023	0.977
0.5800	453.31	0.153	0.846	0.251	0.749
0.5800	454.62	0.150	0.850	0.204	0.796
0.5799	455.04	0.129	0.870	0.098	0.902
0.5800	455.79	0.130	0.870	0.074	0.926
0.5799	455.79	0.139	0.861	0.159	0.841
0.5801	455.87	0.134	0.865	0.079	0.921
0.5801	456.24	0.146	0.853	0.213	0.787
0.5799	456.28	0.139	0.861	0.138	0.862
0.5795	456.39	0.128	0.872	0.121	0.879

[a] In all the binary experiments, I_2 mole fractions in the liquid and the vapor phase were set at 0.001 and 0, respectively. [b] Pressure has an uncertainty of ± 0.000015 MPa. Temperature has an uncertainty of ± 0.01 K. The mole fraction of x_1 and x_2 has an uncertainty of ± 0.01.

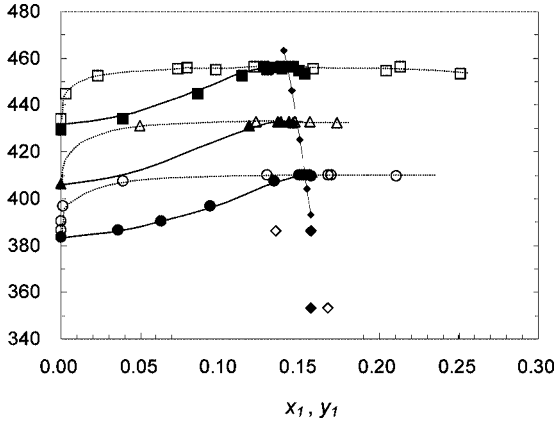

Figure 3. Isobaric VLE data of the binary system HI (1) + H_2O (2). This work: ■, Bubbling Point at 0.58 MPa; □, Dew Point at 0.58 MPa; ▲, Bubbling Point at 0.30 MPa; Δ, Dew Point at 0.30 MPa; ●, Bubbling Point at 0.15 MPa; ○, Dew Point at 0.15 MPa. Broken line with x is the bubbling point curve of the azeotrope calculated by Berndhäuser.[5] ◆, ◇ are experimental data by Doizi:[8] ◆, Bubbling Point; ◇, Dew Point. The pressure of the VLE data at 386 K was 0.016 MPa, and that at 353 K was 0.059 MPa. Solid lines and dotted lines were drawn by eye. The former shows bubbling point curves, and the latter shows dew point curves.

Results and Discussion

A binary system must be defined in this report because the I^- ion in the HI solution is readily oxidized to I_2 by oxygen, light, and heat. During the experiment, some I^- ion was oxidized to I_2. The I_2 is known to dissolve in the solution that contains the I^- ion to form I_3^- or I_5^-[11] and make the solution colored. Therefore, hydriodic acid containing a small amount of iodine which originated from I^- was defined as the binary system.

High Pressure VLE. The VLE data in the binary system are shown in Table 2. Figure 3 shows the T, x, y plots of hydriodic acid at (0.58, 0.30, and 0.15) MPa. The HI mole fraction in the liquid samples and vapor samples shows the B.P. (Bubbling Point) and D.P. (Dew Point), respectively. B.P. marks on the y-axis show the boiling points of pure water at each pressure. At each pressure, an azeotropic point can be found in the range of $x_1 = 0.13$ and 0.15. The HI mole fraction of the azeotropes has a good accordance with the calculation result of the literature.[7] It has been confirmed that the azeotropic concentration shifts to the left, or more dilute side, as the pressure elevates. However, the results of Doizi et al.[8] show the opposite behavior. Even with the same composition of hydriodic acid ($x_1 = 0.157$), the solution was under azeotrope at 386 K (0.016 MPa) and over azeotrope at 353 K (0.059 MPa). Elevating the pressure, the azeotropic concentration shifts to the right, or more concentrated side. It might suggest that the relation between the bubbling point of the azeotrope and the pressure depend on the pressure range. The concentration of I_2 in the liquid samples was so dilute and the amount of each sample was so small that I_2 could not be measured by titration. Therefore, the I_2 concentration in the drained HI solution was measured by titration. The mole fraction of I_2 was 0.001. The drained HI solution was considered to be mainly the liquid phase because the amount of the condensate (the vapor phase) was small compared with that of the liquid phase and most of the condensate was considered to remain in the sampling pot without mixing with the liquid phase. In calculating the composition of the liquid phase, we set the I_2 mole fraction as 0.001 ± 0.001.

The uncertainty in the analysis comes from weighing and titration. As weighing has a smaller error than titration, the main cause of the uncertainty is thought to be from titration. The uncertainty of titration is estimated to be less than 0.1 %. The uncertainty of HI and H_2O mole fraction of binary experiments is thought to be more than 0.1 % because the I_2 mass fraction in the liquid can contain 0.001. Since the HI mole fraction of an azeotrope is about 0.15, the uncertainty would be 0.001/0.15 = 0.6 %. Compared with the uncertainty of titration (0.1 %), the uncertainty from I_2 is larger.

On the other hand, all the samples of vapor phases were almost transparent. It means the I_2 concentration in the vapor phase is far less than that in the liquid phase. Therefore, we neglected the I_2 concentration in the vapor phases.

Atmospheric Pressure VLE. The VLE data in the ternary system at the atmospheric pressure are shown in Table 3. Figure 4 shows the comparison with the results of Lutugina.[4] Figure 5 shows the composition of the vapor and the liquid phases in the ternary graph with the results of Doizi.[8]

At first, the device for the atmospheric pressure was tested by hydriodic acid, and the result had a good accordance with that of Lutugina[4] in Figure 4. In Figure 5, the two marks, ●; and ○, indicate the compositions of the liquid and the vapor in equilibrium, respectively. One data set was obtained in one experiment. First, the HI + H_2O binary solution was placed in the apparatus. After the binary test, some iodine was added to the solution. Then, another experiment in a different condition was carried out. In this way, the I_2 mole fraction in the HI (1) + H_2O (2) + I_2 (3) solution was increased up to $x_3 = 0.423$.

Table 3. Experimental Vapor−Liquid Equilibrium Data for HI (1) + H_2O (2) + I_2 (3) at Atmospheric Pressure[a]

P/MPa	T/K	x_1	x_2	x_3	y_1	y_2	y_3
0.1000	401.4	0.153	0.847	0.000	0.1371	0.8629	0.0000
0.1011	401.2	0.158	0.841	0.001	0.2015	0.7982	0.0003
0.1017	402.3	0.153	0.829	0.018	0.1342	0.8631	0.0026
0.1008	396.0	0.141	0.839	0.020	0.0072	0.9902	0.0025
0.1012	402.5	0.154	0.808	0.038	0.1321	0.8634	0.0045
0.1020	402.5	0.150	0.802	0.048	0.1450	0.8484	0.0065
0.1009	402.0	0.153	0.789	0.057	0.1774	0.8152	0.0074
0.1012	399.0	0.141	0.799	0.061	0.0246	0.9709	0.0046
0.1020	403.2	0.146	0.763	0.091	0.1273	0.8641	0.0086
0.1018	400.7	0.140	0.756	0.104	0.0400	0.9525	0.0075
0.1024	403.9	0.144	0.739	0.117	0.1521	0.8372	0.0107
0.1017	402.9	0.154	0.718	0.128	0.1298	0.8598	0.0103
0.1021	400.7	0.138	0.715	0.147	0.0325	0.9567	0.0109
0.1016	401.6	0.137	0.713	0.151	0.0464	0.9424	0.0112
0.1017	403.7	0.136	0.698	0.166	0.1057	0.8806	0.0137
0.1006	401.8	0.134	0.690	0.175	0.0478	0.9376	0.0146
0.1023	403.9	0.134	0.674	0.192	0.1012	0.8810	0.0178
0.1020	404.6	0.135	0.671	0.194	0.1253	0.8585	0.0163
0.1017	402.7	0.135	0.658	0.207	0.0493	0.9325	0.0182
0.1021	404.7	0.129	0.657	0.214	0.0947	0.8841	0.0212
0.1000	404.7	0.133	0.647	0.220	0.1829	0.7859	0.0312
0.1019	403.8	0.135	0.642	0.223	0.1097	0.8206	0.0698
0.1008	404.6	0.127	0.616	0.257	0.0934	0.8807	0.0259
0.1020	405.4	0.132	0.601	0.267	0.0643	0.9047	0.0310
0.1024	404.9	0.125	0.611	0.264	0.0800	0.8929	0.0271
0.1008	405.9	0.126	0.577	0.297	0.1118	0.8469	0.0413
0.1019	407.7	0.120	0.562	0.318	0.0869	0.8800	0.0331
0.1014	406.6	0.133	0.490	0.377	0.1005	0.8620	0.0375
0.1019	407.7	0.108	0.468	0.423	0.0775	0.8482	0.0743

[a] Pressure has an uncertainty of ± 0.0001 MPa. Temperature has an uncertainty of ± 1.5 K. The mole fraction of x_1 and x_2 has an uncertainty of ± 0.002.

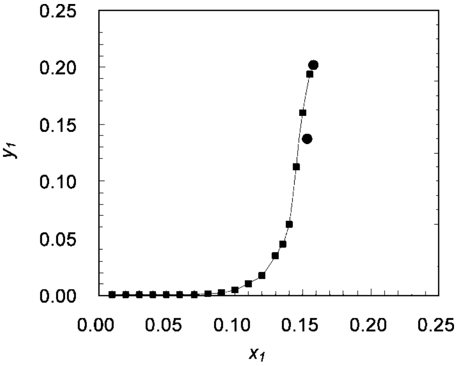

Figure 4. *xy* curve of the binary system HI (1) + H_2O (2) at atmospheric pressure. This work: ●, Lutugina;[4] solid curve with ■.

The solution (x_1, y_1, z_1 = 0.108, 0.468, 0.423) was close to the composition of Doizi et al.'s experiments.[8] In their results, HI concentration in the vapor was demonstrated to be very sensitive when the liquid was close to the pseudoazeotrope. Comparing with their data, the result of this work could be judged as excellent. As the I_2 concentration increased, the HI concentration in the liquid expressed in the two-component system (calculated by [HI]/([HI] + [H_2O])) gradually increased, but HI concentration in the vapor decreased. It means that increasing the I_2 concentration in the liquid makes the pseudoazeotrope more concentrated in HI.

The uncertainty in the analysis comes from weighing and titration. As weighing has a smaller error than titration, the main cause of the uncertainty is thought to be from titration. The uncertainty of titration is estimated less than 0.1 %.

Even though no iodine was added in the binary experiments, the solutions finally included some iodine after the experiment. The I_2 mole fraction in the liquid can be estimated as 0.001 ± 0.001 from the result of "high pressure VLE". The same as "high pressure VLE", the colors of the condensates (vapor phase) were almost transparent, so I_2 concentration in the condensates could be neglected.

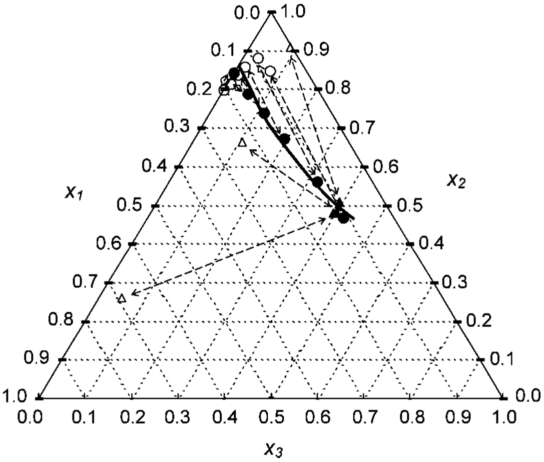

Figure 5. VLE data of the ternary system HI (1) + H_2O (2) + I_2 (3) at atmospheric pressure. This work: ●, liquid; ○, vapor. ▲ and □ are experimental data at close to the atmospheric pressure by Doizi:[8] ▲, Liquid; □, vapor. Solid line: imaginary line of the pseudoazeotrope curve.

Conclusions

To design a distillation column for polyhydriodic acid, two equilibrium stills for measuring VLE data were made and used. VLE data of HI + H_2O and HI + H_2O + I_2 were obtained.

We have realized that as the pressure increases the HI concentration of the azeotrope decreases in the HI + H_2O system and that in the HI + H_2O + I_2 system as the I_2 concentration increases the HI concentration of the azeotrope (expressed in the two-component system) increases. These VLE data must be the basic data for designing the distillation column in the SI cycle.

For acquiring more precise VLE data, the experiments must be carried out in the condition that the decomposition rate of HI should be negligibly small. Especially in the binary system, HI is readily oxidized.

In this paper, there are no VLE data of HI solution which is far more concentrated than the azeotrope. As the HI concentration in the liquid gets more concentrated than the azeotrope, the HI concentration in the vapor is estimated to increase suddenly.[5,8] In the condenser, the HI-rich vapor is condensed at very low temperature. Since the condensate of such vapor has a very low bubbling point, it is difficult to return the condensate to the hot evaporator without bubbling. It is required to install a new device which can properly recycle the condensate. With this device, a wide range of VLE data will be acquired.

There is one more area in the ternary graph where the experiment is hard to perform. It is where the HI concentration is more diluted than the azeotrope, and I_2 dissolved as a complex ion in the liquid. When the solution is supplied, the HI concentration in the condensate gets low enough to precipitate I_2 in the condensate, which makes it impossible to take homogeneous samples. To measure VLE data of the solution, a new apparatus which does not have a cold area is required.

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