

Development of a New Advanced Process for Manufacturing Polyacetal Resins. Part II. Vapor–Liquid Equilibrium of Formaldehyde–Water System

J. MASAMOTO,* K. MATSUZAKI, and H. MORISHITA

Asahi Chemical Industry Co., Ltd., 2767-11, Azanishima, Kojimashionasu, Kurashiki, 711 Japan

SYNOPSIS

To develop a new technology for the purification of formaldehyde from formalin by a distillation method, the vapor–liquid equilibrium of formaldehyde–water binary systems was investigated. Based on the studies of Thompson et al. and Iliceto et al., the vapor–liquid equilibrium of formaldehyde and water are calculated and these calculations are in good agreement with the reported data. It was found that the vapor pressure of formaldehyde is determined by the free formaldehyde concentration (mol/L), whereas the vapor pressure of water is determined by the molar ratio of free water. The vapor–liquid equilibrium of formaldehyde–water in the presence of a diluent was also investigated. It was shown by calculation that in some cases, when the concentration of the diluent is high, the volatility of formaldehyde is always higher than that of water and these calculations are in good agreement with experimental data. Based on this principle, the purification of formaldehyde by an extraction distillation method was established. © 1993 John Wiley & Sons, Inc.

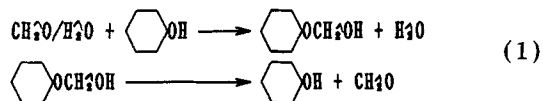
1. INTRODUCTION

Acetal homopolymer can be obtained by the homopolymerization of formaldehyde or trioxane. However, only from formaldehyde polymerization is the acetal homopolymer industrially produced. At present, DuPont and Asahi Chemical are the only producers of the acetal homopolymer.

Although details of the process for acetal homopolymer have never been made public, the general processing step can be outlined as monomer purification, polymerization, end-capping, and finishing. In this article, we will mention mainly the new technologies developed by Asahi Chemical.

Features of the Asahi Chemical's acetal homopolymer process are as follows: First, it uses highly purified formaldehyde gas as the monomer. Second, end-capping during polymerization uses highly purified formaldehyde as the monomer and acetic anhydride as a chain transfer or end-capping agent.

The formaldehyde purification technology will now be discussed in more detail: Anhydrous formaldehyde used in the manufacture of the acetal homopolymer must be extremely pure to achieve a high molecular weight. Even very low concentrations of residual formic acid or hydroxy compounds, such as water and methanol, in the monomer can result in an unacceptably low molecular weight for the finished product; great care is taken to eliminate these species during monomer production. Purification schemes listed in the patent literature^{1–10} include scrubbing with an inert liquid, partial polymerization to remove chain transfer impurities, conversion of formaldehyde to a hemiformal and subsequent decomposition to purified formaldehyde, and treatment with P₂O₅. The hemiformal method is the most prevalent [eq. (1)]. Formaldehyde is treated with cyclohexanol to form cyclohexyl hemiformal^{9,10}:

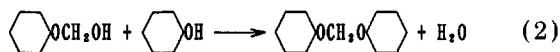


The cyclohexanol containing the hemiformal, along with water and methanol, is sent to a distillation column in which the water–cyclohexanol

* To whom correspondence should be addressed.

azeotrope and methanol are removed overhead. Bottoms from this column are then sent to a pyrolysis column, where they are heated in order to decompose the hemiformal and formaldehyde gas goes overhead. Removal of trace cyclohexanol and any remaining water is accomplished by condensation after chilling the formaldehyde gas.

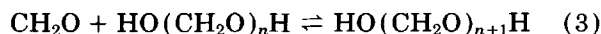
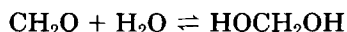
This hemiformal process is rather complicated and, moreover, dicyclohexyl formal is generated as a byproduct [eq. (2)]:



Ube industries proposed poly(ethylene glycol) in place of cyclohexanol to obtain purified formaldehyde.¹¹

Masamoto et al. of Asahi Chemical recently invented the extractive distillation process as a new simple purification technology.¹² We will now introduce this new distillation purification technology.

Formaldehyde itself is a material whose boiling point is -19°C and is a gas at room temperature. However, in aqueous solution, it makes homologs of HOCH_2OH , $\text{HO}(\text{CH}_2\text{O})_2\text{H}$ — — — $\text{HO}(\text{CH}_2\text{O})_n\text{H}$, in addition to a small amount of free formaldehyde. These are the results of the reaction equilibrium [eq. (3)]:



2. WHY CAN'T FORMALDEHYDE BE PURIFIED BY DISTILLATION?

It is well known that the vapor-liquid equilibrium of aqueous formaldehyde solution has an azeotropic point.¹³ Therefore, it was believed to be impossible to purify formaldehyde by the distillation method.

These phenomena were explained to be due to the fact that formaldehyde makes adducts to form the polymer in the high-concentration region. Concerning this point, there are many studies concerning the physical chemistry of aqueous formaldehyde. Therefore, industrially speaking, the hemiformal method⁹⁻¹¹ is used for the purification of formaldehyde, which is very complicated and consists of four steps: formation of the hemiformal, elimination of the water, pyrolysis of the hemiformal, and, lastly, purification of crude formaldehyde.

3. THE PRINCIPLES OF FORMALDEHYDE PURIFICATION BY DISTILLATION

Thompson and Bryant¹⁴ of the DuPont group and Bezzi and Ilicet¹⁵ studied the physical chemistry of aqueous formaldehyde. Based on their research, Masamoto et al. of Asahi Chemical investigated the vapor-liquid equilibrium of aqueous formaldehyde, and they concluded that the vapor pressure of formaldehyde is determined by the free formaldehyde concentration (mol/L), whereas the vapor pressure of water is determined by the molar ratio of free water. Based on these considerations, the vapor-liquid equilibrium of formaldehyde and water in the presence of an inert solvent was calculated, and it was found that there is no azeotropic point under some special conditions, i.e., formaldehyde always has a higher volatility than that of water. This calculation was experimentally confirmed.

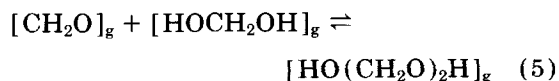
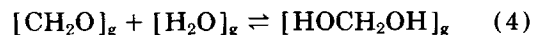
We describe next the vapor-liquid equilibrium of the formaldehyde-water system.

4. THEORY OF VAPOR-LIQUID EQUILIBRIUM OF BINARY SYSTEMS OF FORMALDEHYDE AND WATER

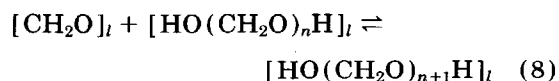
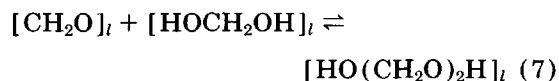
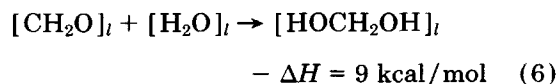
4.1. Principle

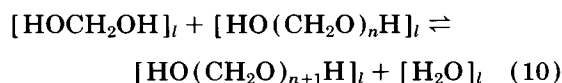
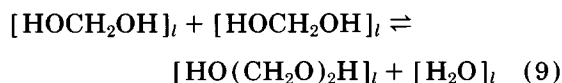
There are three phases of formaldehyde-water systems, i.e., the vapor, liquid, and solid phases. In these three phases, there exist the following chemical equilibria:

- Vapor phase

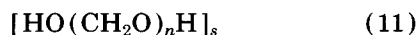


- Liquid phase



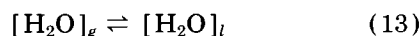
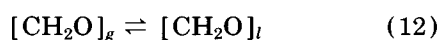


- Solid phase

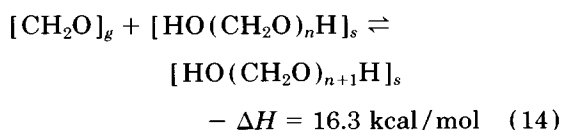


There exists a chemical equilibrium among the vapor-liquid, vapor-solid, and liquid-solid as follows:

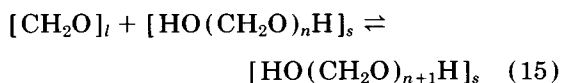
- Vapor-liquid



- Vapor-solid



- Liquid-solid



4.2. CONCEPT OF CALCULATION OF VAPOR-LIQUID EQUILIBRIUM

First, the relationships between formaldehyde concentration and distribution of $[\text{H}_2\text{O}]_l$, $[\text{CH}_2\text{O}]_l$, $[\text{HOCH}_2\text{OH}]_l$, and $[\text{HO}(\text{CH}_2\text{O})_2\text{H}]_l$ — — — — — $[\text{HO}(\text{CH}_2\text{O})_n\text{H}]_l$ were calculated. Next, the molar ratio of $[\text{H}_2\text{O}]_l$, which is indicated as $X \text{H}_2\text{O}$, was calculated.

The vapor pressure of water, $P \text{H}_2\text{O}$, can be described as follows:

$$P \text{H}_2\text{O} = X \text{H}_2\text{O} \cdot P \text{H}_2\text{O}, \text{O} \quad (16)$$

where $P \text{H}_2\text{O}, \text{O}$ is the vapor pressure of pure water. The vapor pressure of formaldehyde $P \text{CH}_2\text{O}$ is equal to the decomposition pressure of solid polyoxymethylene.

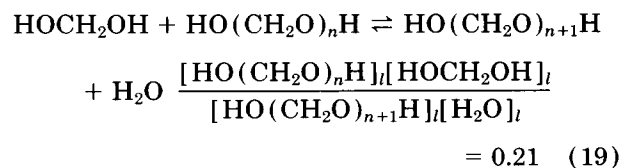
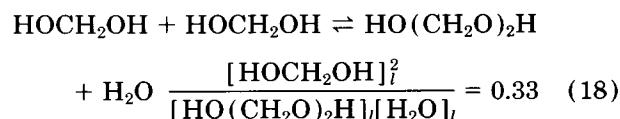
The equilibrium of the CH_2O vapor-polyoxymethylene solid can be described as follows:

$$P \text{CH}_2\text{O} = \exp \left[\frac{-16,300}{R} \left(\frac{1}{T} - \frac{1}{273 + 119} \right) \right]_{\text{atm}} \quad (17)$$

If we know the relationships between the formaldehyde concentration and the polymer precipitation equilibrium temperature, we can calculate the vapor pressure of formaldehyde using eq. (17).

Thompson and Bryant¹⁴ of the DuPont group gave the relationships between equilibrium formaldehyde concentration experimentally, where the equilibrium between the formaldehyde concentration and the solid polymer exists at a definite temperature. Relationships between equilibrium formaldehyde concentration and temperature are shown in Table I based on the experimental data of Thompson and Bryant.¹⁴

Bezzi and Iliceto¹⁵ gave the following equation for the equilibrium in the liquid phase:



Using these equations, distribution of H_2O , HOCH_2OH , $\text{HO}(\text{CH}_2\text{O})_2\text{H}$ — — — — —, and $\text{HO}(\text{CH}_2\text{O})_n\text{H}$ can be calculated. Based on these calculations, the molar ratio free water, $X \text{H}_2\text{O}$, can be calculated and is shown in Table II. Similar results have already been reported by Thompson and Bryant¹⁴ and Bezzi and Iliceto.¹⁵

4.3. Calculation of Vapor-Liquid Equilibrium of the Formaldehyde-Water System

First, we assumed that the vapor pressure of water, $P \text{H}_2\text{O}$, is proportional to the molar ratio of free water, $X \text{H}_2\text{O}$, in the aqueous formaldehyde solution and is given by eq. (16). Second, we simply assumed that the heat of vaporization of formaldehyde from an equilibrium mixture is 14.3 kcal/mol:



Table I Relationship between Equilibrium Formaldehyde Concentration and Temperature

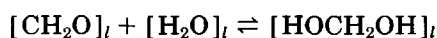
Formaldehyde Concentration (Wt %)	Temperature ¹⁴ (°C)	P CH ₂ O Calcd (atm)	P CH ₂ O Calcd (mmHg) 35°C	P CH ₂ O L ¹³ (mmHg) 35°C
10	—	—	—	1.18
20	23	0.00108	2.11	1.96
30	51	0.01200	2.85	2.56
40	67	0.03990	3.31	—
50	82	0.11100	3.75	—
60	100	0.34200	4.18	—
70	117	0.89900	4.68	—
80	135	2.29000	5.21	—
90	157	6.43000	5.60	—
95	168	10.38000	5.90	—

Thus, we can calculate the vapor pressure of formaldehyde, P CH₂O, at a definite formaldehyde concentration and definite temperature. Therefore, if the formaldehyde concentration and temperature are given, the vapor pressure of water, P H₂O, and vapor pressure of formaldehyde, P CH₂O, can be calculated and, thus, the vapor-liquid equilibrium of formaldehyde and water can be given.

The results are shown in Table III. The calculated vapor-liquid equilibrium is shown in Figure 1 and these calculations give sometimes a slightly higher value for the vapor phase of formaldehyde than does the data reported by Walker.¹³ However, calculations are almost in good accordance with the reported data in the wide range of formaldehyde concentration and temperature.

4.4. Further Consideration of Vapor Pressure of Formaldehyde

Data of free formaldehyde [CH₂O]_l are reported at 30 and 60°C by Bezzi and Iliceto.¹⁵ Based on these data, the free formaldehyde concentration, [CH₂O]_l, was calculated at different formaldehyde concentrations at 100 and 153°C, assuming the following equation:



$$\Delta H = -9 \text{ kcal/mol}$$

$$K = \frac{[\text{CH}_2\text{O}]_l [\text{H}_2\text{O}]_l}{[\text{HOCH}_2\text{OH}]_l} \quad (21)$$

Table II Distribution of H₂O, HOCH₂OH, and Σ HO(CH₂O)_nH

Formaldehyde Concentration (Wt %)	H ₂ O (mol/kg)	HOCH ₂ OH (mol/kg)	Σ HO(CH ₂ O) _n H (mol/kg)	X H ₂ O (mol/mol)	$\frac{\text{HOCH}_2\text{OH}}{\Sigma n\text{HO}(\text{CH}_2\text{O})_n\text{H}}$
0	55.60	0.00	0.00	1.00	—
5	47.70	1.37	1.51	0.97	0.820
10	41.70	2.22	2.69	0.94	0.670
20	36.10	3.08	4.65	0.89	0.460
30	34.50	3.56	5.74	0.86	0.360
40	30.00	3.57	6.53	0.82	0.270
50	23.50	3.17	6.75	0.78	0.190
60	17.50	2.60	6.58	0.73	0.130
70	11.50	1.87	5.86	0.66	0.080
80	6.05	1.07	4.62	0.57	0.040
90	2.36	0.45	3.09	0.43	0.015
95	0.79	0.16	1.83	0.30	0.005
98	0.16	0.03	0.85	0.16	0.001

Table III Vapor-Liquid Equilibrium of Formaldehyde-Water System

Formaldehyde Concentration (Wt %)	35°C			100°C			153°C			X_{H_2O}
	P_{H_2O} (mmHg)	P_{CH_2O} (mmHg)	CH_2O (Wt %)	P_{H_2O} (mmHg)	P_{CH_2O} (mmHg)	CH_2O (Wt %)	P_{H_2O} (mmHg)	P_{CH_2O} (mmHg)	CH_2O (Wt %)	
10	40	1.2	4.7	714	69	13.9	3402	766	27.3	0.94
20	38	2.0	8.0	673	115	22.1	3207	1277	39.9	0.89
30	36	2.6	10.5	651	154	28.3	3102	1737	48.3	0.86
40	35	3.3	13.8	600	200	34.8	2974	2247	55.7	0.82
50	33	3.8	16.0	591	227	39.0	2817	2545	60.0	0.78
60	31	4.2	18.4	552	260	44.0	2631	2969	65.3	0.73
70	28	4.7	21.9	503	291	49.1	2397	3324	69.8	0.66
80	24	5.2	26.6	430	324	55.7	2049	3700	75.1	0.57
90	18	5.6	33.8	329	358	64.5	1568	1568	81.6	0.43
95	13	5.9	43.6	230	377	73.2	1096	4385	87.0	0.30

The results are shown in Table IV. Plotting the free formaldehyde, $[CH_2O]_l$, and the vapor pressure of formaldehyde at 100 and 153°C, a good linear relationship was observed. These results are shown in Figures 2 and 3.

From these observations, we concluded that the vapor pressure of formaldehyde can be determined by the free formaldehyde concentration, $[CH_2O]_l$, and these considerations brought us the concept of purification of formaldehyde by distillation. We will describe this concept in the next section.

5. THEORY OF VAPOR-LIQUID EQUILIBRIUM OF FORMALDEHYDE AND WATER IN THE PRESENCE OF DILUENT

In a previous section, we calculated the vapor-liquid equilibrium of formaldehyde-water binary systems, and we obtained agreement between calculated and the reported results. In this section, we calculated the vapor-liquid equilibrium of formaldehyde and water in the presence of diluent, and we invented a novel method of purification of formaldehyde by extractive distillation.

5.1. Assumption

We first assumed that the vapor pressure of formaldehyde, P_{CH_2O} , is determined by the free formaldehyde concentration, $[CH_2O]_l$ (mol/L), and the temperature in the ternary systems containing diluent, formaldehyde, and water. Calculated values of vapor pressure of formaldehyde, free formaldehyde concentration, and equilibrium constant of 100% formaldehyde solution at various temperatures are

shown in Table V. We assumed that, in the ternary systems containing diluent, formaldehyde, and water, the vapor pressure of formaldehyde is proportional to free formaldehyde concentration (mol/L).

Next, we assumed that each rate constant or equilibrium constant has the same value as in the case of the binary systems of formaldehyde and water, i.e., relationships between vapor pressure of formaldehyde, P_{CH_2O} , and free formaldehyde, $[CH_2O]_l$, is the same for both the ternary and binary systems. We also assumed that the vapor pressure of water, P_{H_2O} , is proportional to the molar ratio of free water, X_{H_2O} , which is defined in eq. (22).

We used poly(ethylene glycol dimethyl ether)

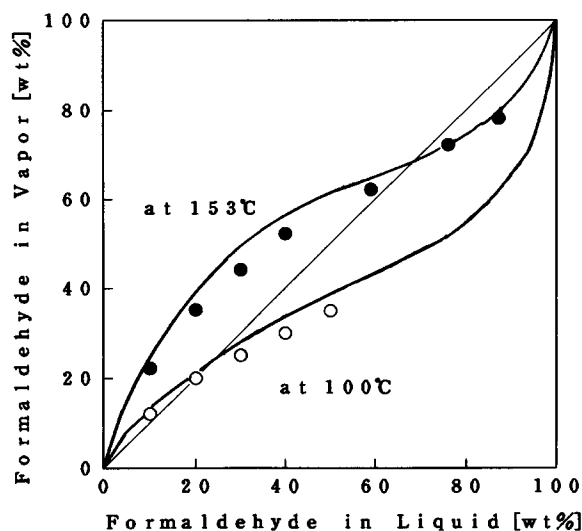


Figure 1 Vapor-liquid equilibrium of formaldehyde aqueous solution: (—) calculation; (○) (●) data from the literature.¹³

Table IV Calculation of Free Formaldehyde

Formaldehyde Concentration (Wt %)	H ₂ O (mol/L)	HOCH ₂ OH (mol/L)	[CH ₂ O] _f (Ref. 15)	Mol/L	[HOCH ₂ OH] [H ₂ O]	[CH ₂ O] _f Calcd Mol/L	
			30°C K : 0.0255	60°C K : 0.096		100°C K : 0.433	153°C K : 2.07
2.0	54.40	0.62	3.33×10^{-4}	1.33×10^{-3}	0.0114	4.94×10^{-3}	2.36×10^{-2}
5.0	51.80	1.37	6.67×10^{-4}	2.57×10^{-3}	0.0287	12.4×10^{-3}	5.94×10^{-2}
10.0	48.50	2.58			0.0532	23.0×10^{-3}	11.0×10^{-2}
15.0	45.30	3.24	17.3×10^{-4}	5.9×10^{-3}	0.0716	31.0×10^{-3}	14.8×10^{-2}
20.0	42.10	3.59			0.0853	36.9×10^{-3}	17.7×10^{-2}
28.4	36.80	3.69	40.0×10^{-4}	9.6×10^{-3}	0.1000	43.3×10^{-3}	20.7×10^{-2}
30.0	35.80	3.69			0.1030	44.6×10^{-3}	21.3×10^{-2}
40.0	29.80	3.55			0.1190	51.5×10^{-3}	24.6×10^{-2}
50.0	25.90	3.50			0.1350	58.5×10^{-3}	27.9×10^{-2}
60.0	19.80	2.95			0.1490	64.5×10^{-3}	30.8×10^{-2}
70.0	13.30	2.07			0.1620	70.1×10^{-3}	33.5×10^{-2}
80.0	7.17	1.27			0.1770	76.6×10^{-3}	36.6×10^{-2}
90.0	2.88	0.55			0.1900	82.3×10^{-3}	39.3×10^{-2}
95.0	0.98	0.20			0.1990	86.2×10^{-3}	41.2×10^{-2}
98.0	0.20	0.04			0.2100	89.2×10^{-3}	42.6×10^{-2}
100.0	0.00				0.2100	91.4×10^{-3}	43.7×10^{-2}

(PEGDME) as the diluent, and we assumed that an apparent molecular weight of PEGDME used was 1000 to obtain good agreement between calculated and observed values of the vapor pressure of water.[†]

[†] Real molecular weight of PEGDME used here was 400. However, the measurement of the vapor pressure of water in the PEGDME-water binary systems gave us an apparent molecular weight of PEGDME of 1000, assuming the vapor pressure of water is given by the molar ratio of water.

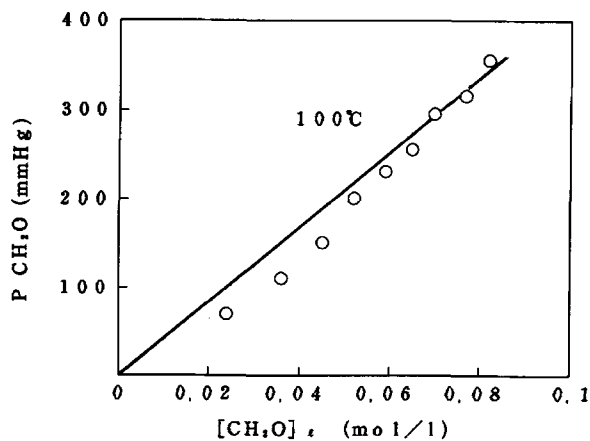


Figure 2 Relationship between free formaldehyde and vapor pressure of formaldehyde at 100°C.

5.2. Methods of Calculation

First, the temperature and, then, the total vapor pressure of formaldehyde and water were determined. Next, the vapor pressure of formaldehyde, P_{CH_2O} , and the vapor pressure of water, P_{H_2O} , were determined. Thus, the vapor-phase composition can be determined.

From these assumptions, we can calculate the concentration of free formaldehyde, $[CH_2O]_f$, and the molar ratio of free water, X_{H_2O} , in the liquid

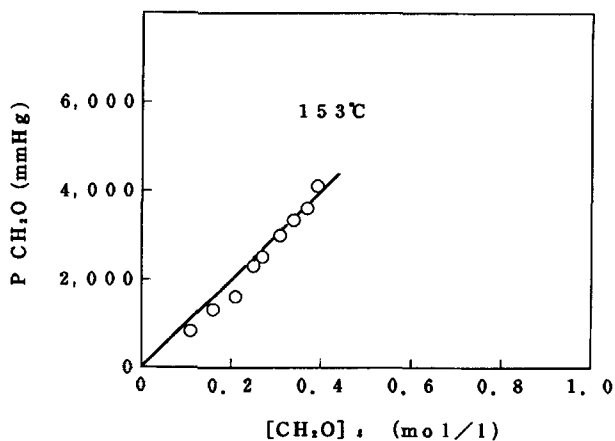


Figure 3 Relationship between free formaldehyde and vapor pressure of formaldehyde at 153°C.

Table V Vapor Pressure of Formaldehyde, Free Formaldehyde Concentration, and Equilibrium Constant of Assumed 100% Formaldehyde Solution at Various Temperatures

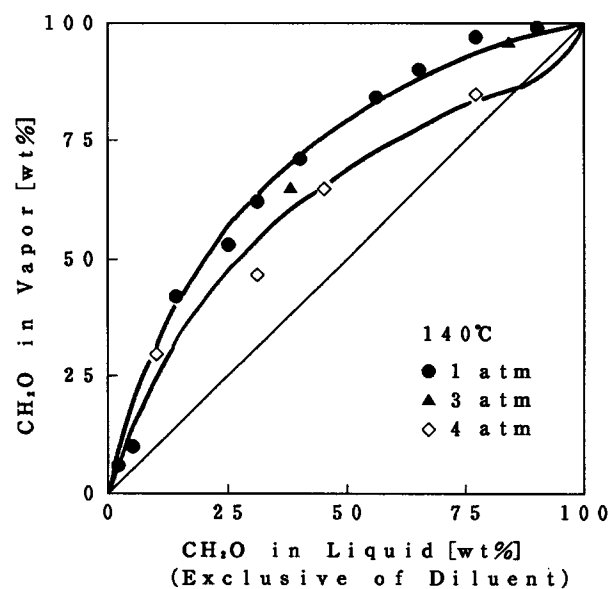
	35°C	100°C	120°C	130°C	140°C	150°C	160°C
$K : \frac{[\text{H}_2\text{O}]_l[\text{CH}_2\text{O}]_l}{[\text{HOCH}_2\text{OH}]_l}$ (mol/L)	0.0310	0.430	0.82	1.10	1.46	1.91	2.46
$P \text{ CH}_2\text{O}$ (atm)	0.0080	0.510	1.25	2.22	3.44	5.25	7.84
$[\text{CH}_2\text{O}]_l$ (mol/L)	0.0065	0.091	0.17	0.23	0.31	0.40	0.52

phase. The molar ratio of free water, $X \text{H}_2\text{O}$, is given by eq. (22):

$$X \text{H}_2\text{O} = \frac{[\text{H}_2\text{O}]_l}{[\text{H}_2\text{O}]_l + [\text{CH}_2\text{O}]_l + [\text{PEGDME}] + \sum_{n=1}^{\infty} \text{HO}(\text{CH}_2\text{O})_n\text{H}} \quad (22)$$

From the molar ratio of the free water, the $X \text{H}_2\text{O}$ value, the free water concentration of $[\text{H}_2\text{O}]_l$ can be determined. Then, from the relationships between $[\text{CH}_2\text{O}]_l$ and $[\text{H}_2\text{O}]_l$, which were described in a previous section, $[\text{HOCH}_2\text{OH}]_l$ can be determined.

From the relationships between $[\text{HOCH}_2\text{OH}]_l$ and $[\text{H}_2\text{O}]_l$, the summation of associated formaldehyde concentration, $\sum_{n=1}^{\infty} n \cdot \text{HO}(\text{CH}_2\text{O})_n\text{H}$, and total moles of water, $[\text{H}_2\text{O}]_l + \sum_{n=1}^{\infty} \text{HO}(\text{CH}_2\text{O})_n\text{H}$,

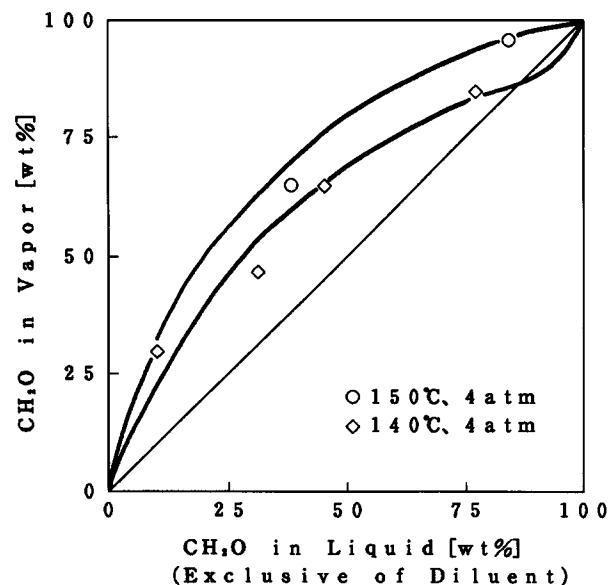
**Figure 4** Calculation of vapor-liquid equilibrium of formaldehyde and water in the presence of diluent.

can be calculated. Thus, the composition of total formaldehyde and water in the liquid phase can be determined.

5.3. Results of Calculation

The vapor-liquid equilibrium of formaldehyde and water in the presence of PEGDME was calculated at 140°C for 1, 2, 3, and 4 atm. The results are shown in Figure 4. At a temperature of 140°C, under a vapor pressure from 1 to 4 atm, it was calculated that the specific volatility of formaldehyde is higher than that of water. If the vapor pressure is elevated (actually, this means a lower concentration of diluent), an azeotropic point will appear.

Figure 5 shows the effect of temperature on vapor-liquid equilibrium. As the temperature increased from 140 to 150°C under a vapor pressure of 4 atm,

**Figure 5** Effect of temperature on vapor-liquid equilibrium of formaldehyde and water in the presence of diluent.

it was calculated that the specific volatility of formaldehyde increased. This is due to two reasons: First, at constant vapor pressure, increase of temperature means a higher concentration of diluent. Second, increase of temperature means a higher concentration of free formaldehyde.

Figure 6 shows the experimental data of the vapor-liquid equilibrium of formaldehyde and water in the presence of PEGDME. The vapor-liquid equilibrium was measured as follows: Definite amounts of aqueous formaldehyde solution was dissolved in PEGDME and then sealed in an SUS 316 microbomb. The microbomb was immersed in the oil bath at 140°C, and the vapor phase was sampled by a microsyringe and analyzed by gas chromatography. When the ratio of PEGDME/formalin increases, i.e., the weight ratio of PEGDME/formalin is 20, then there is no azeotropic point. These experimental results show good agreement with the calculations.

Experimentally, if the concentration of the diluent is reduced, i.e., vapor pressure is increased at

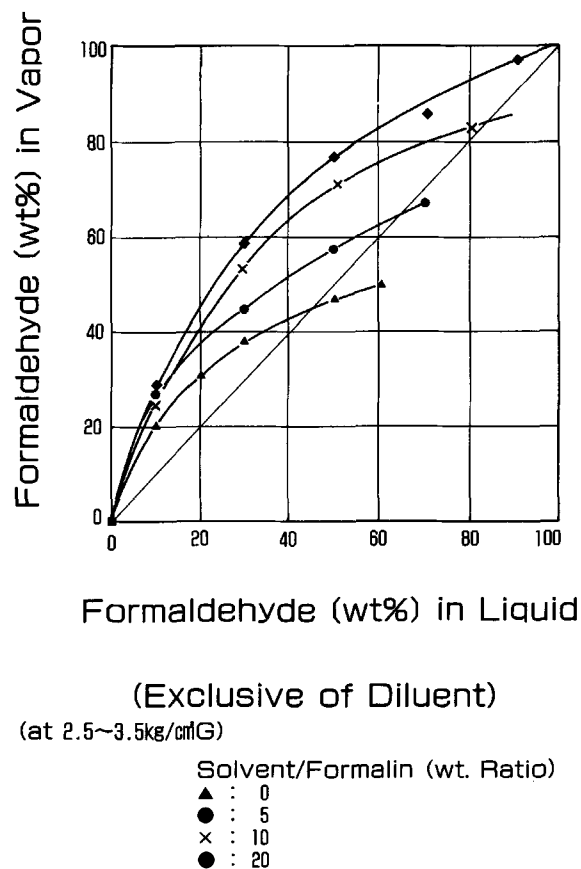


Figure 6 Vapor-liquid equilibrium of formaldehyde and water in the presence of PEGDME at 140°C.

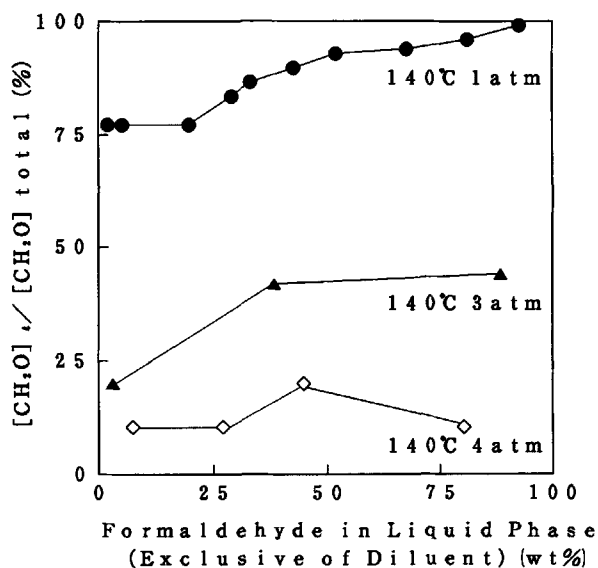


Figure 7 Calculation of free formaldehyde percentage in the presence of the diluent.

the same temperature, the azeotropic composition was observed. In our calculations, the ratio of free formaldehyde, $[\text{CH}_2\text{O}]_l$, to total formaldehyde, $[\text{CH}_2\text{O}]_l + \sum_{n=1}^{\infty} n \cdot \text{HO}(\text{CH}_2\text{O})_n\text{H}$, is shown in Figure 7.

When vapor pressure is low, i.e., the concentration of the diluent is high, the ratio of free formaldehyde is high, and with an increase in vapor pressure (i.e., a decrease in the concentration of the diluent), the ratio of free formaldehyde decreases and then the azeotropic mixture finally appears in vapor-liquid equilibrium.

6. PURIFICATION OF FORMALDEHYDE BY EXTRACTIVE DISTILLATION¹²

For the vapor-liquid equilibrium of formaldehyde and water, the azeotropic point is diminished in the presence of the diluted solvent. Using this discovery, aqueous formaldehyde can be purified by a distillation technique, i.e., an extractive distillation technique.

Figure 8 shows the principle of formaldehyde purification by the distillation method. Aqueous formaldehyde is fed to the middle (A) of the tower (1), and PEGDME, used as the extracting agent, is fed to the top (B) of the tower. Vapor is generated at the bottom of the tower by using a reboiler (2). From the top (B) of the tower, purified formaldehyde gas is obtained, and from the bottom (D), PEGDME

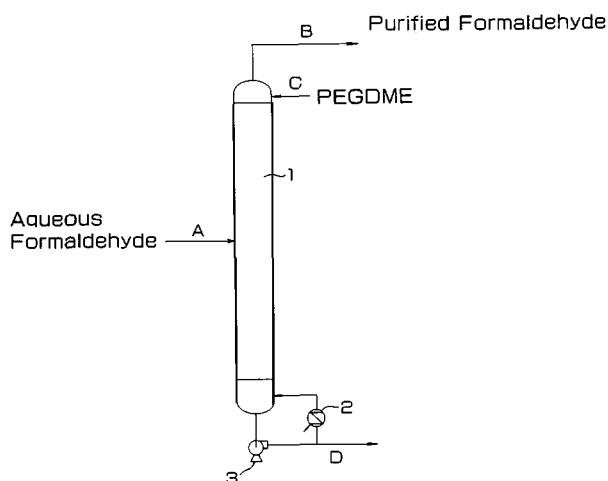


Figure 8 Principle of purification of formaldehyde by distillation: (1) tower; (2) reboiler; (3) reboiler; (A) inlet (middle of the tower); (B) outlet (top of the tower); (C) inlet (top of the tower); (D) outlet (bottom of the tower).

containing dilute aqueous formaldehyde is withdrawn. PEGDME and dilute aqueous formaldehyde are separated and PEGDME is recycled as the extracting agent. Thus, the obtained purified formaldehyde is fed to the formaldehyde polymerization process.

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