

Saturation Vapor Pressure and Critical Constants of H₂O, D₂O, T₂O, and Their Isotopic Mixtures

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Reliable data on the vapor pressure and critical constants of H₂O isotopes and their isotopic mixtures are required for the generation of thermophysical properties data over a wide range of temperatures and pressures. In this study, vapor pressure equations for D₂O and T₂O have been developed based on the latest experimental and theoretical information. Considering the similarity among H₂O isotopes, the functional form of the Saul and Wagner equation, fully proven for H₂O, has been employed. The present equation for D₂O shows a lower trend by up to 0.09% than the widely used Hill and MacMillan equation at temperatures below 150°C. For the vapor pressure of the isotopic mixtures, the available experimental data have been examined for the validity of Raoult's law. Then it has been shown that the critical temperature and the critical pressure of the isotopic mixture can also be predicted as simple mole-fraction average values.

KEY WORDS: critical constants; deuterium oxide; heavy water; isotope effect; saturation vapor pressure; tritium oxide; water.

1. INTRODUCTION

Information on the saturation vapor pressure P_s of H₂O, D₂O, and T₂O and of their isotopic mixtures is needed for research in power engineering. The critical constants of the isotopic mixtures as well as of pure H₂O, D₂O, and T₂O are essential in the prediction of other thermophysical properties by the corresponding-states principle. For pure H₂O and D₂O, experimental vapor pressure data are available in the entire temperature

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ranges from the triple points to the critical points [1–24]. The critical constants of the two isotopes have also been well defined [25]. Although, for T_2O , experimental vapor pressure data are limited to temperatures below $114^\circ C$ [21, 26] and no experimental data on the critical constants are available, the present authors predicted the vapor pressure values up to the critical point [27]. In the case of the isotopic mixtures, the vapor pressure values were measured only for the H_2O-D_2O and the H_2O-T_2O systems at temperatures below $220^\circ C$ [11, 16, 19, 20, 26, 28, 29]. As for the critical constants, the critical temperatures [30] and the critical pressures [31] of the H_2O-D_2O mixtures were studied experimentally.

Vapor pressure equations for pure H_2O , D_2O , and T_2O are valuable not only in obtaining vapor pressure values themselves but also in calculating saturation properties such as the heat of vaporization. For pure H_2O and D_2O , a number of vapor pressure equations have been published [32–40]. There exists only one vapor pressure equation for T_2O , due to the present authors [27]. In Table I, the values of the critical temperature T_c and the critical pressure P_c adopted in recent vapor pressure equations

Table I. Recent Studies on the P_s Equations for H_2O and D_2O

First author	Year	T_c (K) (68)	P_c (MPa)	Ref. No.
H_2O				
1967 IFC	1968	647.377	22.12	32
Keenan	1969	647.245	22.088	33
Ambrose	1972	647.31	22.106	34
Wagner	1973	647.25	22.093	35
Tanishita	1974	647.247	22.088	36
Watanabe	1974	647.247	22.0912	37
Aleksandrov	1981	647.073	22.0477	38
Saul	1985	647.14	22.064	39
IAPS 1983	1985	$647.14 \pm \delta_1$ ($\delta_1 = \pm 0.10$)	$22.064 + 0.27\delta_1$ ± 0.005	25
D_2O				
Tanishita	1974	— ^a	— ^a	36
Watanabe	1974	643.89	21.659	37
Hill	1979	643.89	21.66	40
Aleksandrov	1981	643.887	21.659	38
IAPS 1983	1985	$643.89 \pm \delta_2$ ($\delta_2 = \pm 0.20$)	$21.671 + 0.27\delta_2$ ± 0.010	25

^a Equation of the P_s ratio between D_2O and H_2O .

for H₂O and D₂O are compared with the latest recommendation for the critical constants of H₂O and D₂O by the International Association for the Properties of Steam (IAPS) [25]. For H₂O, equations published after the adoption of the 1967 IFC Formulation for Industrial Use [32] are listed. The latest equation by Saul and Wagner [39] adopts the T_c and P_c values in Ref. 25 and reproduces all the reliable experimental data within their claimed accuracy. In the case of D₂O, the P_c values for vapor pressure equations listed in Table I are all slightly too low, although their T_c values are accommodated to the recommendation in Ref. 25. Furthermore, recent experimental vapor pressure data for D₂O below 90°C [11, 24] revealed that these existing equations gave vapor pressure values slightly too high below 150°C.

In this study, first a vapor pressure equation for D₂O has been developed, with the latest IAPS recommendation for the critical constants [25] adopted and with the recent experimental data [11, 24] taken into account. Considering the similarity among H₂O isotopes, a functional form proven for the vapor pressure data of H₂O [39] has been used. With the same functional form, an equation for T₂O has also been developed based on the experimental vapor pressure data below 114°C [21, 26] and on our estimates [27] at temperatures from 120°C to T_c . Then experimental vapor pressure data on the isotopic mixtures, available for the H₂O–D₂O and the H₂O–T₂O systems, have been reviewed. Although the deviation from the mole-fraction average is significant at low temperatures, Raoult's law applies better with increasing temperatures. The deviation is negative for the H₂O–D₂O mixture and positive for the H₂O–T₂O system. Finally, the critical constants of the isotopic mixtures of H₂O isotopes have been estimated by assuming the validity of Raoult's law at near-critical temperatures.

2. VAPOR PRESSURE EQUATION FOR D₂O

Because of the similarity among H₂O isotopes, it is likely that a functional form proven for the vapor pressure values of one isotope applies satisfactorily also to those of others. The vapor pressure is most accurately known for H₂O, and therefore, it seems reasonable to apply the functional form for H₂O to D₂O and T₂O. In this study, the vapor pressure equations for D₂O and T₂O have been developed based on the following functional form of the Saul and Wagner equation for H₂O [39]:

$$P_s = P_c \exp[(T_c/T)(B_1\tau + B_2\tau^{1.5} + B_3\tau^3 + B_4\tau^{3.5} + B_5\tau^4 + B_6\tau^{7.5})] \quad (1)$$

where

$$\tau = 1 - T/T_c \quad (1a)$$

Table II. Experimental Studies on the Vapor Pressure of Liquid H₂O

First author	Year	Temp. (°C)	Claimed accuracy		Method	Ref. No.
			Temp. (K)	Press. (%)		
Holborn	1908–1910	0–374	0.01	0.03	Static	1
Egerton	1932	170–375	0.02	0.05	Dynamic	2
Osborne	1933	110–374	0.001	0.03	Static	3
Smith	1934	150–370	0.002	0.005	Static	4
Stimson	1969	25–100	0.0004	0.002	Static	5
Douslin	1971	–2.5–20	0.001	0.05	Static	6
Besley	1973	0–25	0.0005	0.025	Static	7
Guildner	1976	0.01	0.001	0.002	Static	8
Bottomley	1978	–14––2	0.001	0.5–1.5	Static	9
Munday	1980	0–49	0.01	0.3	Static	10
Jákli	1980	8–89	0.03	0.3	Static	11

Table III. Experimental Studies on the Vapor Pressure of Liquid D₂O

First author	Year	Temp. (°C)	Claimed accuracy		Method ^a	Sample purity	Ref. No.
			Temp. (K)	Press.			
Lewis	1933	20–110	—	13 Pa	Static (D)	>99 mol %	12
Miles	1936	25–229	0.02–0.1	13–270 Pa	Static (D)	98.1, 98.9 %	13
Riesefeld	1936	22–233	0.2	13 Pa	Static (D)	99.6 mol %	14
Niwa	1939	4–13	0.05	0.2 Pa	Static (T)	99.5 %	15
Combs	1954	10–55	—	—	Dynamic	99.8 %	16
Oliver	1956	208–371	0.01	—	Static (D)	99.87 wt %	17
Rivkin	1962	275–365	0.01–0.02	0.01 %	Static (T)	99.87 wt %	18
Kiss	1966	5–9	0.1	0.7 Pa	Static (D)	—	19
Zieborak	1966	80–222	0.001	3 Pa	Static (D)	99.5 mol %	20
Jones	1968	4–114	0.003	3 Pa	Static (D)	98.7–99.0 mol %	21
Liu	1970	106–300	0.001	10–60 Pa	Static (D)	99.85 mol %	22
Pupezin	1972	4–100	0.001	3 Pa	Static (D)	99.8 %	23
Besley	1973	5–25	0.0005	0.025 %	Static (T)	99.75 wt %	7
Bottomley	1978	–12–2	0.001	0.13 Pa	Static (D)	99.75 wt %	9
Jákli	1980	8–89	0.03	0.3 Pa	Static (D)	99.815 mol %	11
Jákli	1981	7–90	0.005	0.3 Pa	Static (D)	— ^b	24

^a D denotes differential manometry against H₂O, and T denotes measurement of the total pressure.

^b Extrapolated values to 100% D₂O are given.

and P_s is the saturation vapor pressure, P_c is the critical pressure, T_c is the critical temperature, and T is the absolute temperature in terms of IPTS—68. Among the recent vapor pressure equations for H₂O listed in Table I, only the Saul and Wagner (SW) equation [39] can reproduce the reliable experimental data of Osborne et al. [3], of Stimson [5], and of Guildner et al. [8] within their claimed accuracy. (The experimental studies on the vapor pressure of H₂O are listed in Table II.) Recently, the IAPS adopted the SW equation as the basis of its latest release on the skeleton tables for the thermodynamic properties of H₂O [41].

Experimental studies on the vapor pressure of liquid D₂O are listed in Table III. The data of Jákli and Illy [11] at 8–89°C and of Jákli and Van Hook [24] at 7–90°C appeared after the vapor pressure equations listed in Table I had been published. Jákli and Illy's data in Ref. 11 are supposed to be replaced by those in Ref. [24]. The data of Jákli and Van Hook [24] more smoothly connect to those of Zieborak [20] at 80–220°C. The latter seems to be unnoticed by the authors of the studies listed in Table I. The available data were carefully examined and the following studies were finally selected as the reliable ones: Oliver and Grisard [17], Rivkin and Akhundov [18], Zieborak [20], Liu and Lindsay [22], Besley and Bottomley [7], and Jákli and Van Hook [24].

With the new IAPS recommendation for the critical constants [25] adopted, the selected vapor pressure data for D₂O were correlated into the functional form of Eq. (1). The resulting constants are listed in Table IV (the constants of the SW equation for H₂O [39] are also listed). The vapor pressure values of D₂O were calculated by the present equation and are compared with some experimental data and equations in Fig. 1. The selected data smoothly connect to one another and the present equation

Table IV. Constant for Eq. (1)

	H ₂ O ^a	D ₂ O	T ₂ O
T_c (K)(68)	647.14	643.89	641.7 ^b
P_c (MPa)	22.064	21.671	21.41 ^b
B_1	-7.85823	-7.93842	-7.93680
B_2	1.83991	1.83689	1.74017
B_3	-11.7811	-11.9932	-11.5127
B_4	22.6705	22.5871	22.1314
B_5	-15.9393	-16.0363	-16.3809
B_6	1.77516	1.08484	1.54377

^a From Ref. 39.^b From Ref. 27.

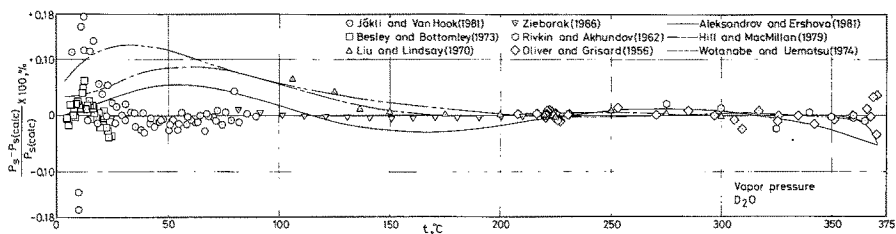


Fig. 1. Comparison of the experimental data and the equations for the vapor pressure of D_2O with the present equation.

reproduces them well. However, Liu and Lindsay's data [22] at 106 and 125°C seem to be slightly too high. Below 150°C, the previous equations compared here all show a systematically higher trend than the selected data.

The vapor pressure values of D_2O calculated with the aid of the present equation are listed in Table V, together with those for H_2O by the SW equation [39].

3. VAPOR PRESSURE EQUATION FOR T_2O

As listed in Table VI, only two sets of experimental data were reported on the vapor pressure of T_2O at temperatures below 114°C [21, 26]. By means of a modified corresponding-states principle, the present authors estimated the critical constants as well as the vapor pressure values of T_2O in the temperature range 120°C– T_c [27].

Based on the data of Jones [21] (those of Popov and Tazetdinov [26] seem less reliable) and on our estimates [27], a vapor pressure equation for T_2O has been developed in the functional form of Eq. (1). The resulting constants are listed in Table IV. In Fig. 2, the vapor pressure values of T_2O calculated by the present equation are compared with Jones' experimental data up to 114°C [21], our estimates at higher temperatures [27], and the only available vapor pressure equation for T_2O [27]. The difference between the two equations is much smaller than the uncertainty for Jones' data [21] ($\pm 0.4\%$) and for our estimates [27] (also $\pm 0.4\%$).

The vapor pressure values of T_2O were calculated with the aid of the present equation and are listed in Table V.

4. VAPOR PRESSURES OF THE ISOTOPIC MIXTURES OF H_2O , D_2O , AND T_2O

Experimental studies on the saturation vapor pressure of the mixtures among H_2O isotopes [11, 16, 19, 20, 26, 28, 29] are listed in Table VII.

Table V. Saturation Vapor Pressure of H₂O, D₂O, and T₂O

T (°C) (68)	T (K) (68)	$P_s(\text{H}_2\text{O})$ (MPa)	$P_s(\text{D}_2\text{O})$ (MPa)	$P_s(\text{T}_2\text{O})$ (MPa)
0.00	273.15	0.000611215		
0.01	273.16	0.000611659		
3.80	276.95		0.000659893	
4.49	277.64			0.000662388
10.00	283.15	0.00122792	0.00102597	0.000983341
20.00	293.15	0.00233849	0.00199813	0.00192801
30.00	303.15	0.00424510	0.00369914	0.00359076
40.00	313.15	0.00738112	0.00654370	0.00638591
50.00	323.15	0.0123446	0.0111115	0.0108949
60.00	333.15	0.0199331	0.0181839	0.0179039
70.00	343.15	0.0311777	0.0287806	0.0284417
80.00	353.15	0.0473759	0.0441947	0.0438152
90.00	363.15	0.0701207	0.0660240	0.0656417
100.00	373.15	0.101325	0.0961983	0.0958764
101.43	374.58		0.101324	
101.51	374.66			0.101312
110.00	383.15	0.143243	0.137001	0.136834
120.00	393.15	0.198483	0.191085	0.191206
130.00	403.15	0.270019	0.261484	0.262068
140.00	413.15	0.361191	0.351614	0.352888
150.00	423.15	0.475712	0.465280	0.467522
160.00	433.15	0.617659	0.606665	0.610215
170.00	443.15	0.791468	0.780326	0.785591
180.00	453.15	1.00193	0.991188	0.998643
188.74	461.89	1.21990		1.21990
190.00	463.15	1.25417	1.24453	1.25473
200.00	473.15	1.55365	1.54597	1.55956
210.00	483.15	1.90617	1.90148	1.91918
220.00	493.15	2.31782	2.31736	2.34001
220.94	494.09	2.35979	2.35979	
230.00	503.15	2.79504	2.80021	2.82876
240.00	513.15	3.34458	3.35702	3.39253
250.00	523.15	3.97350	3.99508	4.03876
260.00	533.15	4.68922	4.72207	4.77528
270.00	543.15	5.49955	5.54607	5.61034
280.00	553.15	6.41272	6.47563	6.55265
290.00	563.15	7.43746	7.51985	7.61150
300.00	573.15	8.58308	8.68848	8.79687
310.00	583.15	9.85970	9.99212	10.1196
320.00	593.15	11.2784	11.4425	11.5916
330.00	603.15	12.8515	13.0528	13.2264
340.00	613.15	14.5934	14.8383	15.0397
350.00	623.15	16.5211	16.8179	17.0508
360.00	633.15	18.6566	19.0163	19.2858
368.55	641.70			21.41
370.00	643.15	21.0329	21.4755	
370.74	643.89		21.671	
373.99	647.14	22.064		

Table VI. Experimental Studies on the Vapor Pressure of Liquid T₂O

First author	Year	Temp. (°C)	Claimed accuracy		Method ^a	Sample purity (mol %)	Ref. No.
			Temp. (K)	Press. (Pa)			
Popov	1960	12-95	0.01	4-8	Static (T)	98.1	26
Jones	1968	4-114	0.003	3	Static (D)	98.6-99.4	21

^a D denotes differential manometry against H₂O, and T denotes measurement of the total pressure.

The data are available for the H₂O-D₂O and H₂O-T₂O mixtures. In Fig. 3, some data for the H₂O-D₂O mixtures [11, 16, 20] are plotted against the mole fraction of D₂O. The vapor pressure of this system shows small negative deviations from Raoult's law at lower temperatures. With increasing temperatures, the difference between the vapor pressure values of pure H₂O and D₂O decreases and Raoult's law applies better. It is known that these deviations can be attributed entirely to the following reaction [42]:



Although the H₂O-D₂O-HDO mixture is ideal, the vapor pressure of HDO is lower than the arithmetic mean of those for H₂O and D₂O at lower temperatures.

Figure 4 shows the composition dependence of the vapor pressure of the H₂O-T₂O mixture. In contrast, with the H₂O-D₂O mixture, positive deviations from Raoult's law are observed in the data of Popov and Tazetdinov [26]. The H₂O-T₂O-HTO mixture is also supposed to be ideal. Therefore, it follows that the vapor pressure of HTO is *higher* than the arithmetic mean of those for H₂O and T₂O, although this seems rather unlikely. The deviation decreases with increasing temperatures also for this mixture.

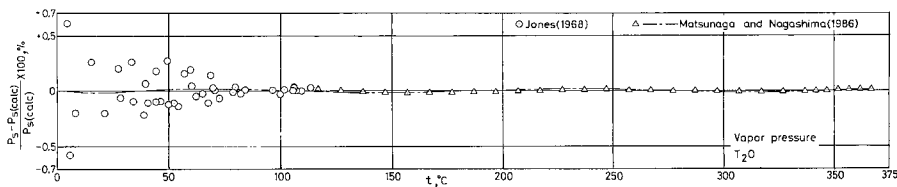
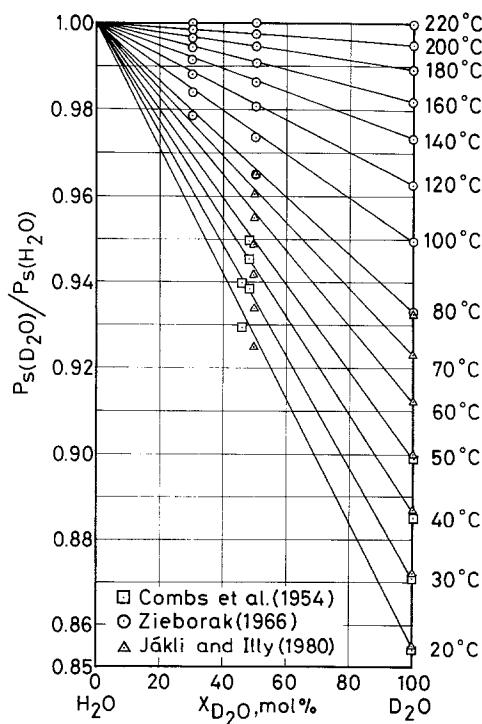


Fig. 2. Comparison of the experimental data and the equations for the vapor pressure of T₂O with the present equation.

Table VII. Experimental Studies on the Vapor Pressure of Liquid Mixtures of H₂O Isotopes

First author	Year	Mixture	Temp. (°C)	Method ^a	Composition	Ref. No.
Smith	1936	H ₂ O–D ₂ O	100	Static (D)	0.3–7 mol% D ₂ O	28
Puddington	1949	H ₂ O–D ₂ O	20	Static (D)	1.4–13 mol% D ₂ O	29
Combs	1954	H ₂ O–D ₂ O	10–55	Static (D)	46–48 mol% D ₂ O	16
Kiss	1966	H ₂ O–D ₂ O	5–9	Static (D)	50 mol% D ₂ O	19
Zieborak	1966	H ₂ O–D ₂ O	80–222	Static (D)	10–89 mol% D ₂ O	20
Jákli	1980	H ₂ O–D ₂ O	8–89	Static (D)	50 mol% D ₂ O	11
Popov	1960	H ₂ O–T ₂ O	20–95	Static (T)	83 mol% T ₂ O	26

^a D denotes differential manometry against H₂O, and T denotes measurement of the total pressure.

**Fig. 3.** Composition dependence of the vapor pressure of the H₂O–D₂O mixture.

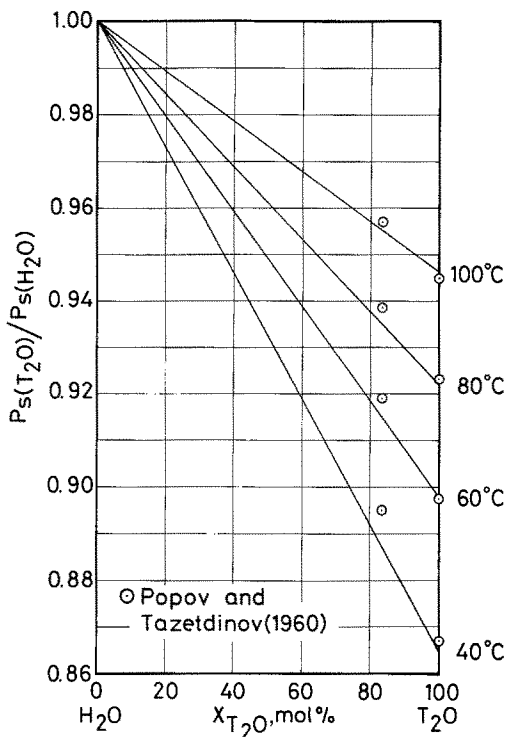


Fig. 4. Composition dependence of the vapor pressure of the H_2O - T_2O mixture.

No experimental data exist for the D_2O - T_2O and the H_2O - D_2O - T_2O mixtures. However, assuming that the D_2O - T_2O - DTO and the H_2O - D_2O - T_2O - HDO - HTO - DTO systems are also ideal and that the vapor pressure of DTO is very close to the arithmetic mean of those for D_2O and T_2O at higher temperatures, one can expect that the vapor pressures of the D_2O - T_2O and the H_2O - D_2O - T_2O mixtures also vary linearly with the mole fractions.

Figure 5 shows the ratios of the vapor pressure values of D_2O and T_2O to that of H_2O . Since the ratios remain close to unity at temperatures above the crossover temperatures, it seems reasonable to assume that the vapor pressure values of the mixtures of H_2O isotopes obey Raoult's law also at temperatures from the crossover temperatures to the critical temperatures. This assumption is applied in the following section for estimating the critical pressure of the isotopic mixtures.

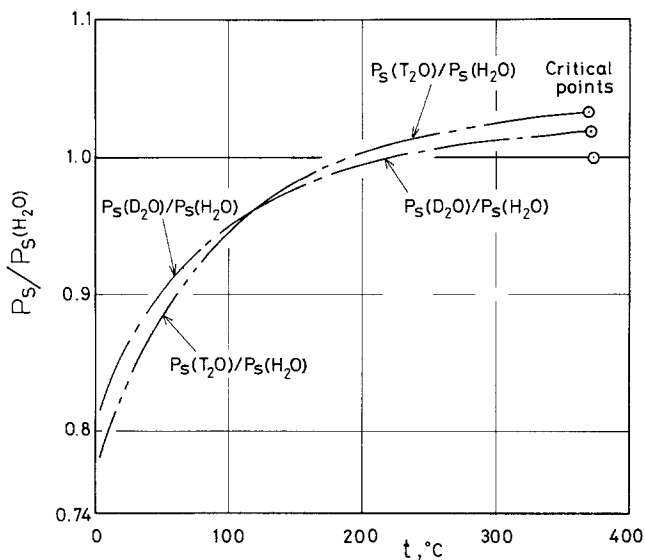


Fig. 5. Ratios of the vapor pressure of D₂O and T₂O that of H₂O.

5. CRITICAL CONSTANTS OF THE ISOTOPIC MIXTURES OF H₂O, D₂O, AND T₂O

Although the critical temperatures T_c of the mixtures are usually not mole-fraction average values [43], Riesenfeld and Chang [30] experimentally showed that the T_c of the H₂O–D₂O mixture varied linearly with the mole fraction. The linear trend of T_c may be expected also for the other isotopic mixtures.

The critical pressures P_c of mixtures are also not mole-fraction average values in most cases and are usually estimated in a quite complicated manner [43]. Although the critical pressure P_c of the H₂O–D₂O mixtures was experimentally studied by Eck [31], because of excessive scatter in his data, one cannot confirm whether P_c is a linear function of the mole fraction or not. However, the linear trend of P_c against the mole fraction can still be predicted for the isotopic mixtures of H₂O isotopes as explained below. As mentioned in the preceding section, one can expect that on an isotherm the vapor pressure P_s of the isotopic mixtures varies linearly with the mole fraction. In Fig. 6, taking the H₂O–D₂O mixture as an example, the vapor pressure values of the mixtures (20, 40, 60, and 80 mol% D₂O) thus predicted are plotted together with those for pure species on the P – T plane near the critical points. Since the critical temperature T_c of the H₂O–D₂O mixtures is empirically known to be a linear function of the

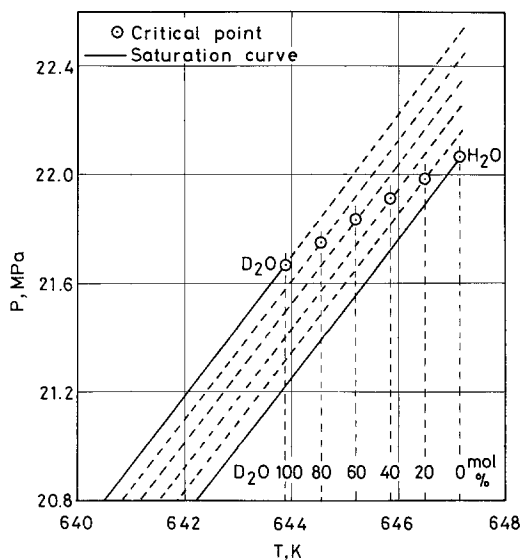


Fig. 6. Estimation of the critical points of the H_2O - D_2O mixtures.

mole fraction [30], one can graphically determine the values of the critical pressure P_c of the mixtures as shown in Fig. 6. The resulting estimates of P_c also show a linear trend against the mole fraction.

To sum up, it is concluded that both the critical temperature T_c and the critical pressure P_c of the isotopic mixtures of H_2O , D_2O , and T_2O may be estimated as simple mole-fraction average values.

The critical molar densities ρ_c of H_2O and D_2O are practically identical [44]. Extending this relationship, the present authors estimated the critical density of T_2O [27]. By further extending this assumption to the isotopic mixtures, their critical density values can readily be estimated.

6. CONCLUSIONS

Adopting the functional form of a vapor pressure equation proven for H_2O [39], those for D_2O and T_2O have been correlated based on the latest experimental and theoretical information. Then the available experimental data on the vapor pressure of the isotopic mixtures have been checked for the applicability of Raoult's law. It has been shown that the critical temperature and the critical pressure of the isotopic mixtures can also be obtained as mole-fraction average values.

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