

Vapor Pressure and Standard Enthalpies of Sublimation of Iron Difluoride, Iron Dichloride, and Iron Dibromide

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Vapor pressures of solid FeF₂, FeCl₂, and FeBr₂, were measured by the torsion and Knudsen methods and the following equations, valid in the reported temperature ranges, were derived: $\log\{p(\text{FeF}_2)/\text{kPa}\} = 10.58 \pm 0.20 - (13740 \pm 200)(\text{K}/T)$ (958–1178 K), $\log\{p(\text{FeCl}_2)/\text{kPa}\} = 10.95 \pm 0.30 - (10390 \pm 300)(\text{K}/T)$ (693–866 K), and $\log\{p(\text{FeBr}_2)/\text{kPa}\} = 11.09 \pm 0.20 - (10320 \pm 250)(\text{K}/T)$ (655–833 K). The following standard sublimation enthalpies were determined by second- and third-law treatment of the vapor pressures: $\Delta_{\text{sub}}H^\circ(298 \text{ K}) = 271 \pm 2$, 204 ± 4 , and $208 \pm 2 \text{ kJ}\cdot\text{mol}^{-1}$ for iron difluoride, dichloride, and dibromide, respectively.

Introduction

Continuing an investigation on the sublimation process of iron halides (FeI₂) Scarozza and Piacente, (1995), new vapor pressures of FeF₂, FeCl₂ and FeBr₂, were measured and their standard sublimation enthalpies evaluated.

As concerns iron difluoride, apparently the standard sublimation enthalpies available in the literature are the values proposed by Brewer et al. (1963) $314 \text{ kJ}\cdot\text{mol}^{-1}$, that measured by Kent and Margrave (1965) by mass spectrometry, $(311 \pm 5) \text{ kJ}\cdot\text{mol}^{-1}$, and the value proposed by Zhuravleva et al. (1976), $(270 \pm 3) \text{ kJ}\cdot\text{mol}^{-1}$, an average of values obtained from a second- and third-law analysis of Knudsen-effusion vapor pressure data. Unfortunately, the only two absolute vapor pressure sets available for this compound over the solid phase (Kent and Margrave, 1965; Zhuravleva et al., 1976) do not agree.

For the sublimation enthalpy of iron dichloride, literature values are the values calculated by Brewer et al. (1963), $218 \text{ kJ}\cdot\text{mol}^{-1}$, and determined from the temperature dependence of its vapor pressure by Schoonmaker and Porter (1958), $(186 \pm 12) \text{ kJ}\cdot\text{mol}^{-1}$, at 640 K, by Sime and Gregory (1960), $(198 \pm 8) \text{ kJ}\cdot\text{mol}^{-1}$, at 298 K, and by Kana'an et al. (1969), $196 \text{ kJ}\cdot\text{mol}^{-1}$, at 298 K. Vapor pressure values available above the melting point are those measured by Maier (1925) and by Schäfer et al. (1952) and two sets obtained during the study of binary systems involving this compound at high temperatures (Buryleva and Sryvalin, 1983; Burylev, 1983); below the melting point, the only vapor pressures are those reported by Schoonmaker and Porter (1958) and Kana'an et al. (1969).

For iron dibromide the only vapor pressure measurements are those of Mac Laren and Gregory (1955) who employed effusion, transpiration and diaphragm techniques to determine the enthalpy of sublimation, $(197 \pm 3) \text{ kJ}\cdot\text{mol}^{-1}$, at an approximate midpoint temperature of 750 K. Later, Sime and Gregory (1960), using a torsion-effusion apparatus, measured a new set of vapor pressures from which they derived $\Delta_{\text{sub}}H^\circ(700) = (196 \pm 8) \text{ kJ}\cdot\text{mol}^{-1}$. Another sublimation enthalpy is available in the literature, that estimated by Brewer et al. (1963), $\Delta_{\text{sub}}H^\circ(298 \text{ K}) = 207 \text{ kJ}\cdot\text{mol}^{-1}$.

For the halides studied the monomers were considered as the predominant species present in the vapor phase.

However, above the dichloride and dibromide small amounts of dimers were mass spectrometrically observed in addition to monomers (Gregory and Thackey, 1950; Schoonmaker and Porter, 1958; Porter and Schoonmaker, 1959). On the contrary Kana'an et al. (1969) measured the vapor pressure of FeCl₂ by two different methods, torsion effusion (p) (this method does not depend on the molar mass of the effused vapor) and mass effusion (π) (depends on the molar mass), and found ratios p/π ranging around the value 1.14 ± 0.01 . This leads to the conclusion that no dimer formation but rather a small amount of partial dissociation of FeCl₂ into FeCl(g) and Cl₂(g) occurs.

Experimental Section

The anhydrous iron halides, having nominal purities of about 99% for FeCl₂ and FeBr₂ and 99.5% for FeF₂, were supplied by Strem Chemicals. However, the principal impurities are more volatile than the iron halides as experimentally observed. For this reason all halide samples were purified by heating under vacuum for several hours before measurements. Vapor pressures of these compounds were measured by the mass-effusion and torsion-effusion methods. The mass-effusion method is based on the Knudsen equation (Knudsen, 1950): $p = K_K(\Delta m/t)(T/M)^{1/2}$, where T is the sample temperature, M the molar mass of the effusing vapor, K_K a constant including the geometrical characteristics of the effusion holes of the cell, and $\Delta m/t$ the rate of mass loss of the sample. Two instruments based on this method were employed: an Ugine-Eyraud Model B60 Setaram thermobalance described by Bardi et al. (1973) and a conventional Knudsen-effusion assembly. The essential part of this last apparatus is a copper block in which a conventional graphite effusion cell is inserted. The block was placed in the isothermal zone of a furnace, and its temperature was determined as an average of two values measured by two calibrated chromel–alumel thermocouples inserted into the top and the bottom of the copper block. In all experiments the two temperature values were never found to differ by more than 1 K. The uncertainty in the absolute temperature values is no larger than 3 K. An efficient pumping system and a nitrogen liquid trap assure an operative vacuum of about 10^{-4} Pa. The rate of mass loss was directly determined from the Knudsen assembly by vaporizing the sample for a known time in the isothermal experiment. Depending on the temperature, the measurements were carried out from 10 to 90 h.

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Table 1. Torsion Cell Constants

cell	material	nominal diameter of the holes d/mm	number of runs	$10^2 K_\alpha / (\text{kPa rad}^{-1})$
A	graphite	1.2	3	1.74 ± 0.07
B	graphite	0.5	4	10.28 ± 0.52
C	steel	0.8	2	5.15 ± 0.13
D	steel	1.0	4	1.52 ± 0.09

The torsion-effusion technique and apparatus have been described by Volmer (1931) and Piacente et al. (1994). The torsion pressure is related to the torsion angle (α) of a tungsten thin wire from which the cell is suspended. The pressure is determined from the relation $p = K_\alpha \alpha$, where the cell constant K_α is related to the torsion constant and length of the wire and the geometrical factors of the effusion holes of the cell. In this study, cells of different materials (graphite and stainless steel) having different diameters and thicknesses of their effusion holes were used. The torsion cell constants and the cell constants used in the mass-effusion experiment were determined (and their values checked during the study) by vaporizing pure reference elements cadmium and lead (Hultgren et al., 1973) in separate runs. The different values of the cell constant influence the final pressure values of 3–5%, an error comparable with the uncertainties usually associated with the method. The average torsion cell constants are reported in Table 1.

In order to minimize oxidation and hydrolysis of the samples, the cells were loaded in a drybox and then rapidly introduced into the apparatus filled with dry nitrogen and then quickly evacuated.

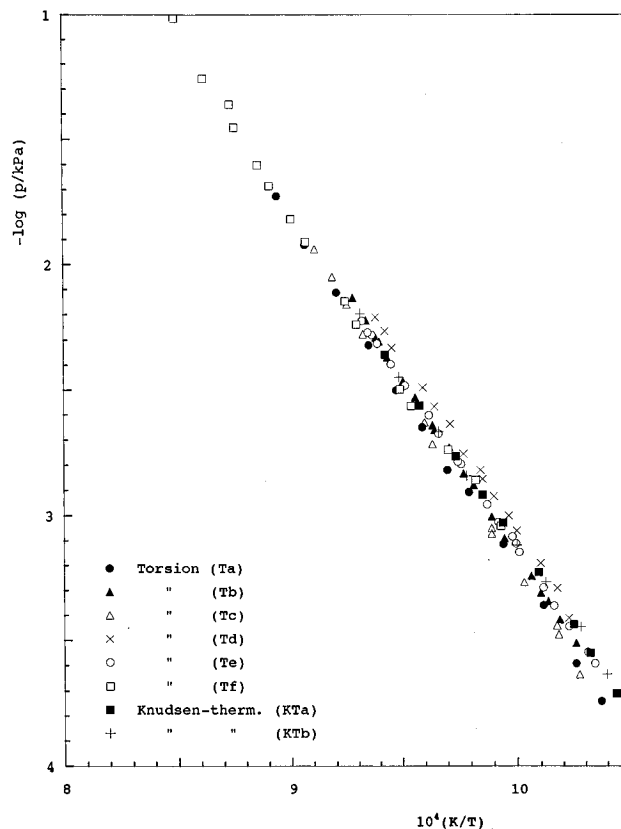
Results and Discussion

A. Iron Difluoride. The vapor pressures measured by the Knudsen and torsion methods in the temperature range 958–1178 K are drawn in Figure 1. Because of the high operating temperatures, no measurements by the conventional Knudsen assembly were made. The torsion results are reported in Table 2, those obtained by the thermobalance in Table 3. The constants of the equation $\log(p)$ vs $1/T$ calculated by a least squares treatment are reported in Table 4. Also the torsion equations obtained with the same procedure in the six experimental runs are reported in Table 4. By weighting the slopes and the intercepts of these equations proportionally to the number of points, the following equation was selected:

$$\log(p/\text{kPa}) = 10.58 \pm 0.20 - (13740 \pm 200)(\text{K}/T) \quad (1)$$

where the uncertainties were estimated assuming an error of about 3 K in the temperature measurements, about 5% in the instrument constants, and about 10% at low torsion angles ($\alpha < 5^\circ$), decreasing to 1% at higher angles ($\alpha > 50^\circ$). This equation, reported for comparison with the literature in Table 5, agrees well with the results of Zhuravleva et al. (1976). From eq 1, the sublimation enthalpy of FeF_2 , $\Delta_{\text{sub}}H^\circ(1068 \text{ K}) = (263 \pm 4) \text{ kJ}\cdot\text{mol}^{-1}$, was derived. The value at 298 K, $\Delta_{\text{sub}}H^\circ(298) = (271 \pm 4) \text{ kJ}\cdot\text{mol}^{-1}$, was obtained using the enthalpy increments of solid and gaseous phases selected by Barin (1993).

At each experimental temperature from the experimental vapor pressure and the corresponding Gibbs energy functions reported by Barin, a third-law $\Delta_{\text{sub}}H^\circ(298 \text{ K})$ value was calculated and reported in Tables 2 and 3. The average third-law value so obtained, $(271 \pm 1) \text{ kJ}\cdot\text{mol}^{-1}$, agrees with the value obtained from the second-law evaluation. In Table 5 are reported the literature third-law

**Figure 1.** Vapor pressures of iron difluoride.

$\Delta_{\text{sub}}H^\circ(298 \text{ K})$ values, and the data confirm the excellent agreement of our value with those found by Zhuravleva et al. (1976) so that the value of $\Delta_{\text{sub}}H^\circ(298 \text{ K}) = 271 \text{ kJ}\cdot\text{mol}^{-1}$ is proposed as a standard sublimation enthalpy of FeF_2 . Considering the absence of evident temperature trends in the third-law $\Delta_{\text{sub}}H^\circ(298 \text{ K})$ values and the agreement between the second- and third-law results, we believe that the uncertainty associated with this enthalpy value should not exceed $\pm 2 \text{ kJ}\cdot\text{mol}^{-1}$.

B. Iron Dichloride. The vapor pressure of iron dichloride was measured by both the Knudsen-effusion and the torsion-effusion assemblies in the range 693–866 K. The data obtained in six torsion runs are reported in Table 6, the results obtained by one thermobalance run are reported in Table 3, and those obtained by the Knudsen assembly are reported in Table 7. All the experimental vapor pressures above solid FeCl_2 are given in Figure 2. The pressure–temperature equations reported in Table 4 were calculated by a least squares treatment of the results of each run.

No evident change in the slopes of the equations obtained from the torsion and the Knudsen results over different temperature ranges was observed; therefore, $\log(p)$ was considered a linear function of $1/T$ in the overall temperature range, and the following equation gives the temperature dependence of the vapor pressure over solid FeCl_2 :

$$\log(p/\text{kPa}) = 10.95 \pm 0.30 - (10390 \pm 300)(\text{K}/T) \quad (2)$$

where the errors were overestimated considering the relative large temperature range in which this equation is valid. The results of this equation is reported in Table 5 for comparison with the literature data. There is substantial agreement between our vapor pressure value calculated at the melting point (950 K), $p \approx 1 \text{ kPa}$, and the values calculated at the same temperature from the equations given by Scafer, Buryleva, and Burylev all obtained over

Table 2. Vapor Pressures of FeF₂ Measured by Torsion Method and Third-Law $\Delta_{\text{sub}}H^{\circ}(298 \text{ K})$

run	<i>T</i> /K	α /rad	<i>p</i> /kPa	$-\Delta_{\text{sub}}[G^{\circ}(T) - H^{\circ}(298 \text{ K})]/(\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1})$	$\Delta_{\text{sub}}H^{\circ}(298 \text{ K})/(\text{kJ}\cdot\text{mol}^{-1})$	run	<i>T</i> /K	α /rad	<i>p</i> /kPa	$-\Delta_{\text{sub}}[G^{\circ}(T) - H^{\circ}(298 \text{ K})]/(\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1})$	$\Delta_{\text{sub}}H^{\circ}(298 \text{ K})/(\text{kJ}\cdot\text{mol}^{-1})$		
Ta	964.0	1.75×10^{-3}	1.80×10^{-4}	172.7	272.6	Td	978.0	7.48×10^{-3}	3.85×10^{-4}	172.6	270.3		
	975.0	2.48×10^{-3}	2.55×10^{-4}	172.6	272.8		983.0	9.86×10^{-3}	5.08×10^{-4}	172.5	269.3		
	989.0	4.22×10^{-3}	4.33×10^{-4}	172.5	272.2		990.0	1.24×10^{-2}	6.39×10^{-4}	172.5	269.3		
	1006.0	7.44×10^{-3}	7.65×10^{-4}	172.3	272.0		1000.0	1.67×10^{-2}	8.62×10^{-4}	172.4	269.4		
	1021.0	1.19×10^{-2}	1.22×10^{-3}	172.2	271.9		1003.5	1.92×10^{-2}	9.90×10^{-4}	172.3	269.2		
	1031.0	1.46×10^{-2}	1.50×10^{-3}	172.1	272.8		1010.0	2.30×10^{-2}	1.18×10^{-3}	172.3	269.4		
	1043.0	2.16×10^{-2}	2.22×10^{-3}	172.0	272.4		1015.0	2.68×10^{-2}	1.38×10^{-3}	172.2	269.4		
	1055.5	3.05×10^{-2}	3.13×10^{-3}	171.9	272.5		1016.0	2.91×10^{-2}	1.50×10^{-3}	172.2	268.9		
	1069.5	4.61×10^{-2}	4.74×10^{-3}	171.7	272.3		1023.5	3.38×10^{-2}	1.74×10^{-3}	172.2	269.6		
	1086.5	7.44×10^{-2}	7.65×10^{-3}	171.6	272.1		1029.5	4.46×10^{-2}	2.29×10^{-3}	172.1	268.7		
	1103.0	1.15×10^{-1}	1.18×10^{-2}	171.4	272.1		1037.0	5.22×10^{-2}	2.69×10^{-3}	172.0	269.2		
	1118.5	1.81×10^{-1}	1.86×10^{-2}	171.3	271.5		1042.5	6.22×10^{-2}	3.20×10^{-3}	172.0	269.1		
	Tb	975.0	2.98×10^{-3}	3.06×10^{-4}	172.6		271.3	1057.5	8.99×10^{-2}	4.63×10^{-3}	171.8	269.6	
		982.0	3.70×10^{-3}	3.81×10^{-4}	172.5		271.4	1061.0	1.05×10^{-1}	5.39×10^{-3}	171.8	269.1	
		987.0	4.38×10^{-3}	4.50×10^{-4}	172.5		271.4	1066.0	1.19×10^{-1}	6.11×10^{-3}	171.8	269.2	
		990.0	4.71×10^{-3}	4.84×10^{-4}	172.5		271.6	Te	967.0	1.67×10^{-2}	2.55×10^{-4}	172.7	270.7
		994.0	5.52×10^{-3}	5.67×10^{-4}	172.4		271.3		970.0	1.85×10^{-2}	2.82×10^{-4}	172.7	270.7
1005.5		7.83×10^{-3}	8.05×10^{-4}	172.3	271.4	978.0	2.35×10^{-2}		3.57×10^{-4}	172.6	270.9		
1011.0		9.55×10^{-3}	9.82×10^{-4}	172.3	271.2	984.5	2.84×10^{-2}		4.32×10^{-4}	172.5	271.1		
1019.0		1.27×10^{-2}	1.31×10^{-3}	172.2	270.9	989.0	3.36×10^{-2}		5.10×10^{-4}	172.5	270.9		
1023.5		1.41×10^{-2}	1.45×10^{-3}	172.2	271.1	999.0	4.66×10^{-2}		7.08×10^{-4}	172.4	270.8		
1030.0		1.79×10^{-2}	1.84×10^{-3}	172.1	270.7	1000.5	5.06×10^{-2}		7.69×10^{-4}	172.4	270.5		
1037.0		2.11×10^{-2}	2.17×10^{-3}	172.0	271.1	1002.0	5.38×10^{-2}		8.18×10^{-4}	172.4	270.4		
1038.0		2.20×10^{-2}	2.26×10^{-3}	172.0	271.0	1008.0	6.14×10^{-2}		9.33×10^{-4}	172.3	270.9		
1046.0		2.84×10^{-2}	2.92×10^{-3}	171.9	270.8	1013.0	7.21×10^{-2}		1.10×10^{-3}	172.3	270.8		
1052.0		3.30×10^{-2}	3.39×10^{-3}	171.9	271.0	1024.5	1.04×10^{-1}		1.58×10^{-3}	172.1	270.6		
1060.0		4.11×10^{-2}	4.22×10^{-3}	171.8	271.0	1026.0	1.07×10^{-1}		1.62×10^{-3}	172.1	270.8		
1064.0		4.76×10^{-2}	4.90×10^{-3}	171.8	270.7	1035.0	1.38×10^{-1}		2.09×10^{-3}	172.0	270.9		
1066.0		4.94×10^{-2}	5.08×10^{-3}	171.8	270.8	1039.5	1.63×10^{-1}		2.48×10^{-3}	172.0	270.6		
1071.0	5.76×10^{-2}	5.92×10^{-3}	171.7	270.7	1046.0	1.79×10^{-1}	2.73×10^{-3}		171.9	271.4			
1078.0	7.09×10^{-2}	7.29×10^{-3}	171.6	270.5	1051.0	2.15×10^{-1}	3.26×10^{-3}		171.9	271.0			
Tc	973.5	2.23×10^{-3}	2.29×10^{-4}	172.6	273.3	1058.0	2.62×10^{-1}		3.98×10^{-3}	171.8	271.0		
	982.5	3.22×10^{-3}	3.31×10^{-4}	172.5	272.7	1065.0	3.16×10^{-1}	4.80×10^{-3}	171.8	271.1			
	983.0	3.50×10^{-3}	3.60×10^{-4}	172.5	272.2	1068.0	3.42×10^{-1}	5.20×10^{-3}	171.7	271.1			
	997.0	5.21×10^{-3}	5.35×10^{-4}	172.4	272.6	1070.0	3.50×10^{-1}	5.32×10^{-3}	171.7	271.4			
	1011.0	8.19×10^{-3}	8.41×10^{-4}	172.3	272.5	1073.0	3.89×10^{-1}	5.92×10^{-3}	171.7	271.2			
	1011.5	8.58×10^{-3}	8.83×10^{-4}	172.3	272.2	Tf	1007.5	8.78×10^{-3}	9.03×10^{-4}	172.3	271.0		
	1038.0	1.85×10^{-2}	1.90×10^{-3}	172.0	272.5		1018.0	1.33×10^{-2}	1.37×10^{-3}	172.2	270.2		
	1042.0	2.26×10^{-2}	2.32×10^{-3}	172.0	271.8		1030.5	1.75×10^{-2}	1.80×10^{-3}	172.1	271.0		
	1055.0	3.08×10^{-2}	3.16×10^{-3}	171.9	272.3		1048.0	2.63×10^{-2}	2.70×10^{-3}	171.9	271.9		
	1072.5	5.09×10^{-2}	5.23×10^{-3}	171.7	272.2		1053.5	3.07×10^{-2}	3.15×10^{-3}	171.9	272.0		
	1080.5	6.66×10^{-2}	6.85×10^{-3}	171.6	271.7		1076.0	5.55×10^{-2}	5.71×10^{-3}	171.7	272.2		
	1088.5	8.58×10^{-2}	8.82×10^{-3}	171.5	271.3		1082.0	6.86×10^{-2}	7.06×10^{-3}	171.6	271.8		
	1098.0	1.11×10^{-1}	1.14×10^{-2}	171.5	271.3		1102.5	1.19×10^{-1}	1.22×10^{-2}	171.4	271.7		
							1110.5	1.46×10^{-1}	1.50×10^{-2}	171.3	271.7		
							1122.5	1.99×10^{-1}	2.04×10^{-2}	171.2	271.6		
							1129.0	2.41×10^{-1}	2.48×10^{-2}	171.2	271.3		
							1142.5	3.39×10^{-1}	3.48×10^{-2}	171.0	271.2		
					1145.0		4.19×10^{-1}	4.31×10^{-2}	171.0	269.7			
					1160.5		5.31×10^{-1}	5.45×10^{-2}	170.9	270.9			
					1178.0		9.31×10^{-1}	9.57×10^{-2}	170.7	269.3			

the liquid phase (1.2, 1.6, and 1.5 kPa, respectively). From our selected equation was calculated the second-law sublimation enthalpy, $\Delta_{\text{sub}}H^{\circ}(770 \text{ K}) = (200 \pm 6) \text{ kJ}\cdot\text{mol}^{-1}$, corrected to 298 K by using Barin's enthalpic functions, $\Delta_{\text{sub}}H^{\circ}(298 \text{ K}) = (210 \pm 6) \text{ kJ}\cdot\text{mol}^{-1}$. From the same source were taken the Gibbs energy functions of both gaseous and solid phases necessary for the third-law calculations. The third-law $\Delta_{\text{sub}}H^{\circ}(298 \text{ K})$ values calculated from the experimental vapor pressures are reported in Tables 3, 6, and 7. Unfortunately our third-law result, $\Delta_{\text{sub}}H^{\circ}(298 \text{ K}) = (201 \pm 1) \text{ kJ}\cdot\text{mol}^{-1}$, is lower than the second-law value, and their single values exhibit a very small temperature dependence. In better agreement are the second- and third-law $\Delta_{\text{sub}}H^{\circ}(298 \text{ K})$ values reported by Sime and Gregory. However, our absolute vapor pressures are in substantial agreement with those reported in the literature over the same temperature ranges. The agreement in the pressure values and the use of the same Gibbs energy functions lead of course to third-law results practically coincident. The small differences between our slopes and those in the literature of the pressure-temperature equations are the cause of the discrepancy between the second- and third-law results. The slopes of our eight equations measured by different techniques and in different temperature ranges

are in good agreement, and an accurate analysis of the error sources in the measurements leads to exclusion of large uncertainties in the corresponding second-law $\Delta_{\text{sub}}H^{\circ}(T)$. For this reason we believe the discrepancy between the second- and third-law sublimation enthalpies is probably due to small errors in the thermodynamic functions selected by Barin (1993) for FeCl₂ and used by us in the reduction to 298 K of the second-law $\Delta_{\text{sub}}H^{\circ}(T)$ and in the third-law evaluations of $\Delta_{\text{sub}}H^{\circ}(298 \text{ K})$. On the basis of these considerations, we propose as the standard sublimation enthalpy of FeCl₂ the value of $(204 \pm 4) \text{ kJ}\cdot\text{mol}^{-1}$.

C. Iron Dibromide. The vapor pressure values of FeBr₂ obtained by both Knudsen-effusion assemblies are reported in Tables 3 and 7. The least squares fit to $\log(p)$ vs $1/T$ equations is reported in Table 4. In the same table are also reported the data from equations determined in four torsion-effusion runs. The experimental torsion data are reported in Table 8 and, with the Knudsen data, in Figure 3. From the final equations the following equation, valid in the overall temperature range 655–833 K, was selected:

$$\log(p/\text{kPa}) = 11.09 \pm 0.20 - (10320 \pm 250)(\text{K}/T) \quad (3)$$

where the uncertainties were estimated on the basis of the

Table 3. Vapor Pressures of FeF₂, FeCl₂ and FeBr₂ Obtained by Thermobalance Measurements and Third-Law $\Delta_{\text{sub}}H^\circ(298\text{ K})$ (Estimated Error in T , $\pm 1\text{ K}$)

compound	run	T/K	$(\Delta m/\delta)/(g\text{ s}^{-1})$	p/kPa	$-\Delta_{\text{sub}}[G^\circ(T) - H^\circ(298\text{ K})]/$ ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)	$\Delta_{\text{sub}}H^\circ(298\text{ K})/$ ($\text{kJ}\cdot\text{mol}^{-1}$)		
FeF ₂	KTa	958	1.78×10^{-7}	1.93×10^{-4}	172.8	270.4		
		969	2.56×10^{-7}	2.79×10^{-4}	172.7	270.5		
		976	3.32×10^{-7}	3.64×10^{-4}	172.6	270.2		
		991	5.33×10^{-7}	5.88×10^{-4}	172.5	270.3		
		1006	8.38×10^{-7}	9.31×10^{-4}	172.3	270.4		
		1015	1.07×10^{-6}	1.20×10^{-3}	172.2	270.6		
		1027	1.51×10^{-6}	1.70×10^{-3}	172.1	270.7		
		1044	2.39×10^{-6}	2.71×10^{-3}	172.0	270.9		
	KTb	1061	3.81×10^{-6}	4.35×10^{-3}	171.8	271.0		
		962	2.11×10^{-7}	2.30×10^{-4}	172.7	270.1		
		973	3.25×10^{-7}	3.55×10^{-4}	172.6	269.6		
		988	4.87×10^{-7}	5.37×10^{-4}	172.5	270.2		
		1000	6.82×10^{-7}	7.56×10^{-4}	172.4	270.5		
		1022	1.27×10^{-6}	1.42×10^{-3}	172.2	270.9		
		1035	1.89×10^{-6}	2.13×10^{-3}	172.0	270.7		
		1054	3.11×10^{-6}	3.54×10^{-3}	171.9	271.1		
		1074	5.48×10^{-6}	6.30×10^{-3}	171.7	270.9		
FeCl ₂	KT	693	1.39×10^{-7}	1.11×10^{-4}	175.8	200.9		
		703	1.92×10^{-7}	1.54×10^{-4}	175.6	201.7		
		710	2.68×10^{-7}	2.16×10^{-4}	175.5	201.7		
		714	3.66×10^{-7}	2.95×10^{-4}	175.4	200.9		
		718	3.78×10^{-7}	3.17×10^{-4}	175.3	201.6		
		721	4.67×10^{-7}	3.79×10^{-4}	175.3	201.3		
		723	5.12×10^{-7}	4.15×10^{-4}	175.2	201.3		
		727	6.24×10^{-7}	5.08×10^{-4}	175.2	201.1		
		731	7.80×10^{-7}	6.37×10^{-4}	175.1	200.8		
		739	1.07×10^{-6}	8.80×10^{-4}	175.0	200.9		
		742	1.22×10^{-6}	1.00×10^{-3}	174.9	200.9		
		748	1.36×10^{-6}	1.13×10^{-3}	174.8	201.7		
		754	2.02×10^{-6}	1.68×10^{-3}	174.7	200.8		
		761	2.68×10^{-6}	2.23×10^{-3}	174.6	200.7		
		FeBr ₂	KTa	655	5.22×10^{-8}	3.09×10^{-5}	191.4	207.1
				675	1.62×10^{-7}	9.76×10^{-5}	191.0	206.7
				703	5.48×10^{-7}	3.59×10^{-4}	190.5	207.3
714	8.77×10^{-7}			5.42×10^{-4}	190.3	207.9		
725	1.60×10^{-6}			9.95×10^{-4}	190.0	207.3		
733	2.05×10^{-6}			1.28×10^{-3}	189.9	207.9		
761	6.52×10^{-6}			4.16×10^{-3}	189.3	208.0		
KTb	671			1.33×10^{-7}	7.96×10^{-5}	191.1	206.6	
	686			2.79×10^{-7}	1.69×10^{-4}	190.8	206.8	
	695			3.96×10^{-7}	2.41×10^{-4}	190.6	207.3	
	709	9.14×10^{-7}	5.63×10^{-4}	190.4	206.3			
		722	1.39×10^{-6}	8.65×10^{-4}	190.1	207.3		
		741	3.35×10^{-6}	2.11×10^{-3}	189.7	207.0		
		750	3.98×10^{-6}	2.52×10^{-3}	189.6	208.3		

Table 4. Experimental Vapor Pressure Data of Iron Halides

compound	assembly	run	crucible	$\Delta T/\text{K}$	no of points	$\log(p/\text{kPa}) = A - B(\text{K}/T)$	
						A ^a	B ^a
FeF ₂	torsion	Ta	B	964–1118	12	10.71 ± 0.10	13931 ± 100
	torsion	Tb	B	975–1078	18	10.92 ± 0.09	14086 ± 90
	torsion	Tc	B	973–1098	13	11.03 ± 0.12	14268 ± 120
	torsion	Td	C	978–1066	15	10.77 ± 0.18	13837 ± 189
	torsion	Te	D	967–1073	21	10.25 ± 0.10	13392 ± 107
	torsion	Tf	B	1007–1178	15	10.64 ± 0.20	13815 ± 212
	thermobalance	KTa	E	958–1061	9	10.16 ± 0.09	13283 ± 92
FeCl ₂	thermobalance	KTb	E	962–1074	8	9.92 ± 0.14	13036 ± 144
	torsion	Ta	B	758–820	11	11.14 ± 0.06	10499 ± 50
	torsion	Tb	A	789–866	9	11.47 ± 0.12	10821 ± 94
	torsion	Tc	B	751–841	12	10.78 ± 0.33	10266 ± 260
	torsion	Td	B	761–840	7	10.44 ± 0.30	10022 ± 241
	torsion	Te	C	745–828	9	10.70 ± 0.44	10163 ± 348
	torsion	Tf	D	743–840	15	10.79 ± 0.22	10347 ± 173
FeBr ₂	thermobalance	KT	E	693–761	14	10.89 ± 0.27	10317 ± 194
	Knudsen	K	F	694–745	8	11.42 ± 0.21	10746 ± 152
	torsion	Ta	B	742–819	11	11.39 ± 0.22	10546 ± 170
	torsion	Tb	B	728–811	13	11.54 ± 0.20	10699 ± 155
	torsion	Tc	D	757–810	10	10.54 ± 0.24	9886 ± 267
	torsion	Td	C	741–833	19	11.43 ± 0.25	10620 ± 193
	thermobalance	KTa	E	655–761	7	10.64 ± 0.20	9916 ± 140
thermobalance	KTb	E	671–750	7	10.52 ± 0.40	9805 ± 281	
Knudsen	K	F	694–745	6	10.84 ± 0.20	10113 ± 144	

^a The quoted errors are standard deviations.

Table 5. Comparison of Vapor Pressures and Sublimation Enthalpies of Iron Halides

compd	method	$\Delta T/K$	\bar{T}/K	$\log(p/\text{kPa}) = A - B(K/T)$		$\Delta_{\text{sub}}H^\circ(\bar{T})/(\text{kJ mol}^{-1})$	$\Delta_{\text{sub}}H^\circ(298\text{ K})/(\text{kJ mol}^{-1})$	
				A	B		second law ^g	third law
FeF ₂	MS ^a	965–1149	1057	11.42 ± 0.24	15820 ± 260	303 ± 5	311 ± 5	296
	Knudsen ^b	848–1142	995	10.604 ± 0.151	13724 ± 153	263 ± 3	270 ± 3	270 ± 1
	this work	958–1178	1068	10.58 ± 0.20	13740 ± 200	263 ± 4	271 ± 4	271 ± 1
FeCl ₂	MS ^c	621–658	640	10.08 ^h	9696 ^h	186 ± 12	193 ± 12	201 ± 1
	torsion ^d	694–745	719	10.22	9890	189 ± 8	198 ± 8	203 ± 1
	torsion ^e		780	11.10 ± 0.05	10532 ± 39	196 ^k	206	202 ± 2
	Knudsen ^{e,i}		780	11.10 ± 0.07	10571 ± 52			
	Knudsen ^{e,j}		780	11.05 ± 0.15	10497 ± 115			
	this work	693–866	769	10.95 ± 0.30	10390 ± 300	200 ± 6	210 ± 6	201 ± 1
FeBr ₂	transpiration ^f	673–962	817	11.16	10294	197 ± 2	210 ± 2	207.5
	effusion ^f	623–718	670	11.00	10300	197 ± 4	206 ± 4	210
	torsion ^d	680–720	700	11.07	10220	196 ± 8	205 ± 8	207
	this work	655–833	744	11.09 ± 0.20	10320 ± 250	197 ± 5	208 ± 5	209 ± 1

^a Kent and Margrave (1965). ^b Zhuravleva et al. (1976). ^c Schoonmaker and Porter (1958). ^d Sime and Gregory (1960). ^e Kana'an et al. (1969). ^f Mac Laren and Gregory (1955). ^g Values recalculated from $\Delta_{\text{sub}}H^\circ(\bar{T})$ using the enthalpy increments reported by Barin. ^h Calculated by us from six experimental vapor pressures reported by Schoonmaker and Porter (1958). ⁱ Measured by using a molybdenum cell. ^j Measured by using an iron cell. ^k Value proposed by the authors; the slopes of their pressure–temperature equations lead to $\Delta_{\text{sub}}H^\circ(T)$ values decidedly higher than 196 kJ·mol⁻¹.

Table 6. Vapor Pressures of FeCl₂ Measured by the Torsion Method and Third-Law $\Delta_{\text{sub}}H^\circ(298\text{ K})$

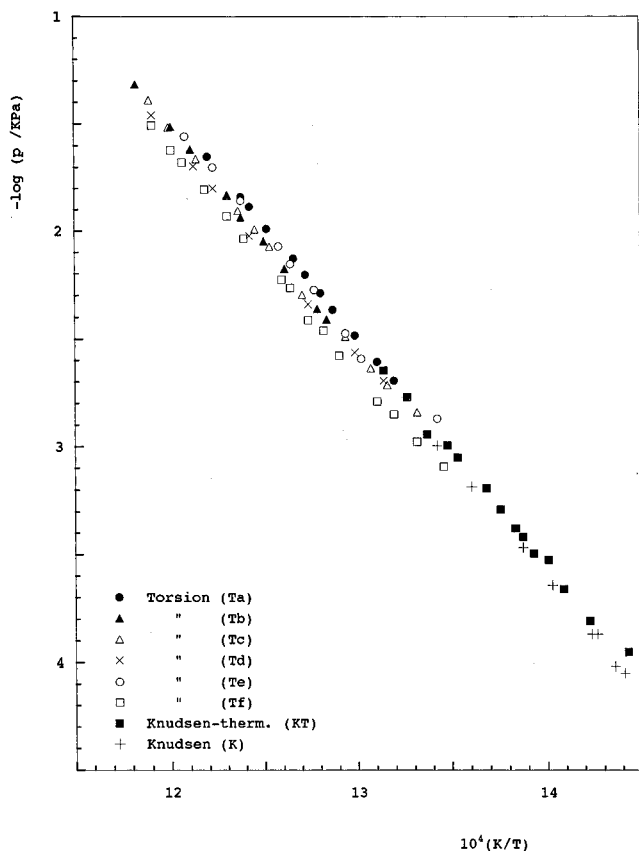
run	T/K	α/rad	p/kPa	$-\Delta_{\text{sub}}[G^\circ(T) - H^\circ(298\text{ K})]/(\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1})$	$\Delta_{\text{sub}}H^\circ(298\text{ K})/(\text{kJ}\cdot\text{mol}^{-1})$	run	T/K	α/rad	p/kPa	$-\Delta_{\text{sub}}[G^\circ(T) - H^\circ(298\text{ K})]/(\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1})$	$\Delta_{\text{sub}}H^\circ(298\text{ K})/(\text{kJ}\cdot\text{mol}^{-1})$
Ta	758	1.94×10^{-2}	2.00×10^{-3}	174.7	200.7	Td	761	1.94×10^{-2}	2.00×10^{-3}	174.6	201.4
	763	2.38×10^{-2}	2.45×10^{-3}	174.6	200.6	770	2.64×10^{-2}	2.72×10^{-3}	174.5	201.7	
	770	3.17×10^{-2}	3.26×10^{-3}	174.5	200.6	785	4.40×10^{-2}	4.57×10^{-3}	174.2	202.1	
	777	4.16×10^{-2}	4.27×10^{-3}	174.3	200.5	805	9.11×10^{-2}	9.37×10^{-3}	173.9	202.1	
	781	4.95×10^{-2}	5.09×10^{-3}	174.3	200.4	818	1.53×10^{-1}	1.57×10^{-2}	173.6	201.7	
	786	6.01×10^{-2}	6.18×10^{-3}	174.2	200.3	825	1.94×10^{-1}	2.00×10^{-2}	173.5	201.7	
	790	7.16×10^{-2}	7.36×10^{-3}	174.1	200.1	840	3.33×10^{-1}	3.43×10^{-2}	173.3	201.4	
	799	9.83×10^{-2}	1.01×10^{-2}	174.0	200.2	Te	745	2.59×10^{-2}	1.33×10^{-3}	174.9	199.9
	805	1.25×10^{-1}	1.28×10^{-2}	173.9	200.0		768	4.92×10^{-2}	2.53×10^{-3}	174.5	201.7
	808	1.39×10^{-1}	1.43×10^{-2}	173.8	200.0	773	6.47×10^{-2}	3.33×10^{-3}	174.4	201.2	
820	2.15×10^{-1}	2.21×10^{-2}	173.6	199.8	783	1.02×10^{-1}	5.27×10^{-3}	174.2	200.6		
Tb	779	2.22×10^{-1}	3.85×10^{-3}	174.3	201.7	791	1.35×10^{-1}	6.93×10^{-3}	174.1	200.8	
	782	2.48×10^{-1}	4.31×10^{-3}	174.3	201.7	795	1.63×10^{-1}	8.39×10^{-3}	174.0	200.5	
	793	3.77×10^{-1}	6.57×10^{-3}	174.1	201.6	808	2.67×10^{-1}	1.37×10^{-2}	173.8	200.3	
	800	5.10×10^{-1}	8.87×10^{-3}	173.9	201.3	818	3.83×10^{-1}	1.97×10^{-2}	173.6	200.1	
	808	6.58×10^{-1}	1.14×10^{-2}	173.8	201.5	828	5.30×10^{-1}	2.73×10^{-2}	173.5	200.2	
	813	8.42×10^{-1}	1.46×10^{-2}	173.7	201.0	Tf	743	5.26×10^{-2}	8.00×10^{-4}	174.9	202.5
	826	1.37	2.38×10^{-2}	173.5	200.7		751	6.86×10^{-2}	1.04×10^{-3}	174.8	203.0
	833	1.74	3.03×10^{-2}	173.4	200.6		758	9.19×10^{-2}	1.40×10^{-3}	174.7	202.9
	846	2.74	4.77×10^{-2}	173.2	200.4		763	1.05×10^{-1}	1.60×10^{-3}	174.6	203.3
	751	1.28×10^{-2}	1.31×10^{-3}	174.8	201.5		775	1.73×10^{-1}	2.62×10^{-3}	174.4	203.2
760	1.85×10^{-2}	1.90×10^{-3}	174.6	201.5	780		2.26×10^{-1}	3.43×10^{-3}	174.3	202.7	
765	2.27×10^{-2}	2.33×10^{-3}	174.5	201.4	785	2.52×10^{-1}	3.83×10^{-3}	174.2	203.2		
773	3.13×10^{-2}	3.21×10^{-3}	174.4	201.4	791	3.54×10^{-1}	5.38×10^{-3}	174.1	202.4		
787	5.39×10^{-2}	5.54×10^{-3}	174.2	201.3	794	3.85×10^{-1}	5.86×10^{-3}	174.0	202.6		
798	8.15×10^{-2}	8.38×10^{-3}	174.0	201.2	807	6.00×10^{-1}	9.11×10^{-3}	173.8	202.8		
803	9.80×10^{-2}	1.01×10^{-2}	173.9	201.2	813	7.64×10^{-1}	1.16×10^{-2}	173.7	202.6		
809	1.22×10^{-1}	1.25×10^{-2}	173.8	201.1	821	1.02	1.55×10^{-2}	173.6	202.5		
813	1.41×10^{-1}	1.45×10^{-2}	173.7	201.1	829	1.37	2.08×10^{-2}	173.5	202.3		
824	2.07×10^{-1}	2.13×10^{-2}	173.5	201.0	833	1.56	2.37×10^{-2}	173.4	202.3		
834	2.93×10^{-1}	3.01×10^{-2}	173.4	200.9	840	2.03	3.08×10^{-2}	173.3	202.1		
841	3.71×10^{-1}	3.81×10^{-2}	173.3	200.8							

Table 7. Vapor Pressures of FeCl₂ and FeBr₂ Measured by Using the Knudsen Assembly and Third-Law $\Delta_{\text{sub}}H^\circ(298\text{ K})$ (Estimated Error in T , ±2 K, in Δm ± 0.1 mg, and in t , ±5 min)

compound	T/K	$\Delta m/\text{mg}$	t/min	p/kPa	$-\Delta_{\text{sub}}[G^\circ(T) - H^\circ(298\text{ K})]/(\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1})$	$\Delta_{\text{sub}}H^\circ(298\text{ K})/(\text{kJ}\cdot\text{mol}^{-1})$	
FeCl ₂	745	20.6	1215	1.00×10^{-3}	174.9	201.7	
	735	14.7	1330	6.47×10^{-4}	175.0	201.8	
	721	6.9	1190	3.36×10^{-4}	175.3	202.0	
	713	5.3	1360	2.25×10^{-4}	175.4	202.2	
	703	6.3	2705	1.33×10^{-4}	175.6	202.4	
	701	2.5	1070	1.34×10^{-4}	175.6	202.0	
	694	6.0	3865	8.83×10^{-5}	175.7	202.5	
	697	9.1	5445	9.53×10^{-5}	175.7	202.7	
	FeBr ₂	745	50.0	623	1.81×10^{-3}	189.6	209.0
		736	37.6	645	1.31×10^{-3}	189.8	208.6
729		23.1	1115	9.27×10^{-4}	190.0	208.8	
721		19.2	1260	6.76×10^{-4}	190.1	208.5	
708		9.9	1230	3.56×10^{-4}	190.4	208.7	
693		11.2	2740	1.79×10^{-4}	190.7	208.5	

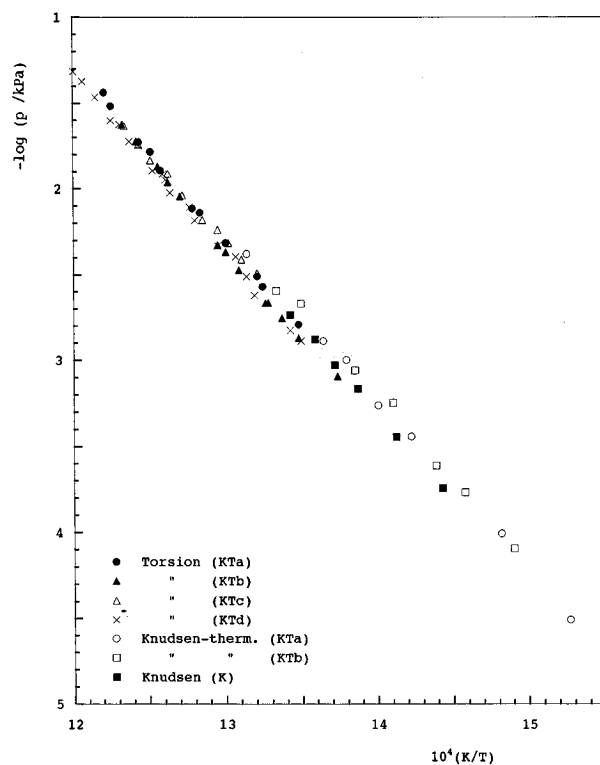
Table 8. Vapor Pressures of FeBr₂ Measured by Using the Torsion Method and Third-Law $\Delta_{\text{sub}}H^\circ(298 \text{ K})$

run	<i>T</i> /K	α /rad	<i>p</i> /kPa	$-\Delta_{\text{sub}}[G^\circ(T) - H^\circ(298 \text{ K})]/(J \cdot \text{mol}^{-1} \cdot \text{K}^{-1})$	$\Delta_{\text{sub}}H^\circ(298 \text{ K})/(kJ \cdot \text{mol}^{-1})$	run	<i>T</i> /K	α /rad	<i>p</i> /kPa	$-\Delta_{\text{sub}}[G^\circ(T) - H^\circ(298 \text{ K})]/(J \cdot \text{mol}^{-1} \cdot \text{K}^{-1})$	$\Delta_{\text{sub}}H^\circ(298 \text{ K})/(kJ \cdot \text{mol}^{-1})$
Ta	742	1.56×10^{-2}	1.60×10^{-3}	189.7	209.0	Tc	757	2.10×10^{-1}	3.20×10^{-3}	189.4	208.6
	755	2.59×10^{-2}	2.67×10^{-3}	189.5	209.2		763	2.53×10^{-1}	3.85×10^{-3}	189.3	209.0
	757	2.98×10^{-2}	3.06×10^{-3}	189.4	208.9		768	3.16×10^{-1}	4.80×10^{-3}	189.2	208.9
	769	4.67×10^{-2}	4.80×10^{-3}	189.2	209.1		772	3.76×10^{-1}	5.71×10^{-3}	189.1	208.8
	779	7.00×10^{-2}	7.20×10^{-3}	189.0	209.1		778	4.30×10^{-1}	6.53×10^{-3}	189.0	209.5
	782	7.39×10^{-2}	7.60×10^{-3}	188.9	209.5		786	5.93×10^{-1}	9.01×10^{-3}	188.9	209.4
	795	1.23×10^{-1}	1.27×10^{-2}	188.7	209.4		792	7.98×10^{-1}	1.21×10^{-2}	188.7	208.9
	799	1.58×10^{-1}	1.63×10^{-2}	188.6	208.7		799	9.55×10^{-1}	1.45×10^{-2}	188.6	209.5
	804	1.80×10^{-1}	1.85×10^{-2}	188.5	209.1		804	1.19	1.80×10^{-2}	188.5	209.3
	816	2.94×10^{-1}	3.02×10^{-2}	188.3	208.7		810	1.51	2.29×10^{-2}	188.4	209.1
	819	3.54×10^{-1}	3.64×10^{-2}	188.2	208.2	Td	741	2.50×10^{-2}	1.29×10^{-3}	189.7	210.1
Tb	728	7.79×10^{-3}	8.01×10^{-4}	190.0	209.4		745	2.88×10^{-2}	1.48×10^{-3}	189.6	210.2
	742	1.30×10^{-2}	1.33×10^{-3}	189.7	210.1		758	4.60×10^{-2}	2.37×10^{-3}	189.4	210.8
	748	1.70×10^{-2}	1.74×10^{-3}	189.6	210.0		761	5.94×10^{-2}	3.06×10^{-3}	189.3	209.9
	753	2.08×10^{-2}	2.14×10^{-3}	189.5	210.1		765	7.76×10^{-2}	4.00×10^{-3}	189.3	209.3
	754	2.07×10^{-2}	2.13×10^{-3}	189.5	210.4		772	8.97×10^{-2}	4.62×10^{-3}	189.1	210.2
	764	3.24×10^{-2}	3.33×10^{-3}	189.3	210.2		781	1.26×10^{-1}	6.47×10^{-3}	188.9	210.3
	769	4.15×10^{-2}	4.26×10^{-3}	189.2	209.9		783	1.50×10^{-1}	7.72×10^{-3}	188.9	209.6
	772	4.56×10^{-2}	4.69×10^{-3}	189.1	210.1		791	1.82×10^{-1}	9.35×10^{-3}	188.8	210.4
	787	8.68×10^{-2}	8.93×10^{-3}	188.8	209.7		793	2.16×10^{-1}	1.11×10^{-2}	188.7	209.8
	792	1.05×10^{-1}	1.08×10^{-2}	188.7	209.7		794	2.33×10^{-1}	1.20×10^{-2}	188.7	209.5
	796	1.30×10^{-1}	1.33×10^{-2}	188.7	209.3		798	2.45×10^{-1}	1.26×10^{-2}	188.6	210.2
	805	1.83×10^{-1}	1.88×10^{-2}	188.5	209.2		804	3.44×10^{-1}	1.77×10^{-2}	188.5	209.4
	811	2.28×10^{-1}	2.35×10^{-2}	188.4	209.2		808	3.62×10^{-1}	1.87×10^{-2}	188.4	210.0
							812	4.56×10^{-1}	2.35×10^{-2}	188.3	209.4
							816	4.82×10^{-1}	2.48×10^{-2}	188.3	210.0
							823	6.61×10^{-1}	3.40×10^{-2}	188.1	209.6
							829	8.19×10^{-1}	4.22×10^{-2}	188.0	209.5
							833	9.34×10^{-1}	4.81×10^{-2}	187.9	209.5

**Figure 2.** Vapor pressures of iron dichloride.

same considerations as used for FeCl₂. Following the same procedure as for the other halides, the second-law sublimation enthalpy of FeBr₂, $\Delta_{\text{sub}}H^\circ(745 \text{ K}) = (197 \pm 5) \text{ kJ} \cdot \text{mol}^{-1}$, was reduced to 298 K using Barin's enthalpic functions to give $\Delta_{\text{sub}}H^\circ(298 \text{ K}) = (208 \pm 5) \text{ kJ} \cdot \text{mol}^{-1}$.

The average third-law $\Delta_{\text{sub}}H^\circ(298 \text{ K}) = (209 \pm 1) \text{ kJ} \cdot \text{mol}^{-1}$, obtained from the values calculated using the experimental vapor pressures reported in Tables 3, 7, and

**Figure 3.** Vapor pressures of iron dibromide.

8 and, again, Barin's Gibbs energy functions, agrees well with our second-law value and with those calculated using literature pressures (see Table 5). This excellent agreement and the absence of substantial temperature trends in the third-law calculations lead us to propose as the standard sublimation enthalpy for FeBr₂ the value of $(208 \pm 2) \text{ kJ} \cdot \text{mol}^{-1}$.

Conclusion

Our best fitting equations of the total vapor pressures as a function of temperature for iron halides selected in

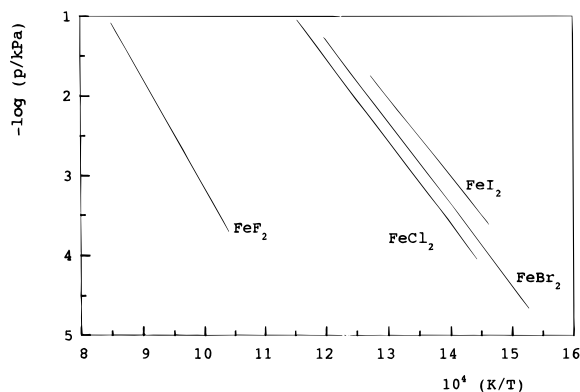


Figure 4. Vapor pressures of the iron halides.

the present work and of FeI_2 studied previously by Scarozza and Piacente (1995) are given for comparison in Figure 4. In contrast with FeI_2 , in which in addition to the monomer about 10% of the dimer was observed in the vapor, for other iron halides the monomers can be considered as the principal gaseous species in the vapor in the temperature ranges studied. This assumption was confirmed by the good agreement, within experimental uncertainties, of the vapor pressure values measured by both the torsion and Knudsen methods. Moreover, no marked dependence of vapor pressures of these compounds on the effusion hole area of the cells was observed, indicating that the accommodation coefficient may be taken as unity.

From the second- and third-law treatment of the vapor pressures of these compounds, their standard sublimation enthalpies were derived. The results confirm that generally the sublimation enthalpies of the difluorides are substantially greater than those of the other halides of the same metal.

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