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Heat Capacity and Thermodynamic Functions of the RFe$_2$ Compounds (R = Gd, Tb, Dy, Ho, Er, Tm, Lu) over the Temperature Region 8 to 300 K

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Experimental heat capacity data for the Laves phase $R\mathrm{Fe}_2$ intermetallic compounds (R = Gd, Tb, Dy, Ho, Er, Tm, and Lu) have been determined over the temperature range 8 to 300 K. The error in these data is thought to be less than 1%. Smoothed heat capacity values and the thermodynamic functions, $(H^\circ_T - H^\circ_0)$ and S°_T, are reported throughout the temperature range for the $R\mathrm{Fe}_2$ series. In addition, $(G^\circ_{298} - H^\circ_0)$ at 298 K is reported for all the $R\mathrm{Fe}_2$ compounds. These data were analyzed and it was shown that the maxima in the thermodynamic functions near HoFe$_2$ are due to the magnetic contribution of the lanthanide element. The lattice contribution to the entropy at 300 K was estimated, and from this quantity the Debye temperature was calculated to be about 300 K, which is in good agreement with the low-temperature heat capacity. Furthermore, this analysis indicates that the apparent electronic specific heat constants, γ', for TbFe$_2$, DyFe$_2$, and HoFe$_2$, reported earlier, are in error.

Introduction

Heat capacity studies of the $R\mathrm{Fe}_2$ compounds have been undertaken in order to provide essential thermal data which have been lacking. These data can provide some insight into the stability of the $R\mathrm{Fe}_2$ compounds and have led to the characterization of the crystal field interaction in several of these compounds (*1*).

This paper reports the heat capacity data for the Laves phase $R\mathrm{Fe}_2$ compounds: GdFe$_2$, TbFe$_2$, DyFe$_2$, HoFe$_2$, ErFe$_2$, TmFe$_2$, and LuFe$_2$. The smoothed heat

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TABLE I

SMOOTH THERMODYNAMIC FUNCTIONS FOR $GdFe_2$

T (K)	C_p (J/mole-K)	$H^\circ_T - H^\circ_0$ (J/mole)	S°_T (J/mole-K)
10.00	0.430	1.61	0.28
20.00	2.593	14.26	1.07
30.00	7.960	65.67	3.08
40.00	14.980	179.17	6.30
50.00	22.600	367.01	10.46
60.00	30.350	631.99	15.27
70.00	37.410	971.34	20.49
80.00	43.700	1377.55	25.90
90.00	49.160	1842.26	31.37
100.00	53.890	2358.26	36.80
110.00	57.850	2917.48	42.13
120.00	61.410	3514.07	47.32
130.00	64.560	4144.56	52.36
140.00	67.080	4803.13	57.24
150.00	69.280	5484.96	61.95
160.00	71.160	6187.31	66.48
170.00	72.820	6907.52	70.84
180.00	74.410	7643.71	75.05
190.00	75.910	8395.39	79.11
200.00	77.400	9162.14	83.05
210.00	78.810	9943.27	86.86
220.00	80.110	10738.00	90.56
230.00	81.200	11544.61	94.14
240.00	81.940	12360.47	97.61
250.00	82.650	13183.33	100.97
260.00	83.520	14014.03	104.23
270.00	84.520	14854.15	107.40
280.00	85.540	15704.51	110.49
290.00	86.550	16564.95	113.51
300.00	87.640	17435.74	116.46

capacity data along with the thermodynamic functions ($H^\circ_T - H^\circ_0$) and S°_T are reported over the temperature range 8 to 300 K for each compound. In addition, the values of S°_{298}, ($H^\circ_{298} - H^\circ_0$), and ($G^\circ_{298} - H^\circ_0$) have been tabulated for the series. This work is part of an ongoing program designed to characterize the thermal properties of the RFe_2 series and their corresponding hydrides (*1*, *2*).

Experimental

The RFe_2 samples (R = Gd, Tb, Dy, Ho, Er, and Tm) were prepared at the University of Pittsburgh by the cold-boat induction heating technique (*3*). Spectrographic analysis supplied by the manufacturer indicated that the rare earth metals had a purity of 99.9 wt% with respect to metallic impurities only. The iron was Johnson and Matthey Chemicals, Ltd. Puratronic grade 1 rod. The $LuFe_2$ compound was prepared at the Ames Laboratory-DOE by arc melting on a water-cooled copper hearth. The Lu metal was 99.99 at.% pure with respect to metallic impurities (H, O, C, and N concentrations were 693, 403, 233, and 25 ppma, respectively). The Fe, which was obtained

TABLE II

SMOOTH THERMODYNAMIC FUNCTIONS FOR $TbFe_2$

T (K)	C_p (J/mole-K)	$H^\circ_T - H^\circ_0$ (J/mole)	S°_T (J/mole-K)
10.00	0.541	1.71	0.28
20.00	2.820	16.72	1.24
30.00	8.040	68.09	3.25
40.00	15.560	184.43	6.54
50.00	23.860	381.09	10.90
60.00	32.000	660.67	15.97
70.00	39.670	1019.32	21.49
80.00	46.600	1451.28	27.24
90.00	52.740	1948.75	33.10
100.00	57.740	2502.24	38.92
110.00	61.730	3100.31	44.62
120.00	65.250	3735.67	50.15
130.00	68.290	4403.70	55.49
140.00	70.910	5100.07	60.65
150.00	73.140	5820.57	65.62
160.00	74.990	6561.37	70.40
170.00	76.640	7319.62	75.00
180.00	78.280	8094.30	79.42
190.00	79.740	8884.46	83.70
200.00	81.050	9688.49	87.82
210.00	82.350	10505.54	91.81
220.00	83.510	11335.11	95.67
230.00	84.310	12174.53	99.40
240.00	85.030	13021.23	103.00
250.00	85.760	13875.25	106.49
260.00	86.410	14736.08	109.86
270.00	87.090	15603.43	113.14
280.00	88.120	16478.94	116.32
290.00	89.130	17365.60	119.43
300.00	89.690	18260.53	122.46

TABLE III

SMOOTH THERMODYNAMIC FUNCTIONS FOR DyFe_2

T (K)	C_p (J/mole-K)	$H^\circ_T - H^\circ_0$ (J/mole)	S°_T (J/mole-K)
10.00	0.501	1.64	0.27
20.00	2.832	15.84	1.16
30.00	8.192	68.19	3.22
40.00	15.880	187.38	6.59
50.00	24.620	389.57	11.07
60.00	33.230	678.70	16.31
70.00	41.210	1051.33	22.04
80.00	48.380	1500.13	28.02
90.00	54.770	2016.58	34.10
100.00	59.950	2591.56	40.15
110.00	64.020	3212.01	46.06
120.00	67.600	3870.53	51.79
130.00	70.630	4562.44	57.33
140.00	73.030	5281.02	62.65
150.00	75.130	6022.10	67.76
160.00	76.820	6782.19	72.67
170.00	78.430	7558.54	77.38
180.00	79.850	8350.18	81.90
190.00	81.150	9155.30	86.25
200.00	82.320	9972.76	90.45
210.00	83.530	10802.11	94.49
220.00	84.570	11642.80	98.40
230.00	85.340	12492.71	102.18
240.00	85.920	13349.01	105.82
250.00	86.530	14211.14	109.34
260.00	87.190	15079.72	112.75
270.00	88.130	15955.97	116.06
280.00	89.160	16842.68	119.28
290.00	89.520	17736.62	122.42
300.00	89.530	18632.05	125.45

from Glidden Iron, Inc., was 99.99 at.% pure with respect to metallic impurities (O, H, C, and N concentrations were 1115, 445, 37, and 24 ppma, respectively). Further details pertaining to the preparation of these samples have been described previously by Germano *et al.* (*1*).

The adiabatic calorimeter used in this study has been described elsewhere (*1*, *4*). Due to the reduced precision of the adiabatic calorimeter in the temperature region 8 to 15 K, the heat capacity data for the $R\text{Fe}_2$ samples in this region were verified by specific heat measurements performed on a pulse calorimeter which is accurate to within 1% of the NBS copper data in this temperature range. Furthermore, based on our results for benzoic acid and copper in the temperature range 15 to 300 K, which agree within 1% of the NBS data, we believe our results on the $R\text{Fe}_2$ phases are accurate within 1% over the temperature range 8 to 300 K.

It had been reported in an earlier paper (*1*) that the heat capacity data had been truncated at 285 K due to experimental scatter above 285 K. Replacement of the platinum resistance thermometer with another, calibrated in house, removed most of

TABLE IV

SMOOTH THERMODYNAMIC FUNCTIONS FOR HoFe_2

T (K)	C_p (J/mole-K)	$H^\circ_T - H^\circ_0$ (J/mole)	S°_T (J/mole-K)
10.00	0.622	1.82	0.28
20.00	4.075	21.69	1.53
30.00	10.975	95.56	4.43
40.00	18.920	244.56	8.66
50.00	27.150	474.81	13.76
60.00	35.320	787.43	19.44
70.00	42.680	1178.30	25.45
80.00	48.920	1636.89	31.57
90.00	54.350	2153.75	37.65
100.00	59.020	2721.23	43.62
110.00	63.100	3332.30	49.44
120.00	66.590	3981.29	55.09
130.00	69.410	4661.87	60.53
140.00	71.800	5368.27	65.77
150.00	73.810	6096.54	70.79
160.00	75.650	6844.03	75.62
170.00	77.300	7609.04	80.25
180.00	78.790	8389.53	84.71
190.00	80.050	9183.92	89.01
200.00	81.280	9990.46	93.15
210.00	82.510	10809.49	97.14
220.00	83.700	11640.69	101.01
230.00	84.570	12482.24	104.75
240.00	85.220	13331.43	108.36
250.00	85.820	14186.55	111.85
260.00	86.730	15049.22	115.24
270.00	87.900	15922.36	118.53
280.00	89.190	16807.69	121.75
290.00	90.290	17705.18	124.90
300.00	91.360	18613.29	127.98

the scatter above 285 K and we now feel justified in reporting our data to 300 K.

Results and Discussion

Tables I through VII[1] contain the smoothed values of the heat capacity and thermodynamic functions, ($H_T^\circ - H_0^\circ$) and S_T°, for the seven RFe$_2$ compounds over the temperature range 8 to 300 K. The enthalpy and entropy at each temperature were obtained by Simpson's-rule integration under the smoothed data curve of interest; C_p vs T for enthalpy, and C_p/T vs T for entropy.

TABLE V

SMOOTH THERMODYNAMIC FUNCTIONS FOR ErFe$_2$

T (K)	C_p (J/mole-K)	$H_T^\circ - H_0^\circ$ (J/mole)	S_T° (J/mole-K)
10.00	0.310	1.20	0.21
20.00	2.708	12.90	0.93
30.00	9.770	72.02	3.23
40.00	18.980	215.18	7.29
50.00	28.850	454.59	12.59
60.00	38.160	790.62	18.69
70.00	45.900	1212.31	25.17
80.00	52.080	1703.39	31.72
90.00	57.120	2250.06	38.15
100.00	61.400	2843.28	44.40
110.00	64.890	3475.50	50.42
120.00	67.490	4137.98	56.19
130.00	69.660	4823.87	61.68
140.00	71.510	5529.94	66.91
150.00	73.200	6253.65	71.90
160.00	74.600	6992.81	76.67
170.00	75.830	7745.02	81.23
180.00	77.000	8509.31	85.60
190.00	78.050	9284.48	89.79
200.00	79.100	10070.26	93.82
210.00	80.100	10866.25	97.70
220.00	81.100	11672.25	101.45
230.00	81.880	12487.16	105.07
240.00	82.580	13309.43	108.57
250.00	83.150	14138.11	111.96
260.00	83.700	14972.16	115.23
270.00	84.490	15812.68	118.40
280.00	85.400	16662.39	121.49
290.00	85.710	17518.37	124.49
300.00	85.810	18375.96	127.40

TABLE VI

SMOOTH THERMODYNAMIC FUNCTIONS FOR TmFe$_2$

T (K)	C_p (J/mole-K)	$H_T^\circ - H_0^\circ$ (J/mole)	S_T° (J/mole-K)
10.00	0.983	2.12	0.31
20.00	5.877	34.02	2.33
30.00	13.240	128.73	6.06
40.00	21.640	302.93	11.02
50.00	30.490	563.61	16.80
60.00	36.860	902.18	22.95
70.00	43.930	1307.20	29.18
80.00	49.890	1776.79	35.45
90.00	54.990	2302.19	41.63
100.00	59.110	2873.40	47.64
110.00	62.420	3481.66	53.44
120.00	65.190	4120.08	58.99
130.00	67.490	4783.85	64.30
140.00	69.450	5468.87	69.38
150.00	71.170	6172.01	74.23
160.00	72.680	6891.46	78.87
170.00	73.970	7624.89	83.32
180.00	75.070	8370.10	87.58
190.00	76.020	9125.61	91.66
200.00	76.920	9890.36	95.58
210.00	77.820	10664.04	99.36
220.00	78.730	11446.79	103.00
230.00	79.450	12238.00	106.52
240.00	80.030	13035.43	109.91
250.00	80.560	13838.38	113.19
260.00	81.110	14646.70	116.36
270.00	81.800	15461.20	119.43
280.00	82.560	16283.03	122.42
290.00	83.310	17112.21	125.33
300.00	84.230	17949.88	128.17

For the GdFe$_2$, ErFe$_2$, and LuFe$_2$ compounds, the heat capacity data in the region $T \leq 8$ K were fit to $C_p = \gamma T + \beta T^3$, using the values of γ and β reported by Butera *et al.* (*2*). For the remaining compounds, the C_p data in this region were fit using the interpolated γ values reported in this paper.

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[1] See NAPS document No. 03777 for 14 pages of supplementary material. Order from ASIS/NAPS c/o Microfiche Publications, P.O. Box 3513, Grand Central Station, New York, N.Y. 10163. Remit in advance, in U.S. funds only, for each NAPS Accession Number. Institutions and organizations may use purchase orders when ordering, however, there is a billing charge for this service. Make checks payable to Microfiche Publications. Photocopies are $5.00. Microfiche are $3.00. Outside of the U.S. and Canada, postage is $3.00 for a photocopy or $1.50 for a fiche.

TABLE VII

SMOOTH THERMODYNAMIC FUNCTIONS FOR LuFe_2

T (K)	C_p (J/mole-K)	$H^\circ_T - H^\circ_0$ (J/mole)	S°_T (J/mole-K)
10.00	0.355	1.20	0.20
20.00	2.487	13.05	0.94
30.00	6.990	58.81	2.73
40.00	13.200	158.36	5.55
50.00	20.435	326.14	9.27
60.00	27.620	566.67	13.63
70.00	34.199	876.74	18.40
80.00	39.999	1247.95	23.35
90.00	45.000	1673.63	28.36
100.00	49.300	2145.83	33.33
110.00	53.100	2658.12	38.21
120.00	56.400	3206.02	42.97
130.00	59.070	3783.56	47.59
140.00	61.300	4385.66	52.05
150.00	63.200	5008.36	56.35
160.00	65.000	5649.44	60.49
170.00	66.630	6307.63	64.48
180.00	68.180	6981.81	68.33
190.00	69.600	7670.76	72.05
200.00	70.780	8372.60	75.65
210.00	71.920	9086.08	79.14
220.00	73.070	9811.05	82.51
230.00	74.120	10547.01	85.78
240.00	75.140	11293.24	88.95
250.00	76.250	12050.13	92.04
260.00	77.350	12818.16	95.06
270.00	78.500	13597.35	98.00
280.00	79.650	14387.90	100.87
290.00	80.820	15190.90	103.69
300.00	81.810	16004.05	106.45

Table VIII contains the tabulated values of $(H^\circ_{298} - H^\circ_0)$, S°_{298}, and $(G^\circ_{298} - H^\circ_0)$ for the $R\mathrm{Fe}_2$ compounds. These values seem to follow a trend in which the maximum value in each occurs at or neat the HoFe_2 compound. A similar trend was noted by Butera *et al.* (*2*) in the electronic contribution to the heat capacity in the temperature range 1.5 to 10 K.

A closer examination, however, shows that the maxima in the thermodynamic functions at holmium are to be expected because of a magnetic contribution from the rare earth metal, as discussed below. Furthermore, this analysis also indicates that the apparent maximum in the electronic contribution is probably due to impurities in the $R\mathrm{Fe}_2$ compounds with R = Tb, Dy, and Ho.

At room temperature (300 K) the total measured entropy, S_T, is given by

$$S_T = S_L + S_E + S^{\mathrm{RE}}_{\mathrm{M}} + S^{\mathrm{Fe}}_{\mathrm{M}}, \qquad (1)$$

where S_L is the lattice contribution, S_E is the electronic contribution, and $S^{\mathrm{RE}}_{\mathrm{M}}$ and $S^{\mathrm{Fe}}_{\mathrm{M}}$ are the respective magnetic contributions from the rare earth and iron atoms to the entropy. S_L is assumed to be given by the Debye theory. S_E is the entropy calculated from the electronic specific heat constant, γ, assuming γ remains constant up to 300 K. The $S^{\mathrm{RE}}_{\mathrm{M}}$ term is given by

$$S^{\mathrm{RE}}_{\mathrm{M}} = R\ln(2J + 1), \qquad (2)$$

where R is the gas constant and J is the total quantum number for the particular magnetic lanthanide metal. But at 300 K the $S^{\mathrm{RE}}_{\mathrm{M}}$ value does not reach the full theoretical value because of the splitting of the J manifold by the crystal field such that at 300 K all states are not occupied. The value of $S^{\mathrm{RE}}_{\mathrm{M}}$ at 300 K is determined from the excess heat capacity after the heat capacity of the LuFe_2 compound is subtracted from the magnetic $R\mathrm{Fe}_2$ phase as outlined by Germano *et al.* (*1*). The full magnetic contribution to the entropy from the iron atom, $S^{\mathrm{theor}}_{\mathrm{Fe}}$, is given by an expression similar to Eq. (2), except J is replaced by the spin quantum number, assumed to be $\frac{3}{2}$ (the

TABLE VIII

Compound	S°_{298} (J/mole-K)	$(H^\circ_{298} - H^\circ_0)$ (J/mole)	$(G^\circ_{298} - H^\circ_0)$ (J/mole)
GdFe_2	115.88	17261	−17272
TbFe_2	121.81	18081	−18242
DyFe_2	124.86	18453	−18776
HoFe_2	127.37	18431	−19561
ErFe_2	126.83	18204	−19591
TmFe_2	127.62	17782	−20249
LuFe_2	105.90	15841	−15718

value for Fe^{3+}). But the full entropy will not be realized until the compound becomes paramagnetic, i.e., above the Curie temperature. Because the ordering in these compounds is due essentially to the iron–iron interactions (*5*), there will be little or no contribution to the entropy at 300 K, since the Curie temperatures, which vary from 565 K for $TmFe_2$ to 800 K for $GdFe_2$ (580 K for $LuFe_2$) (*5*), are far above room temperature. We have estimated S_M^{FE} at 300 K to be given by

$$S_M^{Fe} = (0.1)\,(2)\left(\frac{300}{T_c}\right) S_{Fe}^{theor}, \qquad (3)$$

where T_c is the Curie temperature. The term $300/T_c$ is a rough scaling factor which takes into account the variation of the ordering temperature of the RFe_2 phases. The quantity 2 is required since there are two iron atoms per formula unit and the factor 0.1 is introduced because most of the magnetic entropy is associated with the actual ordering near T_c.

We now use Eq. (1) to calculate S_L from the other quantities. The value of S_E was first calculated from the experimental values reported for the RFe_2 by Butera *et al.* (*2*) but the resultant S_L values, see Table IX, were found to exhibit an overall decrease from $GdFe_2$ to $LuFe_2$. The S_L values did not vary smoothly, but instead showed a sharp dip at $HoFe_2$. This dip is due to the anomalously high apparent electronic specific heat constant, γ', for $HoFe_2$ and also $TbFe_2$ and $DyFe_2$. Band structure calculations for the RFe_2 phases by Cyrot *et al.* (*6*) indicate that the density of states at the Fermi level varies little when one rare earth metal replaces another one. As seen from Fig. 3 in Butera *et al.* (*2*), the γ' values for $GdFe_2$, $ErFe_2$, and $LuFe_2$ lie on a straight line, and are consistent with the calculations of Cyrot *et al.* Thus it is likely that the anomalously high values for $TbFe_2$, $DyFe_2$, and $HoFe_2$ are due to impurities in these samples. It is possible that the γ' values for $GdFe_2$ and $TbFe_2$ are also high because of impurities, but the $LuFe_2$ sample is less likely to be anomalous because it was prepared from much purer starting lanthanide materials than the other RFe_2 compounds. Because of this possibility, we have calculated S_L assuming the electronic specific heat constant remains unchanged for all the RFe_2 phases and is equal to that of $LuFe_2$. Calculations have also been made assuming that the electronic specific constant varies smoothly in a linear fashion from $GdFe_2$ to $LuFe_2$. The results of these calculations are given in Table IX, and it is seen that S_L varies smoothly with no anomalies between $GdFe_2$ and $LuFe_2$ in either case. This behavior is as expected, since the Debye temperature should vary slowly across this series of lanthanide compounds and not show any maxima or minima. The Debye temperatures were calculated from the S_L values using the tables of Kilpatrick and Sherman (*7*) and these are listed in

TABLE IX

LATTICE CONTRIBUTION TO THE ENTROPY, S_L, AT 300 K (e.u./mole)

Compound	Using experimental γ' value[a]	Assuming γ is constant[b]	Assuming γ varies smoothly[c]
$GdFe_2$	102.00	103.94	102.00
$TbFe_2$	101.03	103.48	101.79
$DyFe_2$	98.91	102.73	101.37
$HoFe_2$	97.03	102.22	101.19
$ErFe_2$	101.03	101.78	101.03
$TmFe_2$	101.11	101.47	101.11
$LuFe_2$	100.55	100.55	100.55

[a] Apparent electronic specific heat constant as measured by Butera *et al.* (*2*).

[b] Electronic specific heat constant is the same as that of $LuFe_2$ for all RFe_2.

[c] Electronic specific heat constant varies smoothly from the reported values for $GdFe_2$, $ErFe_2$, and $LuFe_2$, which lie on a straight line in a plot of γ vs atomic number of the lanthanide. The other values were estimated from this plot.

TABLE X

DEBYE TEMPERATURES

Compound	High-temperature values calculated from S_L: Constant γ^a	High-temperature values calculated from S_L: Varying γ^a	Low-temperature (1–10 K) measured values
GdFe$_2$	290	298	270
TbFe$_2$	292	299	280
DyFe$_2$	295	301	284
HoFe$_2$	297	302	284
ErFe$_2$	299	302	300
TmFe$_2$	301	302	—
LuFe$_2$	304	304	296

[a] See text or footnotes b and c of Table IX.

Table X along with the experimental values determined from low-temperature heat capacity measurements (*2*). The agreement between either set of Debye temperatures calculated from the S_L values and the experimental Θ_D values determined below 10 K is good. Exact agreement is not expected since the Θ_D values calculated from S_L correspond to a high-temperature value which, in general, differ by as much as 10% from values determined near 0 K. It is important to note that the Θ_D values calculated from S_L do vary smoothly from GdFe$_2$ to LuFe$_2$ and do change in the same direction as the low-temperature values.

The method used for calculating S_M^{Fe} may be questioned. If in error, the value obtained for S_M^{Fe} is probably too large, but since S_M^{Fe} is only 1 to 1.5% of S_L, it does not significantly affect the values obtained for S_L and Θ_D. If one assumes $S_M^{Fe} = 0$ for all RFe$_2$ compounds, the resultant Θ_D values are 4 to 7 K smaller than those given in Table X and they still vary in a smooth manner, increasing from GdFe$_2$ to LuFe$_2$.

Thus from the above analysis it is seen that the maximum in S_T near HoFe$_2$ is expected and primarily is due to the S_M^{RE} contribution. Furthermore, the above analysis indicates that the large values of γ' of TbFe$_2$, DyFe$_2$, and HoFe$_2$ determined by low-temperature heat capacity measurements are not the true electronic specific heat constant values, but the values for GdFe$_2$, ErFe$_2$, and LuFe$_2$ probably are. It is interesting that the Debye temperatures at 0 K apparently are not affected by the impurities which cause the anomalous γ' values.

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