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UC-32 - Mathematics and Computers

ØNE PHASE: A SIMPLIFIED FORTRAN 63
PROGRAM FOR COMPUTING TABLES OF
THERMODYNAMIC DATA FOR
COMPOUNDS IN ONE PHASE

M. H. Fontana

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SEPTEMBER 1968

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ØNEPHASE: A FORTRAN-63 PROGRAM FOR COMPUTING TABLES OF THERMODYNAMIC
DATA FOR COMPOUNDS IN ONE PHASE

M. H. Fontana

Abstract

A simplified Fortran program was written to compute thermodynamic properties using specific heat equations, and the entropy and the heat of formation at some known temperature. The program computes and prints tables of the following properties: specific heat, enthalpy, entropy, heat of formation, free energy of formation, and the equilibrium constant of formation. Cards may be punched directly for use by computer programs requiring data input in this format.

INTRODUCTION

In the course of performing multicomponent equilibrium calculations on fission product fuel mixtures it became apparent that much of the required thermodynamic data on the compounds of interest were not available.

Other programs were written to compute data for monatomic gaseous elements (MØNAT)¹ and for compounds and elements in any state (TCDATA)², but it soon proved advantageous to have a simple program that was easy to use for cases where data were desired in a temperature range where no phase changes occur. Also, for many uses, tables of data for one phase are required in which the data are extrapolated past the normal range of stability of that phase. Program ØNEPHASE was designed to fill this need.

The program can print tables in the JANAF³ format, i.e., the specific heat, enthalpy, entropy, heat of formation, free energy of formation, and the natural logarithm of the equilibrium constant of formation are presented for any desired temperature. The program can also punch cards for direct use by computer programs requiring data input in this format.

DESCRIPTION OF COMPUTATIONS PERFORMED BY THE PROGRAM

The program computes thermodynamic functions given an equation for the specific heat; the enthalpy for the reactants, referenced to 298⁰K; the heat of formation at some known temperature; the entropy at one temperature; the enthalpy of the compound at one temperature, referenced to 298⁰K; a temperature which is the upper limit of applicability of the specific heat equation used; the temperature at which the properties are

required; and the maximum and minimum temperature to be considered.

Specific Heat

The specific heat is computed by an equation of the form:

$$C_p = A + BT + \frac{C}{T^2} + DT^2 \quad (1)$$

within the range of applicability of the equation; and by

$$C_p = C_{p(\text{med})} + E \left[\frac{T - T_{\text{med}}}{T_{\text{max}} - T_{\text{med}}} \right] \quad (2)$$

where A, B, C, D, and E are constants, $C_{p(\text{med})}$ is the specific heat at the highest temperature for which Eq. (1) holds, T_{med} is the highest temperature for which Eq. (1) holds, and T_{max} is the maximum temperature desired, at temperatures above the range of Eq. (1).

Equation (1) can be made to conform to either of two widely used equations by setting the constants, C or D, equal to zero.

The rationale for using Eqs. (1) and (2) in conjunction with each other is that, for purposes of estimation when experimental data is unavailable, the specific heat of a compound just before the first phase change can be estimated to be approximately 7.25 cal/(°K g-atom) for all elements; it may be about 6.2 cal/(°K g-atom) for inorganic compounds and 7.25 cal/(°K-atom) for intermetallic phases (see Kubaschewski and Evans⁴, p. 183). (These values are multiplied by the number of atoms in the molecule to give the molar heat capacity of the substance.) Estimation procedures are necessary for almost all of the compounds which we dealt with here, so it seemed reasonable to estimate the specific heat relationship as a linear function of temperature from the highest known value to the first known or estimated phase transition.

Enthalpy

The enthalpy is usually required based on a zero value at 298°K. Thus, if a value is given of $H_T - H_{298}$ at a minimum temperature of interest, it is necessary only to compute the enthalpy increment at temperatures higher than this. Thus,

$$H_T - H_{298} = H_{T \text{ min}} - H_{298} + \Delta H_{T \rightarrow (T + \Delta T)} \quad (3)$$

where

$$\Delta H_{(T \rightarrow T + \Delta T)} = C_p \Delta T. \quad (4)$$

Thus, over a large temperature difference

$$H_T - H_{298} = H_{T \text{ min}} - H_{298} + \sum_{\Delta T_i} C_p \Delta T \quad (5)$$

Essentially, the specific heat is computed at the temperature of interest, and the enthalpy change is computed assuming a constant specific heat over the increment of temperature between desired temperatures. Since the normal requirement is to print a table of properties at 100°K temperature increments, this procedure is adequate for our estimation purposes.

Entropy

The entropy is also computed in temperature increments, by first computing the specific heat and assuming that it remains constant over the temperature increment.

$$S_{(T + \Delta T)} - S_{(T)} = C_p \int_T^{T + \Delta T} \frac{dT}{T} = C_p \ln \left(\frac{T + \Delta T}{T} \right) \quad (6)$$

Since the entropy at some temperature is known, the entropy is built up incrementally by Eq. (6).

Enthalpy of Formation

The enthalpy of formation of a compound, ΔH_f° , is computed at temperatures other than the known temperature by use of Kirchoff's Law:

$$\Delta H_{fT}^{\circ} = \Delta H_{fT_{\min}}^{\circ} + \sum_i v_i (H - H_{T_{\min}})_i \quad (7)$$

where the stoichiometric equation is

$$\sum v_i A_i = 0. \quad (8)$$

For example, for the compound, M, formed when a atoms of element A, b atoms of element B, and c atoms of element C, the stoichiometric equation (written so that one molecule of M is formed) is:

$$M - aA - bB - cC = 0, \quad (9)$$

In this case, the enthalpy of formation is

$$\begin{aligned} \Delta H_{fT}^{\circ} = \Delta H_{fT_{\min}}^{\circ} &+ (H_T - H_{T_{\min}})_M - (H_T - H_{T_{\min}})_A - (H_T - H_{T_{\min}})_B \\ &- (H_T - H_{T_{\min}})_C \end{aligned} \quad (10)$$

Since, for computer purposes, $H_T - H_{T_{\min}}$ for the constituent elements must be read in, it is convenient to use the same deck of input cards when computing properties of compounds which contain the same elements, but in different proportions, and possibly with different values of T_{\min} . For this reason Eq. (10) was modified so that values of $H_T - H_{298}$ for the constituent elements could be used regardless of the value of T_{\min} . This was done by adding three more terms to Eq. (10) to obtain:

$$\begin{aligned} \Delta H_{fT}^{\circ} = \Delta H_{fT_{\min}}^{\circ} &+ (H_T - H_{298})_M - (H_{T_{\min}} - H_{298})_M - (H_T - H_{298})_A \\ &- (H_T - H_{298})_B - (H_T - H_{298})_C + (H_{T_{\min}} - H_{298})_A + (H_{T_{\min}} - H_{298})_B \end{aligned}$$

$$+ (H_{T \text{ min}} - H_{298})_C \quad (11)$$

Generalizing,

$$\begin{aligned} \Delta H_{f_T}^{\circ} &= \Delta H_{f_{T \text{ min}}}^{\circ} + (H_T - H_{298})_M - (H_{T \text{ min}} - H_{298})_M \\ &- \sum_{\text{reactants}} (H_T - H_{298}) + \sum_{\text{reactants}} (H_{T \text{ min}} - H_{298}) \end{aligned} \quad (12)$$

Free Energy of Formation

The free energy of formation, ΔF_f° , is computed from its definition

$$\Delta F_f^{\circ} = \Delta H_f^{\circ} - T\Delta S_f^{\circ} \quad (13)$$

where ΔH_f° is the enthalpy of formation and ΔS_f° is the entropy of formation.

Equilibrium Constant of Formation

The equilibrium constant of formation of the compound in the state of interest as formed from its constituent elements in their standard states is computed from:

$$\ln K_p = - \frac{\Delta F_f^{\circ}}{RT} \quad (14)$$

DESCRIPTION OF THE PROGRAM AND ITS USE

As explained in previous sections, program `ONEPHASE` computes the thermodynamic properties for a compound as formed from its elements in their standard state, given some base point data and a specific heat equation.

As presently written, the program prints tables of data for temperatures desired, and punches cards with an identification number, temperature, specific heat in calories/mole-^oK, $H - H_{298}$ in

kilocalories/gram-mole, entropy in cal/mole-^oK, and natural logarithms of the equilibrium constant of formation. This was chosen to be compatible with the JANAF³ tables.

Caution: Although some output values are in kilocalories, all input values must be in calorie units.

Input Required

The first card contains the values of CA, CB, CC, CD, and CE in a (5E15.7) format.

The second card contains the values of A, B, C, HMOR, and NI in the format (4F10.0, I5). Notice that the decimal point must be included (wherever desired) in the first four numbers.

The third card contains the values of HMIN, SMIN, DHMIN, TMED, CMED, TMAX, and TMIN in a (7F10.0) format. The units of HMIN and DHMIN must be in calories per gram-mole (not kilocalories).

The fourth and succeeding cards contain T, HA, HB, HC, DUM, SA, SB, SC, and DUM in the (9F8.0) format. The variable DUM is used so that the same input cards that are used in program TCDATA² which can handle four constituent elements, can be used with ONEPHASE which handles three constituent elements.

One card is needed for each temperature. This is not as bad as it sounds in that one deck of input cards once made (to be used as the fourth and succeeding cards) can be used as input for any compound made up of the elements carried on the cards. This is done by properly manipulating the values of a, b, and c in the stoichiometric equation, Eq. (9).

DICTIONARY OF COMPUTER TERMS*

CA = constant, A, in the specific heat equation (1) (cal/mole-°K)

CB = constant, B, in Eq. (1) (cal/mole-°K²)

CC = constant, B, in Eq. (1) (cal-°K/moles)

CD = constant, D, in Eq. (10) (cal/mole-°K³)

CE = constant, E, in Eq. (2) (cal/mole-°K)

A = stoichiometric constant, a, in Eq. (9) (dimensionless)

B = stoichiometric constant, b, in Eq. (9) (dimensionless)

C = stoichiometric constant, c, in Eq. (9) (dimensionless)

HMR = the sum of the enthalpies of the reactants in going from 298°K to the minimum temperature of interest; the last term in Eq. (12) (cal/mole)

NI = identification number of first card in a deck if the program is to punch cards which are to be numbered (dimensionless)

HMIN = $H - H_{298}$ of the compound at the minimum temperature (cal/mole)

SMIN = entropy of the compound at the minimum temperature (cal/mole-°K)

DHMIN = enthalpy of formation of the compound at the minimum temperature (cal/mole-°K)

TMED = T_{med} in Eq. (2), the highest temperature for which the specific heat Eq. (1) holds (°K)

CMED = C_p med in Eq. (2), the value of the specific heat at the highest temperature for which Eq. (1) holds (cal/mole-°K)

TMAX = T_{max} in Eq. (2), the maximum temperature for which data is to be computed (°K)

TMIN = the lowest temperature for which data is to be computed (°K)

*In order of appearance.

T = temperature ($^{\circ}$ K)

HA = $H - H_{298}$ for element A (cal/mole)

HB = $H - H_{298}$ for element B (cal/mole)

HC = $H - H_{298}$ for element C (cal/mole)

DUM = dummy variable

SA = entropy of element A (cal/mole- $^{\circ}$ K)

SB = entropy of element B (cal/mole- $^{\circ}$ K)

SC = entropy of element C (cal/mole- $^{\circ}$ K)

CP = specific heat (cal/mole- $^{\circ}$ K)

DT = Δt , temperature increment of tabulated values ($^{\circ}$ K)

TSTOR = a storage location for the previous value of T.

HMMS = a storage location for the value of the entropy at the previous value of T

SST = a storage location for the value of the entropy at the previous value of T

HMM = $H - H_{T \text{ min}}$ at the temperature of interest (cal/mole)

S = entropy (cal/mole- $^{\circ}$ K)

DH = Δh , enthalpy increment (cal/mole)

DS = entropy increment (cal/mole- $^{\circ}$ K)

HMTM = $H - H_{T \text{ min}}$, enthalpy of the compound above T_{min} (cal/mole)

DELH = enthalpy of formation at desired temperature (cal/mole)

DELS = entropy of formation at desired temperature (cal/mole- $^{\circ}$ K)

DELF = Gibbs free energy of formation at desired temperature (cal/mole- $^{\circ}$ K)

ALNKP = $\ln K_p$, natural logarithm of the equilibrium constant of formation
(dimensionless)

DELHKC = enthalpy of formation in kilocalories (kcal/mole)

HMMKC = enthalpy ($H - H_{T \text{ min}}$) in kilocalories (kcal/mole)

DELFKC = Gibbs free energy of formation in kilocalories (kcal/mole)

LISTING OF COMPUTER PROGRAM ONEPHASE

```
PROGRAM ONEPHASE
C THE FOLLOWING COMPUTES DATA FOR COMPOUNDS FROM CP EQUATION,
C HEATS OF FORMATION, AND DATA FOR ELEMENTS.
  READ 1,CA,CB,CC,CD,CE
  READ 13,A,B,C,HM8R,NI
  READ 3,HMIN,SMIN,DHMIN,TMED,CMED,TMAX,TMIN
  1 FORMAT (5E15.7)
  2 FORMAT (9F8.0)
  3 FORMAT (7F10.0)
  PRINT 10
  PRINT 11
  NI=NI-1
  4 READ 2,T,HA,HB,HC,DUM,SA,SR,SC,DUM
  NI=NI+1
  IF(T-TMED)5,5,6
  5 CP=CA+CB*T+CC/T**2+CD*T**2
  GO TO 7
  6 CP=CMED+CE*(T-TMED)/(TMAX-TMED)
  10 FORMAT(1H1,11THERMODYNAMIC DATA FOR THE COMPOUND,11//)
  11 FORMAT(1H,6X,3HT,K,12X,2HCP,13X,64H-HMIN,9X,1HS,14X,4HDELH,11X,
  14HDELH,11X,4HLNKP,4X//)
  13 FORMAT(4F10.0,15)
  7 CONTINUE
  8 IF(T-TMIN)9,9,10
  9 DT=T-TMIN
  TSTOR=TMIN
  HMMS=HMIN
  SST=SMIN
  GO TO 12
  10 DT=T-TSTOR
  HMMS=HMM
  SST=S
  12 DH=CP*DT
  DS=CP*LOGF(T/TSTOR)
  HMM=HMMS+DH
  S=SST+DS
  HMTM=HMM-HMIN
  DELH=DHMIN+HMTM-HA*A-HB*B-HC*C+HM8R
  DELS=S-SA*A-SB*B-SC*C
  DELF=DELH-T*DELS
  ALNKP=-DELF/(1.987*T)
  DELHKC=DELH/1000.
  HMMKC=HMM/1000.
  DELFKC=DELF/1000.
  PRINT 14,T,CP,HMMKC,S,DELHKC,DELFKC,ALNKP
  PUNCH 15,NI,T,CP,HMMKC,S,DELHKC,ALNKP
  14 FORMAT(7F15.4)
  15 FORMAT(15,6F12.4)
  TSTOR=T
  GO TO 4
  END
```

SAMPLE OUTPUT

Table 1 shows a sample output, in this case, the compound UO_3 in the condensed state, extrapolated beyond the limits of stability.

Table 1. Sample Output from Program ONEPHASE - UO₃
(Condensed State, Extrapolated)

THERMODYNAMIC DATA FOR THE COMPOUND, UO₃(c)

T,K	CP	H-MIN	S	DELH	DELF	LNKP
1100.0000	21.8081	16.5170	49.3000	-291.0000	+220.7006	100.9748
1200.0000	21.8842	18.7054	51.2042	-290.9986	-214.3098	89.8900
1300.0000	21.9683	20.9015	52.9619	-291.0030	-207.9193	80.4922
1400.0000	22.0364	23.1051	54.5950	-291.0114	-201.5287	72.4454
1500.0000	22.1125	25.3163	56.1206	-295.7237	-194.8226	65.3557
1600.0000	22.1886	27.5352	57.5526	-295.7323	-188.9949	59.1542
1700.0000	22.2647	29.7617	58.9024	-295.7438	-181.3660	53.6919
1800.0000	22.3408	31.9957	60.1794	-295.7582	-174.6378	48.8279
1900.0000	22.4169	34.2374	61.3914	-295.7741	-167.9096	44.4758
2000.0000	22.4930	36.4867	62.5451	-295.7898	-161.1811	40.5586
2100.0000	22.5691	38.7436	63.6463	-295.8053	-154.4487	37.0141
2200.0000	22.6452	41.0082	64.6998	-295.8238	-147.7171	33.7917
2300.0000	22.7213	43.2803	65.7098	-295.8417	-140.9869	30.8498
2400.0000	22.7974	45.5608	66.6800	-295.8590	-134.2532	28.1518
2500.0000	22.8735	47.8474	67.6137	-295.8786	-127.5215	25.6710
2600.0000	22.9496	50.1424	68.5138	-295.8977	-120.7836	23.3796
2700.0000	23.0257	52.4449	69.3828	-295.9181	-114.0464	21.2579
2800.0000	23.1018	54.7551	70.2230	-295.9354	-107.3120	19.2882
2900.0000	23.1779	57.0729	71.0363	-295.9546	-100.5739	17.4538
3000.0000	23.2540	59.3983	71.8247	-295.9737	-93.8343	15.7414
3100.0000	23.3301	61.7313	72.5897	-295.9912	-87.1952	14.1395
3200.0000	23.4062	64.0719	73.3328	-296.0056	-80.5599	12.6384
3300.0000	23.4823	66.4201	74.0554	-296.0233	-73.9215	11.2276
3400.0000	23.5584	68.7760	74.7587	-296.0410	-66.8806	9.8997
3500.0000	23.6345	71.1394	75.4438	-296.0526	-60.1430	8.6481
3600.0000	23.7106	73.5105	76.1117	-296.0635	-53.3991	7.4951
3700.0000	23.7867	75.8892	76.7635	-296.0748	-46.6575	6.3463
3800.0000	23.8628	78.2755	77.3998	-296.0790	-39.9147	5.2863
3900.0000	23.9389	80.6693	78.0217	-296.0822	-33.1799	4.2817
4000.0000	24.0150	83.0708	78.6297	-296.0837	-26.4383	3.3264
4100.0000	24.0911	85.4799	79.2245	-296.0820	-19.6951	2.4176
4200.0000	24.0910	87.8890	79.8051	-403.0724	-13.9776	1.3154
4300.0000	24.0910	90.2982	80.3719	-403.1953	-1.6340	-.1912
4400.0000	24.0910	92.7073	80.9258	-403.3347	7.7010	-.8808
4500.0000	24.0910	95.1164	81.4672	-403.4771	17.1425	-1.9060
4600.0000	24.0910	97.5255	81.9967	-403.6235	26.3947	-2.8978
4700.0000	24.0910	99.9346	82.5148	-403.7814	35.7458	-3.8276
4800.0000	24.0910	102.3437	83.0220	-403.9528	45.1968	-4.7283
4900.0000	24.0910	104.7528	83.5187	-404.1242	54.4485	-5.5923
5000.0000	24.0910	107.1619	84.0054	-404.3026	63.8077	-6.4225
5100.0000	24.0910	109.5710	84.4825	-404.4810	73.1790	-7.2214
5200.0000	24.0910	111.9801	84.9503	-404.6749	82.5427	-7.9987
5300.0000	24.0910	114.3892	85.4092	-404.8663	91.9203	-8.7285
5400.0000	24.0910	116.7983	85.8595	-405.0637	101.2889	-9.4400
5500.0000	24.0910	119.2074	86.3015	-405.2611	110.6716	-10.1269
5600.0000	24.0910	121.6165	86.7356	-405.4690	120.1495	-10.7888
5700.0000	24.0910	124.0256	87.1620	-405.6649	129.4355	-11.4283
5800.0000	24.0910	126.4347	87.5810	-405.8753	138.8287	-12.0463
5900.0000	24.0910	128.8438	87.9928	-406.0812	148.2188	-12.6431
6000.0000	24.0910	131.2529	88.3977	-406.2846	157.6200	-13.2209

NOMENCLATURE

A, B, C, D = constants in specific heat equation (1)

a, b, c, d = stoichiometric coefficients

A_i = generalized name for compound

C_p = specific heat at constant pressure (cal/mole-°K)

C_p(med) = specific heat at the highest temperature for which
equation (1) holds (cal/mole-°K)

ΔF_f⁰ = Gibbs free energy of formation of the compound at standard
state (cal/mole)

ΔH_f⁰ = enthalpy of formation of compound at standard state (cal/mole)

ΔS_f⁰ = entropy of formation of compound at standard state (e.u.)

ΔT = temperature increment (°K)

E = constant in specific heat equation (2) (cal/mole-°K)

H = enthalpy (cal/mole)

K_p = equilibrium constant of formation (dimensionless)

R = universal gas constant (1.987 cal/mole-°K)

S = entropy (e.u.)

T = temperature (°K)

T_{med} = highest temperature for which specific heat equation (1) holds (°K)

T_{min} = lowest temperature for which calculations are to be made (°K)

T_{max} = highest temperature for which calculations are to be made (°K)

v_i = generalized stoichiometric coefficient (dimensionless)

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