Thermodynamic Calculation of Melting Heat

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Abstract

In the present paper, the method for approximate calculation of phase space volumes proposed in Ref. [1] is extended. Expressions for the melting heat of materials with f.c.c., b.c.c. and h.c.p. lattices are derived. Experimental results are compared to numerical calculations for a number of elements.

Keywords: Thermodynamic, Heat; First-order phase transitions

Calculation of a Phase Volume for Solids

A principally new approach to the calculation of the heat of first-order phase transitions was proposed in Ref. [1]. It is based on an approximate calculation of the phase volumes of a system in different phases: solid, liquid or gaseous and calculation of the change of entropy given as a logarithmic ratio of the volumes of old and new phases. The volumes of phase space for liquid and gaseous phases are also calculated therein:

\[
V_{phl} = \frac{(3\pi m k T)^{3/2}}{(3N/2)!} V_N^L
\]

(1)

\[
V_{phg} = \frac{(3\pi m k T)^{3/2}}{(3N/2)!} V_N^G
\]

(2)

Where \( V_N^L \) and \( V_N^G \) are the volumes of liquid and gaseous respectively. The expression for the evaporation heat is obtained in Ref. [1] in terms of other thermodynamic parameters of the process. A good agreement of numerical results and experimental data suggests that the method is versatile and can be used for calculating the transition heat of other first-order phase transitions. As it was shown in Ref. [1], the calculation should take into account the work performed against external forces during a phase transition with employment of the free volume. The expression for the phase volume of liquid was obtained in Ref. [1], whereas the calculation of a melting heat requires the phase space volume of solid state. The following model is used to find this volume. Every atom (molecule, ion) of a solid is assumed to vibrate near the equilibrium state, and its energy in a self-consisted periodic field is

\[
ed = \frac{4\pi}{3} \left( \frac{8m_1m_2m_3}{m_1m_2m_3} \right)^{1/3} \left( e_n - U_n(\vec{r}) \right)^{1/3}.
\]

Integration over \( n \) in the phase space gives:

\[
V_{phs} = \frac{4\pi}{3} \left( \frac{8m_1m_2m_3}{m_1m_2m_3} \right)^{1/3} \left( e_n - U_n(\vec{r}) \right)^{1/3} d^3 \vec{r}.
\]

It was assumed in Ref. [1] that near a point of first-order phase-transition, the majority of atoms are in the state with the average kinetic energy. This assumption is proved by a good agreement between numerical results and experimental data on evaporation heat. Consequently, according to the equipartition theorem for kinetic energy [3], one can substitute \( 3kT/2 \) for \( e_n - U_n(\vec{r}) \). Thus, the approximate volume of phase space for solid is given by:

\[
V_{phs} = \frac{4\pi}{3} \left( \frac{8m_1m_2m_3}{m_1m_2m_3} \right)^{1/3} \left( \frac{3kT}{2} \right)^{1/3} V_N^S.
\]

(3)

Calculation of the Melting Heat

The volume of phase space for solid is given by (3). In the first part of the present work [1] it was shown that the employment of the free volume (for unit mole, the free volume is \( V_f = V - N_f V_s \), where \( V \) is the geometrical volume, \( N_f \) is the Avogadro constant, and \( V_s \) is the volume of atom (molecule, ion)) gives a substantially better agreement between numerical and experimental results. The phase space volume for solid state is expressed as:

\[
V_{phs} = \frac{4\pi}{3} \left( \frac{8m_1m_2m_3}{m_1m_2m_3} \right)^{1/3} \left( \frac{3kT}{2} \right)^{1/3} V_N^S.
\]

(4)

and the phase space volume for liquid state is, respectively, (1):

\[
V_{phl} = \frac{4\pi}{3} \left( \frac{8m_1m_2m_3}{m_1m_2m_3} \right)^{1/3} V_N^L.
\]

Correspondingly, the logarithm of the ratio \( V_{phl}/V_{phs} \) is equal to:

\[
\ln \left( \frac{3\pi m k T}{N f} \right)^{3/2} \left[ V_{phl} / (3N/2)! \right] - \ln \left[ \frac{4\pi}{3} \left( \frac{8m_1m_2m_3}{m_1m_2m_3} \right)^{1/3} \left( \frac{3kT}{2} \right)^{1/3} V_N^S / V_f \right] / 3.
\]

By using the Stirling formula \( \ln (3N/2)! = 3N \ln (3N/2) / 2 - 3N / 2 \) and taking into account \( R = kN_f \), we obtain:

\[\Delta S = \frac{R}{2} \ln \frac{\pi e^2}{6} + \ln \frac{m^2}{m_{1}m_{2}m_{3}} + R \ln \frac{V_f}{V_N^S}.\]

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Thus, the expression for the melting heat has the form:

$$\lambda = \frac{RT}{2} \left( \ln \frac{\pi e^3}{6} + \ln \frac{m}{m_{xx} m_{yy} m_{zz}} \right) + RT \ln \left( 1 + \frac{\Delta V}{V_f} \right) + P\Delta V$$

(5)

Under normal pressure, the term $P\Delta V$ is on the order of $10^5 \times 10^{-6}$, and the value of the first term is $10^4$, hence, up to pressures of $10^{10}$ Pa the term $P\Delta V$ can be neglected. $V_f = V_f + \Delta V$. Finally, the expression for the melting heat at pressures below $10^{10}$ Pa has the form:

$$\lambda = \frac{RT}{2} \left( \ln \frac{\pi e^3}{6} + \ln \frac{m}{m_{xx} m_{yy} m_{zz}} \right) + RT \ln \left( 1 + \frac{\Delta V}{V_f} \right)$$

(6)

Let us term the dimensionless expression

$$S_m = \frac{1}{2} \ln \left( \frac{\pi e^3}{6} + \ln \frac{m}{m_{xx} m_{yy} m_{zz}} \right)$$

(7)

the structural melting constant. Its first term is determined by a substantial difference between liquid and solid states, and the second makes allowance for the corrections related to symmetry and the present contribution is determined by a substantial difference between liquid and solid states, and the second makes allowance for the corrections related to symmetry and forces of interaction between atoms for a particular crystal. Note that the expression

$$\ln \frac{m}{m_{xx} m_{yy} m_{zz}}$$

is sensitive to the values of effective masses. Let the effective masses be slightly greater than $m$, then

$$m_{xx} = m(1 + 0.0n), m_{yy} = m(1 + 0.0n), m_{zz} = m(1 + 0.0n)$$

and we have

$$\ln \frac{m}{m_{xx} m_{yy} m_{zz}} = 3 \ln \left( 1 + \frac{n}{100} \right) = -3 \cdot \ln \left( \frac{6}{\pi e^3} \right) = 2.353.$$  

Taking effective masses into account may reduce the value of the structural melting constant by 10-15% since the effective mass is, as a rule, greater than the mass of a free particle.

Calculation of effective masses is beyond the scope of the present work because it is a hard computational task for a particular material. The effective masses can be found from the analysis of phonon spectra as well, but this is the subject to a separate work. The present work considers only geometric factors in the expressions for effective masses for three different types of lattice: the face-centered lattice (f.c.c.), body-centered lattice (b.c.c.), and hexagonal close-packed lattice (h.c.p.). In calculations of metal ion volumes, the ion radii are used, and in calculations of the volumes of atoms Ne, Ar, Kr, and Xe the corresponding van der Waals radii are used [4,5].

The expression for the melting heat (6) has the form:

$$\lambda = \frac{RT}{2} \left( \ln \frac{\pi e^3}{6} + \ln \frac{m}{m_{xx} m_{yy} m_{zz}} \right) + RT \ln \left( 1 + \frac{\Delta V}{V_f} \right)$$

(8)

where $\Delta V$ is on the order of $10^5 \times 10^{-6}$, and the value of the first term is $10^4$, hence, up to pressures of $10^{10}$ Pa the term $P\Delta V$ can be neglected. $V_f = V_f + \Delta V$. Finally, the expression for the melting heat at pressures below $10^{10}$ Pa has the form:

$$\lambda = \frac{RT}{2} \left( \ln \frac{\pi e^3}{6} + \ln \frac{m}{m_{xx} m_{yy} m_{zz}} \right) + RT \ln \left( 1 + \frac{\Delta V}{V_f} \right)$$

(9)

The structural melting constant for the f.c.c. lattice is $S_m = 1.1765$.

For low-melting elements with weak interaction between atoms the effective masses are approximately equal to atomic masses, whereas for hard-melting materials the difference can be substantial. Results of calculations of the melting heat for f.c.c. lattices by formula (9) and solutions of equation (8) are given in Table 1.

Table 1: Calculation results of melting heat for f.c.c. lattices.
Table 2: Calculation results of melting heat for b.c.c. lattices.

<table>
<thead>
<tr>
<th>El-t</th>
<th>T</th>
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<th>ΔV</th>
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Table 3: Calculation results of melting heat for h.c.p. lattices.

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Experimental values of the melting heat and the line of a simple regression are shown in Figure 1. The slope of the regression line is 1.1670, which well coincides (with a good accuracy of ~ 0.85%) with the structural melting constant of the f.c.c. lattice \( S_0 = 1.1765 \).

Melting heat for the b.c.c. lattice

To the b.c.c. lattice in the \( \bar{p} \)-space corresponds the f.c.c. lattice. Hence, taking into account geometrical factors we take...
m_w = m_m = m_a = m_c = \frac{m}{\sqrt{2}}. Then the expression for the melting heat in the case of b.c.c. lattice takes the form

\lambda = \frac{1}{2} RT \left\{ \ln \left( \frac{\pi c^2}{6} \right) + \ln \left( \frac{a}{2\sqrt{2}c} \right) + RT \ln \left( 1 + \frac{\Delta V}{V_S} \right) \right\},

or

\lambda = \frac{1}{2} RT \left\{ \ln \left( \frac{\pi c^2}{6} \right) + RT \ln \left( 1 + \frac{\Delta V}{V_S} \right) \right\} (10)

The structural melting constant for the b.c.c. lattice is \( S_0 = 1.0032 \).

Results of calculations of the melting heat for b.c.c. lattices by formula (10) are given in Table 2 along with the results of solving equation (8).

T is the melting temperature in K, \( \lambda_{\text{ex}} [\text{J/mol}] \) is the experimental value of the melting heat, \( V_m^* [\text{m}^3/\text{mol}] \) is the molar volume of the solid state, \( \Delta V^* [\text{m}^3/\text{mol}] \) is the volume jump in melting, \( r^* [\text{m}] \) is the radius of atom (ion), \( V_S^* [\text{m}^3/\text{mol}] \) is the free volume, \( (dT/dP)^* [\text{K}]/[\text{Pa}] \) is the initial slope of the melting curve, \( \lambda_{\text{min}} [\text{J/mol}] \) is the molar melting heat obtained by solving formula (10), \( \lambda_{\text{min}} [\text{J/mol}] \) is the molar melting heat calculated by formula (12), \( \lambda_{\text{in}} [\text{J/mol}] \) is the molar melting heat obtained by solving equation (8), \( \delta_1 \) and \( \delta_2 [%] \) are the inaccuracies of \( \lambda_{\text{min}} \) and \( \lambda_{\text{in}} \), respectively.

Experimental values of the melting heat and the line of simple regression are presented in Figure 2 for the most low-melt elements. The slope of the regression line is 0.967 and matches the structural melting constant of the b.c.c. lattice \( S_0 = 1.0032 \) with a good accuracy of ~ 3.6%.

Melting heat for the h.c.p. lattice

h.c.p lattice transforms into itself in the \( \hat{\rho} - \) space. The effective masses are, respectively, \( m_w = m_m = m_a = m_{\text{c}} = \frac{2m}{\sqrt{3}} \). Thus, the expression for the melting heat for h.c.p. lattice has the form

\lambda = \frac{1}{2} RT \left\{ \ln \left( \frac{\pi c^2}{6} \right) + \ln \left( \frac{a}{2\sqrt{3}c} \right) + RT \ln \left( 1 + \frac{\Delta V}{V_S} \right) \right\}, \quad (11)

where \( a \) and \( c \) are parameters of the h.c.p. lattice. Since the ratio \( c/a \) for the elements does not differ much from the ideal case \( c/a = \sqrt{3}/2 \), we may replace real values of the ratio \( a/c \) in the expression for \( \lambda \) with the ideal value and obtain the relationship for the melting heat for the h.c.p. lattice:

\lambda = \frac{1}{2} RT \left\{ \ln \left( \frac{\pi c^2}{6} \right) + RT \ln \left( 1 + \frac{\Delta V}{V_S} \right) \right\} \quad (12)

The structural melting constant for the h.c.p. lattice is \( S_0 = 1.075 \).

Results of calculations by formula (12) are given in Table 3. The results of solving equation (8) for h.c.p. lattices are also presented in the table.

T is the melting temperature in K, \( \lambda_{\text{ex}} [\text{J/mol}] \) is the experimental value of the melting heat, \( V_m^* [\text{m}^3/\text{mol}] \) is the molar volume of the solid state, \( \Delta V^* [\text{m}^3/\text{mol}] \) is the volume jump in melting, \( r^* [\text{m}] \) is the radius of atom (ion), \( V_S^* [\text{m}^3/\text{mol}] \) is the free volume, \( (dT/dP)^* [\text{K}]/[\text{Pa}] \) is the initial slope of the melting curve, \( \lambda_{\text{min}} [\text{J/mol}] \) is the molar melting heat calculated by formula (12), \( \lambda_{\text{in}} [\text{J/mol}] \) is the molar melting heat obtained by solving equation (8), \( \delta_1 \) and \( \delta_2 [%] \) are the inaccuracies of \( \lambda_{\text{min}} \) and \( \lambda_{\text{in}} \), respectively.

Experimental values of the melting heat are presented in Figure 3 along with the line of a simple regression. The slope of the regression line is 1.1186 and matches the structural melting constant of h.c.p. lattice \( S_0 = 1.0075 \) with a good accuracy of ~ 4%.

Analysis and Conclusions

As one can see from Tables 1-3, the proposed model gives a satisfactory description of the melting heat for the elements considered. In Ref. [7], experimental data on temperatures and melting heat values for eight well characterized elements are given. As one can see from Table 4, even for these elements the experimental results noticeably differ.

\( \lambda_{\text{min}} \) and \( \lambda_{\text{in}} \) are the minimal and maximal values of experimental melting temperature, \( \lambda_{\text{ex}} \) and \( \lambda_{\text{in}} \) are the lowest and highest values of experimental melting heat. The same trend holds true for the values given in various handbooks. The results only coincide if data are taken from a single source. Consequently, it would not be correct to hope for a good agreement between experimental data and the numerical results calculated by formulae (9-10, 12).

The substantial difference between the experimental value of the melting heat for lead and the corresponding value calculated for the f.c.c. lattice along with the satisfactory agreement with the value of the melting heat calculated for the b.c.c. lattice suggest that a polymorphic transition from the f.c.c. lattice to b.c.c. lattice occurs near the melting point with the phase transition heat equal to \( \lambda = 1700 \text{–} 1800 \text{ J/mol} \). The phase transition heat is calculated in the frameworks of the proposed model.
Thus, one can assert that the molar melting heat at pressures below $10^{10}$ Pa is given by the expression

$$\lambda = R T S_0 + RT \ln \left(1 + \frac{\Delta V}{V_f}\right)$$

or by a solution of the equation

$$\lambda = R T S_0 + \ln \left(1 + \frac{\Delta T}{T S_0} \frac{dP}{dP}\right),$$

Where $S_0 = 1.1765$ for the f.c.c. lattice, $S_0 = 1.0032$ for the b.c.c. lattice and $S_0 = 1.075$ for the h.c.p. lattice. The rest values have been defined earlier.

References

7. Stolovich NN, Minitskaya NS (1975) Temperaturyne zavisimosti svoistv nekotorykh metallov (Temperature dependencies of the properties for certain metals).

Table 4: Spread of experimental values on temperatures and melting heat.

<table>
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<th>Elt</th>
<th>$T_{min}$</th>
<th>$T_{max}$</th>
<th>$\lambda_{min}$</th>
<th>$\lambda_{max}$</th>
<th>$T_{min}$</th>
<th>$T_{max}$</th>
<th>$\lambda_{min}$</th>
<th>$\lambda_{max}$</th>
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<td>7928.93</td>
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<td>3683</td>
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</tbody>
</table>

Figure 3: Experimental values of melting heat for elements with the h.c.p. lattice and the line of simple regression.