Patterns in new dimensionless quantities containing melting temperature, and their dependence on pressure

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ABSTRACT

The relationships existing between melting temperature and other macroscopic physical quantities are investigated. A new dimensionless quantity $Q_n$ not containing the Grüneisen parameter proves to be suited for serving in future studies as a tool for the determination of the melting temperature in the outer core of the Earth. The pressure dependence of more general dimensionless quantities $Q_n$ is determined analytically and, for the chemical elements, numerically, too. The patterns of various interesting dimensionless quantities are shown in the Periodic Table and compared.

ZUSAMMENFASSUNG

Die Beziehungen der Schmelztemperatur zu anderen makroskopischen physikalischen Meßgrößen wurden untersucht. Eine neue dimensionslose

It is the objective of this study to approximately express the melting temperature $T_m$ of elements and anorganic chemical compounds as a function of other physical quantities. The specific requirements are that (1), to permit an application in future studies, we endeavor to use only such quantities that are reliably known also for the outer core of the Earth from modern seismological and other geophysical models (e.g., after Dziewonski et al. 1975) and (2) the relationship is to be valid at least for high pressures ranging between 1354 and 3289 kbars (1 kbar = 100 MPa).

We start the search for this relationship in the normal pressure range. The starting point is a paper by Plendl (1974) from which Fig. 1 has been taken. $Z_*$ is the atomic coordination valency which is defined as the product of the atomic valence in the elemental state and the ratio of the first order atomic coordinations of two successive polymorphs. $C_s$ is the structural parameter, $C_{s(r)}$ is proportional to the density of
paching of the atoms $C^\infty$ is also proportional to the number of significant approaches of adjacent oscillating atoms. If we compare Fig. 1 with Fig. 2, we find that although there is an indication of the anticipated proportionality $T_m \sim Z_a C_{slr}$, but scattering unfortunately is considerably greater than given by Plendl. According to Plendl and Gielisse (1969, 1970), there is also a proportionality between bulk modulus $\alpha$ and $Z_a C_{str}$. Consequently, we may also expect a proportionality between melting temperature and bulk modulus. To verify this, we plot the melting temperature data of the elements after Gschneidner (1964) versus the bulk modulus for zero pressure after the data collected.

Fig. 1 - Linear relationship between the experimental data of the melting point ($T_m$) and the atomic data of $Z_a C_{slr}$ for 20 metallic elements with strong bondings (Figure and caption after Plendl, 1974).
by Ullmann and Pan'kov (1976) (see Fig. 3). The coordination numbers in the diagram are designated by symbols. There is, in fact, an indication of proportionality. But scattering is so great here, too, that it is hardly suited for practical purposes. If, however, we form in section 3 dimensionless quantities as few material-dependent as possible, we can assume from Fig. 3 that it must contain the quotient $T_m/c$.

2. COMPARISON OF A FEW DIMENSIONLESS QUANTITIES FOR ZERO PRESSURE

In this section, let us form a few dimensionless quantities for the chemical elements and study their correlations at the pressure $P = 0$. We define

$$N_1 = \frac{\rho}{R_e} \cdot \frac{\Delta x}{T_m P}$$

and plot this quantity in Fig. 4 versus the group numbers of the Periodic Table. $R_e = 8.31441 \text{ J/(K} \cdot \text{mol)}$ is the universal gas constant. We take the numbers for the formula weight $F$ in kg/mol from Ebert (1976), for the bulk modulus $\kappa$ and density $\rho$ from Ullmann and Pan'kov (1976) and for the melting temperature $T_m$ from Gschneidner (1964). Moreover, we define

$$N_2 = \frac{\rho}{R_e} \cdot \frac{3 \Delta x \Delta \rho}{\rho}$$

and plot this quantity, too, in the Periodic Table (see Fig. 5), using the linear coefficient of thermal expansion $\alpha$, the atomic volume $\rho/v$ and density $\rho$ from Gschneidner (1964). According to Gschneidner (1964) the product $\Delta \rho \Delta v = 0.0186 \pm 0.0080$ is nearly...
Fig. 3. Melting temperature $T_m$ vs. bulk modulus for zero pressure $x$. The legend shows the assignment between symbols and coordination number.
constant for all elements, $x$ being the coefficient of thermal expansion. However, the parallelism between the curves in Figs. 4 and 5 is not as good as expected. Incidentally, there is a
surprisingly good parallelism between $N_2$ and the Grüneisen parameter

$$\gamma = \frac{\alpha T_x}{\beta C_v},$$  \[3\]

where $x_1$ is the adiabatic bulk modulus, $x_2$ the isothermal bulk modulus, $C_v$ the specific heat at constant pressure, $C_p$ the specific heat at constant volume.

This is true irrespective of whether we use the generally, i.e., irrespective of the author, somewhat problematic $\gamma$-values after Gschneidner (1964) or those after Guinan and Steinberg (1974). This is revealed by a comparison between Fig. 5 and Figs. 6 and 7. As it is obvious when considering the relations 2 and 3, this implies an approximate proportionality between specific heat and the reciprocal value of the formula weight.

3. COMPUTATION OF NEW DIMENSIONLESS QUANTITIES $Q_2$ AS A FUNCTION OF PRESSURE

Following the considerations presented above, we now study the pressure dependence of dimensionless quantities containing the melting temperature. Expecting these quantities to be little dependent on pressure in the outer core of the Earth, we now define dimensionless quantities $Q_2$.

$$Q_2 = N^{-\gamma},$$  \[4\]

$n$ being a real number. A look at formulas (1) and (4) shows that, if a certain $Q_2$ in the outer core of the Earth is known as a function of depth, the quotient $T_c/F$ can be determined, when $\rho$, $x$ and $\gamma$ are taken from seismological models of the Earth. $T_c$ is the melting temperature and $F$ the formula weight. The problematic Grüneisen parameter $\gamma$ (see discussion by Anderson
Fig. 4. The dissociation energy $E_d$ of the elements of the second, third, fourth, fifth and sixth periods of the Periodic Table.
Fig. 5 - The dimensionless quantity $N_2$, defined by Eq. [2], of the elements of the second, third, fourth, fifth and sixth periods of the Periodic Table.
and Mulargia (1977), is not required for \( Q \). The quantity \( Q \), could, for instance, become known as a function of pressure in the outer core by the possibility that \( Q \) of the core-candidate materials for core pressures lie closely adjacent to one another. To be able to study the pressure dependence of the quantities \( Q \), we have to make an assumption as to the form of the interatomic potential. It has already been pointed out by Boschi...
et al. (1979) that the behaviour of the melting temperature in the outer core considerably depends on the interatomic potential assumed. In the following, we assume the Lennard-Jones potential. This is a rough but realistic approximation. The author has shown in another study (Walzer, 1982b) that another, more complicated expression for the potential arrived at by means of quantum mechanics reveals the best agreements with experimental data for metals, halides and some oxides. However, a Lennard-Jones potential can be used as a rough first approximation, even more so since this greatly facilitates computation:

\[ \Phi = \frac{A}{x^{12}} - \frac{B}{x^{6}} \]  \[ (5) \]

\( A, B \) and \( x \) are constants which depend only on the material, \( x = V/V_0 \), \( V \) being the volume, \( V_0 \) the volume at vanishing pressure \( P \). Starting from \( (5) \) and a dislocation model, the author has proven (1982a) that Lindemann’s melting law

\[ T_m = \frac{d T_m}{d P} = \frac{2}{3} (\gamma - 1/3)/x \]  \[ (6) \]

holds for pressure ranges where the solid is present in the densest spherical packing (i.e., in fcc or hcp structure). Stacey and Irvine (1977), using a different, i.e., a thermo-dynamical, approach also achieved this result. \( T_m \) is the melting temperature, \( \gamma \) the Grüniesen parameter \( \gamma \) depends on pressure \( P \) in the following manner:

\[ \gamma = \frac{1}{2} \frac{d x}{d P} \frac{5}{9} + \frac{2}{9} \frac{P}{x} \frac{d x}{d P} \frac{1}{5} \]  \[ (7) \]
This formula had already been deduced in a completely different way from the free volume theory by Vashchenko and Zubarev (1963). Using a simple method that has already been employed by Kittel (1953), we deduce the equation of state belonging to [5]. From [5] we obtain

\[ P = \frac{\partial \Phi}{\partial x} = \frac{2nA}{3\bar{v}^{n+1}} - \frac{nB}{3\bar{v}^{n+1}}. \quad (9) \]

\[ x = - \frac{2n}{3} \frac{3P}{\partial x} = \frac{2n}{3} \left( \frac{2n}{3} + 1 \right) \frac{A}{\bar{v}^{n+1}} + \]

\[ - \frac{nm}{3} \left( \frac{m}{3} + 1 \right) \frac{B}{\bar{v}^{n+1}} \quad (9) \]

as well as

\[ x = - \frac{2n}{3} \frac{3P}{\partial x} = \frac{2n}{3} \left( \frac{2n}{3} + 1 \right) \frac{A}{\bar{v}^{n+1}} + \]

\[ - \frac{nm}{3} \left( \frac{m}{3} + 1 \right) \frac{B}{\bar{v}^{n+1}} \quad (10) \]

If we note that \( \frac{2n}{3} = -x \frac{2n}{3} \frac{1}{x} \)

and if we use \( x_1 = \frac{x}{x-1} \)

and \( x_1 = \frac{2n}{3} \frac{1}{x} \)

the constants A, B, n can be expressed through \( x_1 \) and \( x_1 \) with the help of (8), (9) and (10). This is very advantageous, because the latter quantities are macroscopically observable. In this
In this way, using a different approach, we obtain from (8) the equation of state M1 by Ullmann and Pan'kov (1976)

\[
P = - \frac{3 n}{2 - n} \left( \frac{n}{3} - \frac{1}{2} \frac{\Delta H}{n} - \frac{1}{2} \frac{\Delta H}{\Delta H} \right) \left( T + \frac{P}{2} \right)
\]

We now use the compatible formulas [6], [7] and [11] for an examination of the pressure dependence of the dimensionless quantities \( \Omega \) and \( \Theta \).

From formula (1) it follows that

\[
\frac{\partial N}{\partial P} = \frac{1}{T_e} \frac{\partial x}{\partial P} - \frac{x}{T_e} \frac{\partial x}{\partial P} \left( \frac{\partial}{\partial P} + \frac{\partial}{\partial T} \right) \frac{\partial N}{\partial T}.
\]

which is reformulated by means of Lindemann's law into

\[
\frac{\partial \ln N}{\partial P} = \frac{\partial x}{\partial P} - 2 \left( \frac{1}{3} - \frac{x}{P} \frac{\partial x}{\partial P} \right) \left( \frac{1}{3} - \frac{x}{P} \right) \frac{\partial N}{\partial T}.
\]

\( \tau \) can be eliminated by formula (7). Furthermore, we note that

\[
\frac{\partial \tau}{\partial P} = \frac{1}{x}.
\]

Thus, we obtain from (13)

\[
\frac{\partial \ln N}{\partial P} = \frac{1}{\frac{3}{4} - P} \frac{\partial x}{\partial P} \left( \frac{3}{4} - \frac{x}{P} \right) \frac{\partial N}{\partial P}.
\]
Through similar computations we obtain from Vashchenko's and Zubarev's (1963) formula [7]

\[
\frac{\partial \ln \tau}{\partial P} = \frac{4/1}{9/1} \left( \frac{P}{x^2 P} \right) - \frac{4/1}{9/1} \left( \frac{P}{x^2 P} \right)
\]

From equations [1], [4] and [14] we obtain

\[
q_s = \frac{2 \ln \Omega_s}{2 P} = \frac{P}{x} \frac{\partial x}{\partial P} - 1 + \frac{2 \ln y}{2 P} .
\]

If [15] is substituted in [16], we obtain a formula by means of which it is principally possible to determine $Q_s = Q_s(P)$ by integration. So far, no use has been made of the specially chosen equation of state [11]. One mathematical difficulty arises because of the fact that the Ullmann-Pan'kov equation of state (like its special case, the Birch equation of state) cannot be analytically transformed from $P = P(x)$ to $x = x(P)$. This difficulty has been eluded by the following transformations

\[
\frac{3 \ln \Omega_s}{\partial P} = \frac{P(x)}{s(x)} \frac{\partial x}{\partial P} - \frac{3}{4} \frac{x}{s(x)} - P(x).
\]

\[
x = -x_0 \frac{\partial P}{\partial x} .
\]

\[
\frac{x}{x_0} = -x \frac{\partial x}{\partial x}
\]
Fig. 7 - Grüneisen ratio $\gamma$ after values given by Guinan and Steinberg (1974), plotted in the Periodic Table. This means in cases where the column has a disappearing height that no numerical value is known.

$$\frac{\partial \gamma}{\partial P} = \left( \frac{\partial P}{\partial x} \right)^{-1} \left( \frac{\partial x}{\partial P} \right) - \frac{\partial x}{\partial x} \left( \frac{\partial P}{\partial x} \right)$$
In the auxiliary function \( q' \) defined by [16] we eliminate \( P, x, \frac{3x}{P} \) and \( \frac{2x}{P^2} \) through the equations [11], [15] and [17] to [20]. Thus, \( q \) is known as a function of \( x \). The equation of state effects that \( P \)-values bi-uniquely correspond to the \( x \)-values. Using a computer, we calculated \( Q' \) as a function of pressure \( P \) by means of numerical integration of [16].

4. The systematics of the quantities \( Q \) in the Periodic Table

To study the distribution of the dimensionless quantity \( Q' \) (i.e., \( Q \) for \( n = 1 \)), we plotted this quantity in Fig. 8 against the groups of the Periodic Table. Using the formulas of the previous section, we computed \( Q \) for the pressure at the core-mantle boundary (CMB) for the chemical elements and plotted it in Fig. 9 against the groups. Fig. 10 shows the corresponding distribution for the pressure at the inner-core boundary (ICB). A comparison of these three figures shows that the change seen in the characteristic curves is less considerable for the outer core than for the mantle. The material parameters required for the computation have been listed in Tab. 1, their origin is given in the legend of the Table. In Figs. 11 to 13, the quantities \( Q \) have been plotted as a function of pressure. Generally, no strong concentration of the curves can be found for the elements of periods 4 to 6. It is particularly apparent for the 5th and 6th periods that the elements with the densest spherical packing (i.e., those present in hcp or fcc structure) are predominant in the upper curves. Much more favourable results are obtained for the dimensionless quantity \( Q \), which is plotted against the pressure in Fig. 14 for the 4th period, in Fig. 15 for the 5th period and in Fig. 16 for the 6th period. In Fig. 14, all fcc and hcp materials are found in the strong middle bundle. Only the element Sc does not fit in this scheme. In Fig. 15, the same results are found for the elements of the 5th period. With the exception of Y and Sr, all elements with fcc or hcp lattices are found in the strong middle bundle of curves. Two clearly separated curve bundles can also be seen for the elements of the 6th period in Fig. 16. Except Hf and Re, all elements with fcc and hcp structures
Fig. 8. Plot of the dimensionless quantity $Q_0$ in the Periodic Table for the elements of the 4th period (continuous line), 5th period (dotted line) and 6th period (point line) at pressure $P = 2$. 
are found in the lower narrow bundle of curves, while the box structure is predominant in the upper bundle. Since, if the temperatures were lower, one would have to reckon with the densest spherical packings for the pressures existing in the outer core, the fcc-hcp bundles of $Q$ provide a possibility for estimating the quotient $T_s/P$ for the outer core without becoming too specific in regard of the chemical composition. In Fig. 17, $Q$ has been plotted against the groups of the Periodic Table, the pressure being $P = 1354$ kbars. These characteristics do not significantly vary for the higher pressures existing in the interior of the outer core. The characteristics curves of $Q$ at $P = P_{crit}$ reveal a slight similarity to the Leibfried number $L$, the modified Leibfried number $L'$, and the Bragg number $B$, as can be seen from a comparison of Fig. 17 with Figs. 18 to 20. However, these quantities are given for zero pressure. The Figures were plotted according to the numerical values given by Gschneidner (1964).

As defined in the paper by Leibfried (1950), the Leibfried number is

$$L = \frac{R_s T_s}{\pi \sigma v}.$$  \[21\]

where $v$ is the atomic volume and $\sigma$ the shear modulus. This quantity was modified by Gschneidner (1964):

$$L' = \frac{K T_s}{\pi \sigma v}.$$  \[22\]

In this modified Leibfried number, $K$ is a constant which, however, has a different value for each crystal structure, while $R_u$ denotes the universal gas constant as before. The Bragg (1948) number, on the other hand, is defined by

$$B = \frac{1}{\pi \sigma v}.$$  \[23\]
Fig. 9: Plot of the quantity $Q_i$ in the Periodic Table at the pressure relating to the non-elastic boundary. Designations as in Fig. 8.
Fig. 10 - Plot of the quantity $Q$ in the Periodic Table at the pressures existing at the inner-core boundary. Designations as in Fig. 8.
Fig. 11 - Dependence of the dimensionless quantity Q on pressure for the chemical elements of the 4th period of the Periodic Table.
Fig. 13 - Dependence of the quantity $Q$ on pressure for the elements of the 6th period.
Fig. 14 - Dependence of the dimensionless quantity Q on pressure for the chemical elements of the 4th group of the Periodic Table. A comparison with Fig. 11 reveals a significantly closer proximity of the curves.
Fig. 15 - Pressure dependence of the quantity $Q_0$ for the elements of the 5th period.
Fig. 17: The dimensionless quantity $Q$, subjected to the pressure existing at the core-mantle boundary (CMB). Representation in the Periodic Table for the elements of the 4th period (continuous line), 5th period (dotted line) and 6th period (spaced line).
Fig. 18 - The Liebfried number $L$ plotted in the Periodic Table for zero pressure. Designations as in Fig. 17.
Fig. 19. The modified Leibfried number $L'$, plotted in the Periodic Table for zero pressure. Designations as in Fig. 17.
Fig. 20: The Bragg number, plotted in the Periodic Table for zero pressure. Designations as in Fig. 17.
where $AH$ is the heat of fusion. In view of Richard's rule $AH = R \Delta T_m$, it does not surprise that $L$, $E'$, and $B$, when plotted in the Periodic Table, yield very similar characteristic curves (cf. Figs. 18 to 20). However, these quantities are not suited for estimating $T_m$ in the outer core, because the shear modulus $\mu$ disappears in the liquid state. In contrast to this, the bulk modulus $K$ does not vary considerably during melting. Therefore, $Q_i$ appears to be a quantity useful for geophysical purposes. It goes without saying that the pressure dependence of the individual quantities must be taken into account. Thus, Fig. 17 shows a comparable representation of $Q_i$ for the pressure at the core-mantle boundary.

5. Conclusions

A systematic correlation between the melting temperature and other physical quantities has been found. In particular, two new, dimensionless quantities, $Q_i$ and $Q_0$, depending on $T_m$, were investigated with respect to their dependence on pressure as well as to their distribution in the Periodic Table. While the $Q_i$ for pressures in the Mbar range strongly differ from one another for the individual chemical elements, it was found for $Q_0$ that the curves of materials having the same lattice structure prior to melting run very close to one another. The results suggest that it should be possible even without detailed knowledge of the chemical composition of the outer core to determine the melting temperature to formula weight ratio as a function of depth.
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<td>457.8</td>
<td>152.60</td>
<td>130.04</td>
<td>6.73</td>
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### Table I (continuation)

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<th>Element</th>
<th>( T_m [K] )</th>
<th>( \rho [g/cm^3] )</th>
<th>( x_{\text{kl}} [\mu m] )</th>
<th>( F [\text{kg/mol}] )</th>
<th>( N_1 )</th>
<th>( x_1 )</th>
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<td>Hf</td>
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<td>360</td>
<td>208.19</td>
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TABLE I  -  Physical constants of chemical elements used for $P = 0$. Melting temperature $T_m$ according to Gschneidner (1964), formula weight $F$ according to Ebert (1976), density $\rho$ and bulk modulus $\kappa$ for the lanthanons and actinons according to Kittel (1973), density, bulk modulus and its derivative with respect to pressure for the other elements according to Ullmann and Pan'kov (1976). $N$ is the dimensionless quantity for $P = 0$ defined by Eq. [1]. $\gamma$ is the Ginzburg parameter used, if relevant, for the indicated element and phase. $\gamma$ applies to bcc iron. If the hypothetical melting point curve for bcc iron by Liu (1975) is drawn for low pressures, one attains the same value. The Ginzburg parameters $\gamma$ used are not listed in the Table. The values given by Guinan and Steinberg (1974) were used. Only for Fe, the value $\gamma = 1.65$ according to Plendl (1975) was preferred.
REFERENCES


PLENDL J.N., 1974 - Phys. stat. sol. a (3), 65, 519.


