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LITHIUM EQUATION-OF-STATE

James A. Blink

Previously published internally as E&MA 82-176
(Dec. 30, 1982).

September 1983

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LITHIUM EQUATION-OF-STATE

James A. Blink

INTRODUCTION

In 1977, Dave Young published an equation-of-state (EOS) for lithium.¹ This EOS was used by Lew Glenn in his AFTON calculations of the HY LIFE inertial-fusion-reactor hydrodynamics. In this paper, I summarize Young's development of the EOS and demonstrate a computer program (MATHSY) that plots isotherms, isentropes and constant energy lines on a P-V diagram.

THE EOS MODEL

Young's EOS uses the soft sphere model of Hoover, et al.² The soft sphere model is based on Monte Carlo calculations for particles interacting with pair potentials of the form

$$\phi(r) = \epsilon \left(\frac{\sigma}{r} \right)^n, \quad 4 \leq n \leq 12 \quad (1)$$

The Helmholtz free energy for Young's EOS is

$$A = NkT \left[-\ln \frac{V_e}{N\lambda^3} + C_n \rho^{n/3} \left(\frac{\epsilon}{kT} \right) + \frac{1}{2}(n+4)Q\rho^{n/9} \left(\frac{\epsilon}{kT} \right)^{1/3} - \rho^m \left(\frac{\epsilon}{kT} \right) \right] + E_{coh} \quad (2)$$

In the brackets, the first term represents the kinetic energy, the second term is the static lattice potential energy, the third term is the thermal lattice potential energy correction, and the fourth term is the attractive potential energy. In the first term, λ is the deBroglie wavelength, $h/\sqrt{2\pi mkT}$. In the second term, C_n is the fcc Madelung constant, and ρ is the normalized density, $N\sigma^3/\sqrt{2}V$. In the third term, Q is a multiplier to account for electronic effects on the heat capacity. The fourth term is a mean-field attraction, since the actual attractive potential is complex and poorly known.

The pressure, energy, and entropy may be found from

$$P = - \left(\frac{\partial A}{\partial V} \right)_{N,T} \quad (3)$$

leg

$$E = A - T \left(\frac{\partial A}{\partial T} \right)_{N,V} \quad (4)$$

$$S = - \left(\frac{\partial A}{\partial T} \right)_{N,V} \quad (5)$$

The results of the differentiations are

$$P = \frac{NkT}{V} \left[1 + \frac{1}{3} n c_n \rho^{n/3} \left(\frac{\epsilon}{kT} \right) + \frac{1}{18} n(n+4)Q \rho^{n/9} \left(\frac{\epsilon}{kT} \right)^{1/3} - m \rho^m \left(\frac{\epsilon}{kT} \right) \right] \quad (6)$$

$$E = NkT \left[\frac{3}{2} + c_n \rho^{n/3} \left(\frac{\epsilon}{kT} \right) + \frac{1}{6} (n+4)Q \rho^{n/9} \left(\frac{\epsilon}{kT} \right)^{1/3} - \rho^m \left(\frac{\epsilon}{kT} \right) \right] + E_{coh} \quad (7)$$

$$S = (E-A)/T \quad (8)$$

Eqs. (6) and (7) are equivalent to Young's Eqs. (4) and (5).¹

The parameters ϵ , σ , n , m , and Q are adjustable and are chosen by fitting available enthalpy, volume, and sound speed isobars. Also, n is chosen such that $(n+2)/6 \approx \gamma_0$, the Gruneisen parameter at normal solid volume, as predicted by harmonic lattice theory. Similarly, m should be ~ 1.0 to agree with van der Waals EOS and its good fits to molecular fluid data. Finally, $Q = 1$ except where electronic corrections are clearly warranted.

Young's best fit values for Li are

$$\begin{aligned} n &= 4 & m &= 0.7 & Q &= 1.30 \\ \epsilon &= 1.99436 \times 10^{-17} \frac{\text{J}}{\text{atom}} & \sigma^3 \sqrt{2} &= 1.29408 \times 10^{-31} \text{ m}^3/\text{atom} \end{aligned}$$

Three comparisons of the EOS and experimental data are shown in Fig. 1 for enthalpy, thermal expansion, and sound speed; the agreement is reasonable. The EOS also produces the following critical point values:

$$T_c = 3741 \text{ K} \quad P_c = 114 \text{ MPa} \quad V_c/V_0 = 5.86 \quad Z_c = P_c V_c / NkT_c = 0.28$$

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Fig. 1. Soft-sphere fit (smooth curves) to experimental lithium thermodynamic data (points). (From Ref. 1.)

The predicted Gruneisen constant at V_0 is 1.0 (comparable to a measurement of 0.9).

The independent variables are $V(\text{m}^3/\text{kg})$ and $T(\text{K})$. Other useful Li data include

$$\text{Li atomic weight} = 0.006939 \text{ kg/mole}$$

$$N = \text{atoms/kg} = 6.0225 \times 10^{23} / 0.006939 = 8.679 \times 10^{25}$$

$$k = \text{Boltzman constant} = 1.38066 \times 10^{-23} \text{ J/atom K}$$

$$\lambda = \text{de Broglie wavelength} = 6.62618 \times 10^{-23} (2\pi kT/N)^{-1/2} T^{-1/2} \text{ (meters)}$$

$$E_{\text{coh}} = \text{Cohesive energy} = 23.03 \times 10^6 \text{ J/kg}$$

$$C_n = \text{fcc Madelung constant} = 12.669$$

$$V_0 = \text{Volume at room temperature} = 0.001876 \text{ m}^3/\text{kg}$$

$$T_m = \text{Melt temperature} = 453.7 \text{ K}$$

$$V_m = \text{Volume at melt} = 0.001931 \text{ m}^3/\text{kg}$$

$$H_m = \text{Enthalpy at melt} = 1.034 \times 10^6 \text{ J/kg (H = 0 at room temperature)}$$

After inserting the constants, Eqs. (2), (6), and (7) become

$$\begin{aligned} A\left(\frac{\text{J}}{\text{kg}}\right) = & 1198.16 T \left[-\ln(107.573 VT^{3/2}) + 4.60365 V^{-4/3} T^{-1} \right. \\ & \left. + 3.71071 V^{-4/9} T^{-1/3} - 495.542 V^{0.7} T^{-1} \right] + 2.303 \times 10^7 \end{aligned} \quad (9)$$

$$\begin{aligned} P(\text{Pa}) = & 1198.16 TV^{-1} \left[1 + 6.13820 V^{-4/3} T^{-1} \right. \\ & \left. + 1.64920 V^{-4/9} T^{-1/3} - 346.879 V^{-0.7} T^{-1} \right] \end{aligned} \quad (10)$$

$$\begin{aligned} E \frac{\text{J}}{\text{kg}} = & 1198.16 T \left[1.5 + 4.60365 V^{-4/3} T^{-1} \right. \\ & \left. + 1.23690 V^{-4/9} T^{-1/3} - 495.542 V^{-0.7} T^{-1} \right] + 2.303 \times 10^7 \end{aligned} \quad (11)$$

THE COMPUTER PROGRAM

A MATHSY³ macro was written to plot the equations for pressure, internal energy, and entropy. A copy of the macro is attached as Appendix A. It will run on the LLNL Cray computers. To obtain the macro, sign onto a Cray and type

```
XPORT RD .080975:EOS:EOS
```

The macro file EOS is used with the public file MATHSY. To begin, type

```
MATHSY TTT BNN ID  
RMAC EOS HCINIT
```

where TTT is the three digit TMDs monitor number, BNN is the box number, and ID is a one-word identifier.

There are five executable programs in the EOS macro.

POINT

```
POINT DDD,TTT
```

This command prints eight values on the teletype. The two inputs are the density (DDD) in kg/m^3 , and the temperature (TTT) in $^{\circ}\text{C}$. Default input values are 500 kg/m^3 and 500°C . The eight outputs are the temperature in Kelvins and $^{\circ}\text{C}$, the density in kg/m^3 , the specific volume in m^3/kg , the pressure in MPa, the internal energy in J/kg, the Helmholtz free energy in J/kg, and the entropy in J/kg K.

PLOT1

```
PLOT1 TTN,DDN, PPL,PPU, DDL,DDU, TTL,TTU
```

This command plots the pressure in MPa between the values PPL and PPU (MPa), as a function of specific volume (m^3/kg). The specific volume range is specified by inputting lower and upper densities DDL and DDU (kg/m^3). Temperature is a parameter, i.e., isotherms will be plotted between the values TTL and TTU ($^{\circ}\text{C}$). There will be TTN isotherms, and each isotherm will be plotted based on DDN equally spaced

density points. PLOT1 also indicates the coexistence curve and critical point supplied by Dave Young. Default input values are 33,100, -1500,500, 20,500, 400,3600. As an example, Fig. 2 demonstrates the plot obtained from the command

PLOT1 16,100, 0,130, 50,300, 2750,3500

The lines are the isotherms from 2750°C to 3500°C in 50°C increments. The labeled line "x" is Young's coexistence curve. The solitary "x" is Young's critical point. The coexistence curve peak should be at the critical point, indicating that Young's coexistence curve calculation is rough; however, the coexistence curve is accurate enough in the lower temperature region of interest to us.

PLOT2

PLOT2 TTN,DDN, DDL,DDU, TTL,TTU

This command plots the internal isotherms (in J/kg) as a function of the specific volume. The inputs and defaults are just as for PLOT1, except a range for the y-axis is not specified.

PLOT3

PLOT3 TTN,DDN, DDL,DDU, TTL,TTU, EEE

This command produces the same plot as the similar PLOT1 command (pressure isotherms as a function of specific energy). In addition, each isotherm is labeled with an "E" at the point where one line of constant internal energy (EEE in J/kg) crosses. The default value for EEE is 3.1495×10^6 J/kg, i.e., the internal energy immediately after 800 J/g of energy is added isochorically to 500°C saturated liquid.

PLOT4

PLOT4 TTN,DDN, DDL,DDU, TTL,TTU, SSS

This command also produces the same plot as the similar PLOT1 command (pressure isotherms as a function of specific energy). In addition, each isotherm is labeled with an "O" at the point where one line of constant entropy (SSS in J/kg·K) crosses. The default value for SSS is 7809 J/kg K, which is the approximate entropy after 800 J/g is isochorically added to 500°C saturated liquid.

Fig. 2. P-V diagram produced by the PLOT1 command. The 16 isotherms are at 50°C intervals between 2750°C and 3500°C.

As an example, Fig. 3 shows the plot obtained from the command

PL0T4 21,200, 0,500, 450,500, 400,800, 7809

For convenience, the isentrope points have been connected, and the abscissa relabeled. By the time isentropically expanding liquid reaches the coexistence curve, it has cooled down from 755.1°C to 695.4°C.

Figure 4 shows the plot obtained from the command

PL0T4 26,100, -1600,500, 300,500, 300,800, 7809

After the isentrope enters the two phase region, it continues to expand to lower (and then negative pressures). The isentrope is not continued beyond the spinodal line (where the isotherms have zero slope) since the fluid is then unstable to phase separation (back to the spinodal) on a very short time scale. Between the coexistence curve and the spinodal, the fluid is metastable, and it eventually separates into saturated liquid and vapor phases (on the coexistence curve) by the sequential processes of nucleation and growth.

THERMODYNAMIC VALUES AT POINTS OF INTEREST IN FIG. 3

The points of interest are the initial condition (500°C saturated liquid), the state after isochoric heating, the state after the isentropic expansion reaches the coexistence curve, and the state after the expansion reaches the spinode. By using the PL0T4 and POINT commands, these points were specified (Table 1).

During the heating, the internal energy increases by 799.9 J/g.. When the isentropic expansion back to the saturated liquid line is included, the net internal energy change is 783.9 J/g. The missing 16 J/g must be kinetic since no work or potential energy changes are possible in this isolated system and the change in surface energy is small. The kinetic energy of expansion is $mv^2/2$ where v is found from $p = \rho cv$. Substituting $m = 1$ g, $\rho = 484.364$ kg/m³, $c = 4500$ m/s, and $P = 380.15$ MPa, values of 174.4 m/s expansion velocity and 15.2 J/g kinetic energy are obtained.

If the isentropic expansion continues deep into the two phase region, phase separation will eventually occur, and some volume will be occupied by vapor. When the expansion reaches the spinodal, the specific

Fig. 3. P-V diagram produced by the PLOT4 command. The isochoric heating and isentropic expansion back to the saturated liquid line are shown for an inner HYLIFE jet.

Fig. 4. P-V diagram produced by the PLOT4 command. Isentropic expansion to the spinodal line is shown.

Table 1. Thermodynamic Values of the Inner HYLIFE Jets Before and After Heating

Point	T(°C)	$\rho\left(\frac{\text{kg}}{\text{m}^3}\right)$	$v\left(\frac{\text{m}^3}{\text{kg}}\right)$	P(MPa)	$E\left(\frac{\text{kJ}}{\text{kg}}\right)$	$A\left(\frac{\text{kJ}}{\text{kg}}\right)$	$S\left(\frac{\text{J}}{\text{kg K}}\right)$
Initial condition	500	484.364	0.00206456	0.000685	2349.56	-2988.00	6904.99
After heating	735.1	484.364	0.00206456	380.15	3149.46	-4723.10	7809.30
After expansion to coexistence curve	695.4	464.796	0.00215148	0.00337	3133.42	-4429.03	7809.22
After expansion to spinodal	372	305.705	0.00327113	-1537.5	4349.51	-686.68	7808.44

volume will be increased by about one-half. At a temperature of 500°C, the vapor pressure is $\sim 3 \times 10^{-3}$ torr, and the vapor specific volume is $\sim 2.34 \times 10^6 \text{ m}^3/\text{kg-vapor}$. Since the additional volume to be filled by vapor is $\sim 0.001 \text{ m}^3/\text{kg-liquid}$, 0.43 ng ($0.001/2.34 \times 10^6$) vapor must be vaporized for each gram of original liquid. Since vaporization requires $\sim 20.7 \text{ kJ/g}$ ($23.0 e_{\text{coh}} - 1.0 H_m - 1.3$ specific heat), only $\sim 10^{-5} \text{ J}$ are required for vaporization to fill the space due to the expansion of one gram of original liquid. Thus, energy used to vaporize lithium will be small, as will be the mass fraction of vapor in the expanded liquid.

References

1. D. A. Young, A Soft-Sphere Model for Liquid Metals, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-52352 (Nov. 1977).
2. W. G. Hoover, G. Stell, E. Goldmark, and G. D. Degani, "Generalized van der Waals Equation of State," J. Chem. Physics, **63**, No. 12, 5434 (Dec. 1975).
3. G. Peterson and D. Chato, Mathsy Manual, Lawrence Livermore National Laboratory, Livermore, CA, UCID-18875 (Oct. 1982).

Appendix A

```

mathsy macro eos                                updated Dec 15, 82
plots D. Young's soft sphere EOS for LI
legend! :aa is first term multiplier, :bb is second term multiplier,
:cc is third term multiplier, :cn is Cn, :dd is # density points,
:db is the debroglie wavelength (without T), :dl & :du
are SI density range, :ec is Ecoh, :el & :eu are the SI int. energy range,
:ep is epsilon, :er is epsilon/k, :fn is n/9, :ft is n/3, :mm is m,
:na is N, :nn is n, :pl & :pu are the MPa pressure range, :qq is Q,
:rp is R/A=k, :sg is sigma, :th is -1/3, :ss is the max dimension,
:tl & :tu are the Cent. temp range, :tm is the constant energy,
:tn is the constant entropy,
:tt is # of temp points. a is the critical vol (dim1), x & c are the
coexistence v & p points (dim17), p & v & t & e & f & s are the
pressure, volume, temperature, int. energy, helmholz free en. and entropy
arrays (dim:tt,:dd except for T).
---
"point prints the temperature in kelvins and centigrade, the density,
the volume, the pressure (MPa), the internal energy, the helmholz free
energy, and the entropy. Inputs are the density and
temperature (defaults 500 kg/m**3 and 500 C).
"point nosave dim4 save dim1 1 :tt=:dd=:ss=:g#or500 :dl=:du=:gc#or500 +273
:tl=:tu=:const pres intengy helm
format1(" temp(k) temp(c) density volume")
format3(" pressure internal energy helm energy entropy")
format2(4e16.8) wtty1 p :zp=:e :ze=:f :zf=:s :fs=:tl-273 :tc=
dim4 htty :tl,:tc,:dl,:vl wtty2,1,4 rtty:zp,:ze,:zf,:fs wtty3 wtty2,1,4 return
---
"plot1 plots p vs v, with t as a parameter, also plots coex curve & c.p.
Inputs for plot1 are the # of T & rho points, the lower & upper pressure
(MPa), lower and upper density (kg/m**3), & lower & upper temp (C)
plot1 defaults are 33,100, -1500,500, 20,500, 400,3600
"plot1 nosave g#or33 :tt=:gc#or100 :dd=:*:tt :ss=:dim:ss save dim1
gc#or-1500 :pl=:gc#or500 :pu=:gc#or20 :dl=
gc#or500 :du=:gc#or400 +273 :tl=:gc#or3600 +273 :tu=
const pres xaxv zaxt mapl,:vl,:vu,:pl,:pu,:tl,:tu front wtv1,...4,..02,h
wtv2,...02,..4,v build coex build crit return
---
"plot2 plots internal energy vs v, with t as a parameter
Inputs for plot2 are the # of T & rho points,
lower and upper density (kg/m**3), & lower & upper temp (C)
plot2 defaults are 33,100, 20,500, 400,3600
"plot2 nosave g#or33 :tt=:gc#or100 :dd=:*:tt :ss=:dim:ss save dim1
gc#or20 :dl=:gc#or500 :du=:gc#or400 +273 :tl=:gc#or3600 +273 :tu=
const pres xaxv zaxt intengy mapl,:vl,:vu,...:tl,:tu front wtv1,...4,..02,h
wtv3,...02,..4,v return
---
"plot3 plots p vs v with t as a parameter. It also indicates one
constant internal energy path. the coex curve & c.p. are also plotted.
plot3 inputs and defaults are as in plot1, the internal energy
is an added input with default 3.1495e6.
"plot3 nosave g#or33 :tt=:gc#or100 :dd=:*:tt :ss=:dim:ss save dim1
gc#or-1500 :pl=:gc#or500 :pu=:gc#or20 :dl=
gc#or500 :du=:gc#or400 +273 :tl=:gc#or3600 +273 :tu=:gc#or3.1495e6 :tm=
const pres xaxv zaxt mapl,:vl,:vu,:pl,:pu,:tl,:tu front wtv1,...4,..02,h
wtv2,...02,..4,v build d.chE intengy -:tm zeros -1 neg *1e6 +p
build coex build crit return
---
"plot4 plots p vs v with t as a parameter. It also indicates one
constant entropy path. the coex curve & c.p. are also plotted.
plot4 inputs and defaults are as in plot1. the entropy

```

Appendix A, continued

```

is an added input with default 7809.
"plot4 nosave g#or33 :tt= gc#or100 :dd= *:tt :ss= dim:ss save dim1
gc#or-1500 :pl= gc#or500 :pu= gc#or20 :dl= gc#or500 :du= gc#or400 +273 :tl
gc#or3600 +273 :tu= gc#or7809 :tn=
const pres xaxv zaxt mapl,:vl,:vu,:pl,:pu,:tl,:tu front wtv1,...4,.02,h
wtv2,...02,.4,v build d.ch0 intengy helm -:tn zeros -1 neg *1e6 +p
build coex build crit return
---
"const dim1 i2.669 :cn= 8.314/.000939 :rp= 23.03e6 :ec= mapp,1,.15,.95,.1,1
4 :nn= /3 :ft= 1.73075e9 :op= / :rp :er= 1.12316e-5 :sg= 1/3 neg :th= :nn/9 :fu=
.7 :mm= format1("Specific Volume, m**3/kg") format2("Pressure, MPa")
6.0225e23/.0006939 :na= format3("Internal Energy, J/kg") 1.3 :qq=
2*pi/:na*1.38066e-23*.5 inv *6.62618e-34 :db= return
---
"pres dim1 :er*:cn*:nn/3 :aa= :er*:th inv *:nn :bb= :nn+4*:bb*:qq/18 :bb=
:mm*:er :cc= :dl inv :vu= :du inv :vl= dim:dd,:tt span:tl,:tu trans t=
span:dl,:du inv v= dl t*:th p= :sg/v*:fn*p*:bb p= :sg/v*:mm/t*:cc neg +p p=
:sg/v*:ft/t*:aa+p+1*:rp*t/v/1e6 p= return
---
"coex dlchX dim17 rttY488.633,468.765,445.9,417.6,377.4,329.3,264.5,262.2,123.8
rtty9.2226,8.8321,1.9548,.322669,.02585,.0013125,.000027967,.0000001364 inv x=
xaxx rttY.11946,31.15,1834.7,45385,715854,5084330,24132000,25167300,100181000
rtty25167300,24132000,5084330,715854,45385,1834.7,31.15,.11946 /1e6 c= return
---
"crit dim1 rttY.0109991 a= xaxa rttY114.278 return
---
"intengy dim1 :er*:th inv :bb= :nn+4*:qq/6*:bb :bb= :er*:cn :aa=
dim:tt,:dd t*:th e= :sg/v*:fn*e*:bb e=
:sg/v*:mm*:er/t neg +e e= :sg/v*:ft/t*:aa+e+1.5*t*:rp+:ec e= return
---
"helm dim1 :db**3*:na :aa= 1 exp /:aa :aa= :er*:cn :bb= :er*:th inv :cc=
:nn/2+2*:qq*:cc :cc= dim:tt,:dd t*:th f= :sg/v*:fn*f*:cc f=
t**1.5*v*:aa ln neg +f f= :sg/v*:ft/t*:bb+f f= :sg/v*:mm*:er/t neg +f*t*:rp+:ec
f= e-f/t s= return

```

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