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Estimation of Surface Free Energies of Metals

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Abstract

The experimentally determined surface tensions of over twenty metals and twenty-five organic liquids have been compared with the predicted values by various equations. For liquid metals there is an excellent correlation between the molar surface tensions and the heats of vaporization, $\gamma_{\ell m} = 0.15\ \Delta H_{vap}$ while similar correlation does not hold for organic liquids. For solid metal surfaces the molar surface tensions can be correlated with the heats of sublimation, $\gamma_{sm} = 0.16\ \Delta H_{sub}$. These simple relationships have strong physical basis and can be utilized to estimate surface tensions to evaluate surface compositions for multicomponent systems, adhesion, or other thermodynamic parameters.

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Introduction

One of the fundamental thermodynamic parameters which characterize surfaces is the surface free energy per unit area, G^s, which is identical to the surface tension, γ, for one-component systems. It is the source of various other thermodynamic parameters, such as the surface entropy which is obtained from the temperature dependence of G^s. The specific surface free energy is also used to calculate the work of adhesion or wetting. The specific surface free energy is important in determining the surface composition of multi-component systems such as alloys and solid solutions. Utilizing Auger electron spectroscopy, the composition of the top-most layer can be readily determined to within 1% of a monolayer (about 10^{13} atoms/cm^2). As a result the surface composition may be determined by experiment as a function of the bulk composition for various systems. Since the solid or liquid multi-component system in equilibrium will have minimum surface free energy, equilibrium will be achieved by enrichment of the surface with the component that has the lowest surface free energy. Thus the surface composition of multi-component systems may be markedly different from the bulk composition.

Since the surface composition has an important effect on the surface reactions (gas-solid, liquid-solid), that may take place, it would be desirable to be able to determine the surface composition from the available thermodynamic data. The surface free energies of each component of the multi-component system are the thermodynamic functions that determine the surface composition. Their experimental determination for liquid metals, although far from being simple, can readily be carried out by contact angle or several other techniques that have been reviewed recently.[1] For solids, however, the measurement of the surface free energy is very difficult indeed. There are techniques to determine the ratio of the surface free energies of various crystal faces, however, using grain boundary grooving or other measurements.[2] It would thus be important to be able to estimate the surface free energy where such data are not available from experiments.

We have reviewed the surface free energies that were reported for over 20 liquid and solid metals and have found a useful correlation that permits estimation of the surface tension of metals to within 8% from the well known heats of vaporization and sublimation. This correlation can then be used to estimate unknown surface free energies and to predict the surface compositions of several solid solutions that are commonly utilized or to calculate other thermodynamic parameters of the studied systems.

The Surface Tension of Liquid Metals

The surface tension of many liquid metals has been measured over the past decade.[3,4,5] The surface tension was frequently determined as a

function of temperature in a finite temperature range. The surface tension of liquid metals decreases with increasing temperature, and of course it must vanish at the critical point, T_c. One equation used to describe this behavior is the Guggenheim equation[6] which is based on the corresponding states principle:

$$\gamma = \gamma_o \left(1 - \frac{T}{T_c}\right)^{1+R} \tag{1}$$

where γ_o, R, and T_c are adjustable parameters to give the best fit to the experimental data. R is usually taken as R = 2/9. Another equation frequently employed, especially to determine interfacial tension, assumes linear temperature dependence:

$$\gamma = \gamma_o - \left(\frac{\partial\gamma}{\partial T}\right)_P T \quad . \tag{2}$$

This equation derives from the expression of the Gibbs specific surface free energy, $G^s(T) = H^s - TS^s$, since for the one-component system $\gamma(T) = G^s(T), S^s = (d\gamma/\partial T)_P$ and $\gamma_o = H^s_o$. For the case when H^s and S^s are independent of temperature, it is possible to obtain both functions from the temperature dependence of γ . The intercept of the straight line (γ vs. T) at absolute zero, γ_o, yields a specific surface enthalpy, and the slope $(d\gamma/dT)_P$, is the specific surface entropy.

The first three columns of Table I show the values of the parameters of the Guggenheim equation (γ_o, T_c, 1+R) that give the best least squares fit to the experimental surface tension values for 15 different metals.

The last three columns of Table I give the values of the parameters γ_o and $\left(\frac{\partial\gamma}{\partial T}\right)_P$ in equation 2 that give the best fit to the same data. The references identify sources of experimental data. The γ_o values that were determined from fitting the experimental values to both equations are almost identical. Thus, one has no reason to prefer one equation over the other on this basis. The two equations give different results in their prediction of the critical temperature. This is due to the different functional dependence of γ on T and the long extrapolation required to reach the point where the surface tension is zero. The linear and Guggenheim relations are equally accurate in the region where data are available, but they begin to deviate at about 1000-3000°K, depending on the metal.

Although the temperature dependence of the surface tensions of liquid metals are certainly not negligible, inspection of the $\left(\frac{\partial\gamma}{\partial T}\right)_P$ values listed in Table I reveals that most surface tensions change by no more than 5% in a 100° temperature interval. This variation is not greater than the uncertainty of most surface tension experiments. Thus, the surface tension may be taken as constant in most cases, as long as the temperature range of experimental interest is limited.

There are anomalies reported in measurements of the temperature dependence of γ that indicate deviations from the straight line γ vs. T behavior. A change of slope of the γ vs. T curve indicates a change of surface entropy that may signify ordering on the surface. The consideration of these anomalies is outside the scope of this paper.

Correlation Between Surface Tension of Liquid Metals and Their Heats of Vaporization

The specific surface free energy for an unstrained phase is equal to the increase of the total free energy of the system per unit increase of the surface area: $G^s = (\partial G/\partial A)_{T,P}$. Thus, creation of more surface always increases the total free energy of the system. Since atomic bonds must be broken to create surfaces, it is expected that the specific surface free energy be related to the heat of vaporization, which reflects the energy input necessary to break all the bonds of atoms in the condensed phase. The heat of vaporization is a molar quantity (energy/g-atom), while the specific surface free energy is defined as energy per unit area (energy/cm^2). In order to compare the two values, we must convert the specific surface free energy to molar surface free energy (energy/g-atom).[7] Because of differences in the densities of various materials, they will have differing numbers of atoms occupying a unit area. Let us define an area, A, as the area occupied by Avogadro's number of atoms, N . The atomic volume V_a is given by

$$V_a = \frac{V_m}{N} = \frac{M}{N\rho} \tag{3}$$

where V_m is the molar volume, ρ is the density, and M the atomic weight. Thus the area per atom A_a is given by[7]

$$A_a = f(V_a)^{2/3} = f\left(\frac{M}{N\rho}\right)^{2/3} \tag{4}$$

where f is a structure factor that corrects for the assumption that the surface is the (100) face of a simple cubic lattice (this was implicitly assumed in using $V^{2/3}$ as the surface area).* The value of f is 1.09 for melts of fcc solids, 1.12 for melts of bcc solids, and 1.14 for molten Bi, Sn, and Sb (orthorhombic in the solid state).[7] The molar surface area is then given by

$$A = N A_a = fN^{1/3}\left(\frac{M}{\rho}\right)^{2/3} \tag{5}$$

and the molar surface tension or molar surface free energy of the liquid is defined as

$$\gamma_{\ell m}(T) = A\gamma_{\ell}(T) \tag{6}$$

Now we can proceed to compare $\gamma_{\ell m}$ directly to the heat of vaporization since both quantities are known from experiments for the 22 liquid metals that are listed in Table II. The plot of $\gamma_{\ell m}$ at the melting point for each metal vs. their heats of vaporization is shown in Fig. 1. A least-squares fit yields the relationship

$$\gamma_{\ell m} = 0.15\ \Delta H_{vap} \tag{7}$$

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* Following McLachan,[8] the area of an atom is expected to be proportional to the square of some dimension of the atom and the volume proportional to its cube: $A = bD^2$, $V = cD^3$. Thus $A = \frac{b}{c^{2/3}}\ V^{2/3}$. For the (100) face of a simple cubic structure b = c = 1, so f = 1 as expected. For the (111) face of an fcc metal, $b = \sqrt{3}/4$ and c = 1/4, which yields f = 1.09.

All of the experimental data fit this equation with a standard deviation of 8%.

The $\gamma_{\ell m}$ vs. ΔH_{vap} correlation that is expressed in Eq.(7) that appears to hold so well for liquid metals does not hold for organic liquids of various types. In Fig. 2, surface tensions of various organic liquids at the melting point are plotted against their heats of vaporization, assuming f = 1. As can be seen the data are widely scattered, reflecting the diversity and complex bonding characteristics and packing of these liquids. The first three columns of Table III give the values of the parameters γ_o, T_c and 1+R of the Guggenheim equation that give the best least-squares fit to the experimental surface tension values for 25 organic liquids. The last three columns of Table III show the values of the parameters of equation 2 (γ_o and $\left(\frac{d\gamma}{dT}\right)_P$) that give the best fit to the same data.

Correlation Between the Surface Tension of Solids and Their Heats of Sublimation

For monatomic solids surface tension determination is more difficult and the available experimental data are scarce and often determined only at one temperature.[2] Nevertheless, we have collected most of the available data, which are tabulated in Table IV. In Fig. 3 the molar surface tensions of the solids, γ_{sm}, are plotted against the heats of sublimation, ΔH_{sub}, for various metals. A least-squares fit yields the relationship

$$\gamma_{sm} = 0.16\ \Delta H_{sub}\ . \tag{8}$$

The temperature dependence of γ_{sm} was disregarded in the correlation as discussed earlier. The validity of this approximation can be seen by examining the data for copper and nickel: the correction for the temperature dependence is well within the experimental error. There is excellent agreement between the experimental values and those calculable from Eq.(8), and the standard deviation is 8%. Thus it appears that at least for monatomic solids the surface tension may be estimated when direct experimental determination is difficult or lacking.

Discussion

The surface free energy is defined as the increase of the total free energy of the system per unit increase of the surface area. For metals, the creation of more surface requires the breaking of chemical bonds which is accompanied by charge redistribution of the electron gas. Theoretical computations of the surface tension of metals have been performed by considering these contributions separately.

The model which takes into account only the breaking of chemical bonds correlates the surface tension with the heat of vaporization or heat of sublimation. Skapski[7] and McLachan[8] considered the breaking of only the nearest neighbor bonds in the condensed phase. For a close-packed plane of a solid[9] an atom in the surface has nine bonds to the interior of the solid; thus, the heat of sublimation corresponds to the energy necessary to break 18 half-bonds. The surface free energy is approximately equal to the energy of breaking the bonds by transferring

a bulk atom to the surface; since this is a close-packed solid (12 nearest neighbors in total), there will be 3 half-bonds per atom directed out of the plane at the interface. Thus the ratio of γ_{sm} to ΔH_{sub} should be 3:18 or 1/6, which is approximately the same as the empirically determined value that is given in Eq.(8). Such a simple model does not explain the surface tension of organic substances as these simple assumptions are no longer valid. More detailed calculations should take into account longer range interactions, relaxation of the newly created surface atoms into their new equilibrium positions, and the excess of binding energy the surface atoms may have as compared to those in the bulk due to the availability of surplus bonding orbitals.[10] Such a model, when developed, would include both the bond breaking and the charge redistribution that take place on creating new surfaces.

The simplest version of the free electron gas model used to calculate the charge redistribution that takes place at the freshly made surface is the particle in a box, with the surfaces of the metal corresponding to the walls of the box, which contains a uniform density of electrons.[11] This model was improved by various workers,[12,13] but until Hohenburg, Kohn and Sham [14,15] devised a more general formalism which can treat inhomogeneous electron distributions, the change of electron density at the surface of a metal was ignored. Using this new model Lang and Kohn[16] predicted metal surface free energies within about 25% of the experimental values. More recently Schmidt and Lucas[17,18] proposed that the surface free energies of metals are mainly due to the change in plasmon density caused by the introduction of a new surface. Their computed surface

free energies fall within 30% of the measured values and there is no attempt to fit the experimental data.

These different types of calculations of the surface tensions of metals provide the physical basis of the observed correlation between surface tension and the heat of vaporization or heat of sublimation. It appears that Eqs.(7) and (8) can be used with confidence to estimate surface tensions and utilize them in evaluating many important properties of surfaces, their composition, adhesion or other surface thermodynamic parameters.

Acknowledgements

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Table I.

Values of the parameters for equations 1 and 2 that give the best least squares fit to the experimental surface tension data for several liquid metals.

Metal	Corresponding States			Linear		
	$\gamma_o\left(\frac{ergs}{cm^2}\right)$	T_c °K	1+R	$\gamma_o\left(\frac{ergs}{cm^2}\right)$	$\frac{\partial\gamma}{\partial T}$	T_c °K
Al[a,b]	943.85	14481	1.22	943.17	0.0782	12061
Sb[a,b]	403.17	12834	1.22	402.78	0.0375	10741
Bi[a,b]	416.44	7719	1.25	415.54	0.0656	6334
Cd[a,b]	700.54	6736	1.22	699.35	0.1238	5649
Cs[a,c,d]	86.78	1958	1.19	85.57	0.0491	2087
Cu[a,b]	1291.81	63540	1.21	1291.46	0.0244	52927
Pb[a,b]	489.06	9106	1.27	488.15	0.0663	7363
Li[c]	483.27	3316	1.23	475.88	0.1640	2902
Mg[a,b]	656.34	6350	1.22	654.09	0.1217	5375
K[c,d]	144.06	3126	1.03	138.52	0.0739	1898
Rb[c,d]	105.52	2185	1.33	103.56	0.0573	1807
Ag[a,b,e]	1277.11	5333	1.22	1262.43	0.2729	4626
Na[a,b,c]	252.51	2452	1.22	249.73	0.1161	2151
Sn[a,b,e]	578.30	11876	1.22	577.93	0.0585	9879
Zn[a,b]	832.74	9703	1.25	830.89	0.1037	8012

[a] Handbook of Chemistry and Physics, 53rd edition, Chemical Rubber Publishing Co., Cleveland (1972).

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[e] International Critical Tables, McGraw-Hill Book Co., Inc., New York (1928).

Table II

The densities, molar surface areas, heats of vaporization, and calculated and experimental values of γ_ℓ for several liquid metals.

Metal	ρ [a] $\frac{gm}{cc}$	$A\left(\frac{cm^2}{mole}\right)$	ΔH_{vap} [a] $\left(\frac{ergs}{mole}\right)$	$\gamma_\ell(calc)\frac{ergs}{cm^2}$	$\gamma_\ell(exp)\frac{ergs}{cm^2}$ this paper	others
Al	2.29	4.61×10^8	2.90×10^{12}	937	870	914[b], 825[a], 860[a], 865[a]
Sb	6.13	6.67×10^8	2.00×10^{12}	447	369	383[a]
Bi	9.15	7.13×10^8	1.75×10^{12}	366	380	378[a], 376[c]
Cs	1.57	16.8×10^8	0.69×10^{12}	61	58	60[a]
Cu	7.70	3.68×10^8	3.11×10^{12}	1259	1258	1270[a], 1300[a], 1220[d], 1350[d]
Ir	20.0	4.16×10^8	5.86×10^{12}	2113		2250[d]
Pb	6.80	6.63×10^8	1.82×10^{12}	409	448	451[a]
Li	10.16	5.3×10^8	1.39×10^{12}	391	400	
Mo	9.33	4.44×10^8	6.16×10^{12}	2081		2250[d], 2080[d]
Ni	7.8	3.54×10^8	3.79×10^{12}	1606		1780[d], 1725[d], 1720[d]
Nb	7.83	4.95×10^8	6.99×10^{12}	2118		1900[d]
Pd	10.7	4.26×10^8	3.74×10^{12}	1317		1500[d]
Pt	19.7	4.25×10^8	5.12×10^{12}	1807		1800[d], 1699[d], 1865[d], 1740[d]
K	0.72	12.4×10^8	0.81×10^{12}	97	114	114[e]
Rh	11	4.08×10^8	5.33×10^{12}	1960		2000[d]
Rb	1.45	14.1×10^8	0.78×10^{12}	82	86	76[a], 92[e]
Ag	9.00	4.74×10^8	2.59×10^{12}	814	926	785[a], 930[f]
Na	0.74	7.77×10^8	1.00×10^{12}	192	207	206[c], 220[f], 191[a]
Ta	15.0	4.97×10^8	7.34×10^{12}	2215		2150[d], 2360[d], 2020[d]
Sn	6.29	6.24×10^8	2.35×10^{12}	561	549	526[a], 550[b]
W	17.6	4.53×10^8	7.84×10^{12}	2596		2500[d]
V	5.55	4.15×10^8	4.82×10^{12}	1742		1950[d]

[a] Handbook of Chemistry and Physics, 53rd edition, Chemical Rubber Publishing Co., Cleveland (1972).

[b] V. K. Sementchenko, Surface Phenomena in Metals and Alloys, Pergamon Press, New York (1962).

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Table III.

Values of parameters for equations 1 and 2 that give the best least-squares fit to the experimental surface tension data for several organic liquids.

Compound[a,b]	Corresponding States			Linear		
	$\gamma_o\left(\frac{ergs}{cm^2}\right)$	T_c °K	1+R	$\gamma_o\left(\frac{ergs}{cm^2}\right)$	$\frac{\partial\gamma}{\partial T}$	T_c °K
C_2H_2	73.38	287	1.22	64.64	0.2380	272
CH_3OCH_3	61.81	397	1.22	56.64	0.1530	370
CH_2CH_2O	76.86	480	1.22	72.54	0.1646	441
$(CH_3)_2NH$	44.52	522	1.22	42.87	0.0910	471
$(CH_3)_3N$	48.37	472	1.22	46.13	0.1074	430
$C_2H_5NH_2$	51.86	563	1.34	49.02	0.1019	481
CO	29.57	131	1.75	27.22	0.2266	120
$CHCl_3$	72.00	523	1.18	67.47	0.1373	491
HCN	59.97	466	1.22	54.95	0.1266	434
HCOOH	71.91	711	1.22	69.17	0.1077	642
CH_3COOH	58.17	593	1.08	55.90	0.0962	581
C_2H_5COOH	57.24	631	1.22	53.75	0.0925	581
CH_3CHO	64.67	467	1.13	61.22	0.1366	448
CH_3COCH_3	63.99	507	1.15	60.46	0.1254	482
$C_2H_6O_2$	77.35	889	1.21	75.55	0.0952	794
$HCOOC_2H_5$	66.03	508	1.20	57.67	0.1168	494
CH_3COOCH_3	70.71	507	1.22	60.48	0.1230	492
$n\text{-}C_6H_{14}$	51.97	494	1.15	49.02	0.1041	470
$n\text{-}C_8H_{18}$	53.75	561	1.22	50.69	0.0985	515
C_5H_5N	86.40	598	1.22	81.17	0.1476	550
$C_6H_5NO_2$	82.60	775	1.33	77.23	0.1140	678
C_6H_5OH	79.70	704	1.11	72.75	0.1086	670
$C_6H_5NH_2$	80.38	729	1.22	76.68	0.1153	665
C_6H_5CHO	76.80	709	1.22	74.22	0.1167	636
$C_6H_5CH_3$	62.21	606	1.21	58.65	0.1048	560

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(Table III continued)

[a] Handbook of Chemistry and Physics, 53rd edition, Chemical Rubber Publishing Co., Cleveland (1972).

[b] International Critical Tables, McGraw-Hill Book Co., Inc., New York, (1928).

Table IV.

The densities, molar surface areas, heats of sublimation, and calculated and experimental values of γ_s for several metallic solids.

Metal	$\rho^a\left(\frac{gm}{cc}\right)$	$A\left(\frac{cm^2}{mole}\right)$	$\Delta H_{sub}{}^b\left(\frac{ergs}{mole}\right)$	γ_s(calc) $\frac{ergs}{cm^2}$	γ_s(exp) $\frac{ergs}{cm^2}$	T°K of γ_s(exp)
Al	2.70	4.24×10^8	3.14×10^{12}	1198	1140±200[c]	450
Cu	8.96	3.7×10^8	3.39×10^{12}	1484	1670[d]	1320
					1710[e]	1273
					1750[f]	1173
Au	19.3	4.44×10^8	3.68×10^{12}	1343	1400±65[g]	1315, 1290
					1370±150[h]	1313
					1410[d]	1300
Ni	8.90	3.22×10^8	3.39×10^{12}	1706	1850[d]	1523
					1860±200[i]	1493
Nb	8.60	4.56×10^8	7.20×10^{12}	2557	2100±100[d,j]	2523
					2550±550[k]	1773
Pt	21.45	4.06×10^8	5.56×10^{12}	2219	2300±800[ℓ]	1310
					2340[d]	1311
Ag	10.5	4.34×10^8	2.85×10^{11}	1064	1140±90[m,d]	1203, 1180
Ta	16.6	4.64×10^8	7.78×10^{12}	2713	2680±500[k]	1773
Sn	5.76	6.89×10^8	2.30×10^{11}	541	600±75[n]	488
Ti	4.5	4.47×10^8	4.73×10^{12}	1712	1700[o]	1873

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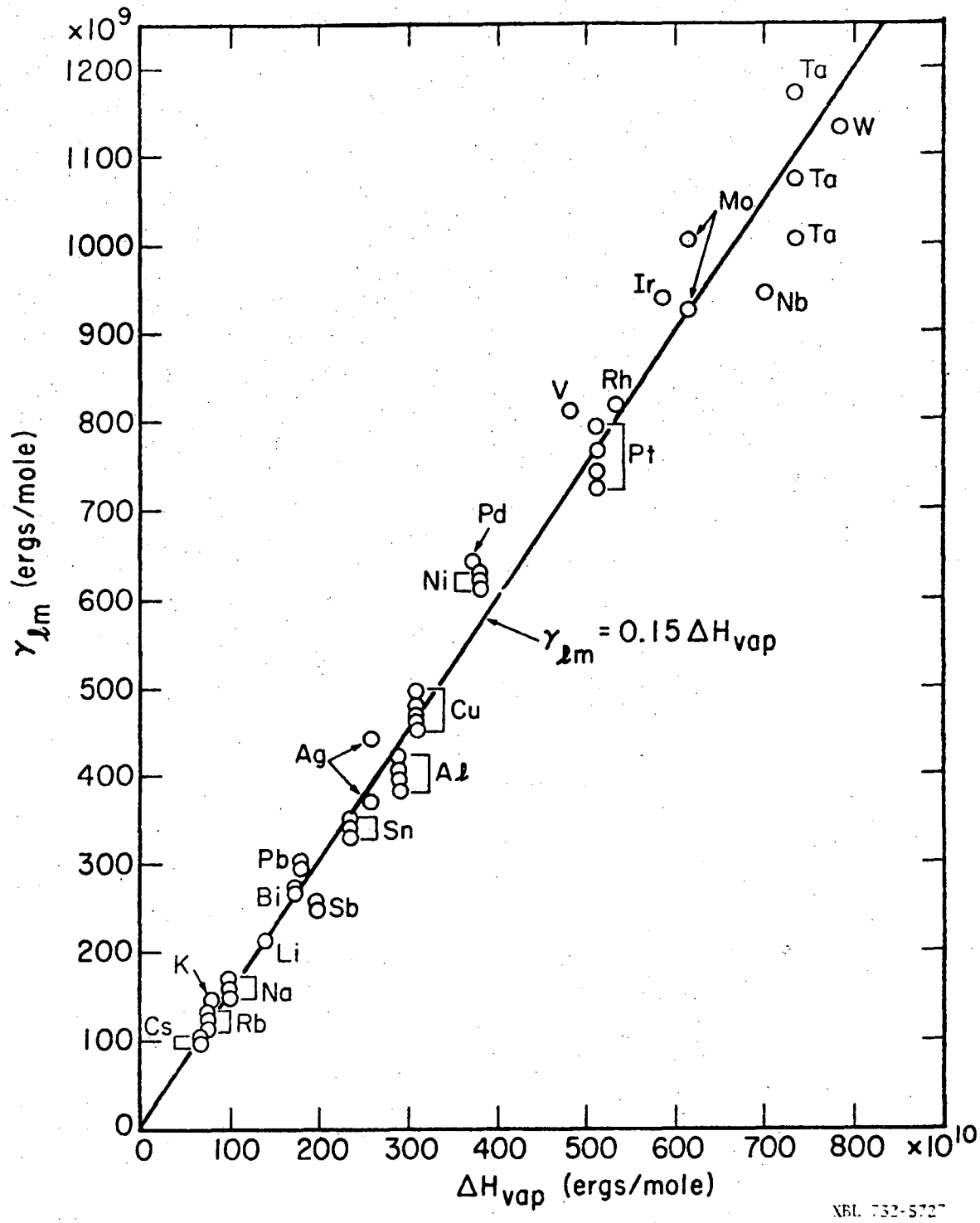
Figure Captions

Figure 1 Molar surface tensions of liquid metals, $\gamma_{\ell m}$, as a function of their heats of vaporization.

Figure 2 Molar surface tensions of organic liquids, $\gamma_{\ell m}$, (assuming f = 1) as a function of their heats of vaporization.

Figure 3 Molar surface tensions of several solid metals as a function of their heats of sublimation.


×10^9
1200
1100
1000
900
800
700
600
500
400
300
200
100
0
γℓm (ergs/mole)
0
100
200
300
400
500
600
700
800
×10^10
ΔHvap (ergs/mole)
Ta
W
Ta
Mo
Ta
Ir
Nb
V
Rh
Pt
Pd
Ni
γℓm = 0.15 ΔHvap
Cu
Ag
Aℓ
Sn
Pb
Bi
Sb
Li
K
Na
Rb
Cs

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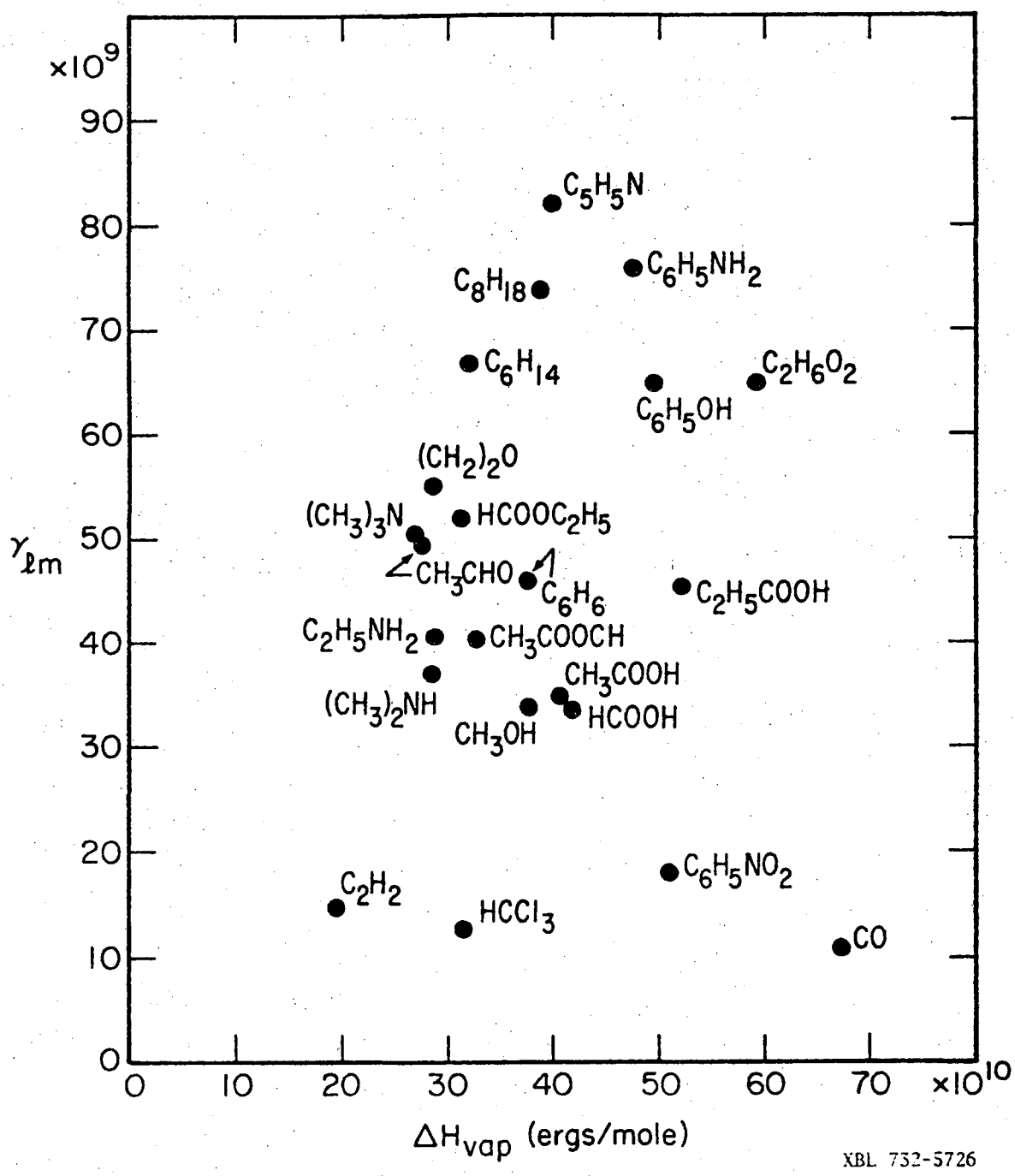
×10^9
90
80
70
60
50
40
30
20
10
0
γℓm
C5H5N
C6H5NH2
C8H18
C6H14
C2H6O2
C6H5OH
(CH2)2O
(CH3)3N
HCOOC2H5
CH3CHO
C6H6
C2H5COOH
C2H5NH2
CH3COOCH
CH3COOH
(CH3)2NH
HCOOH
CH3OH
C6H5NO2
C2H2
HCCl3
CO
0
10
20
30
40
50
60
70
×10^10
ΔHvap (ergs/mole)

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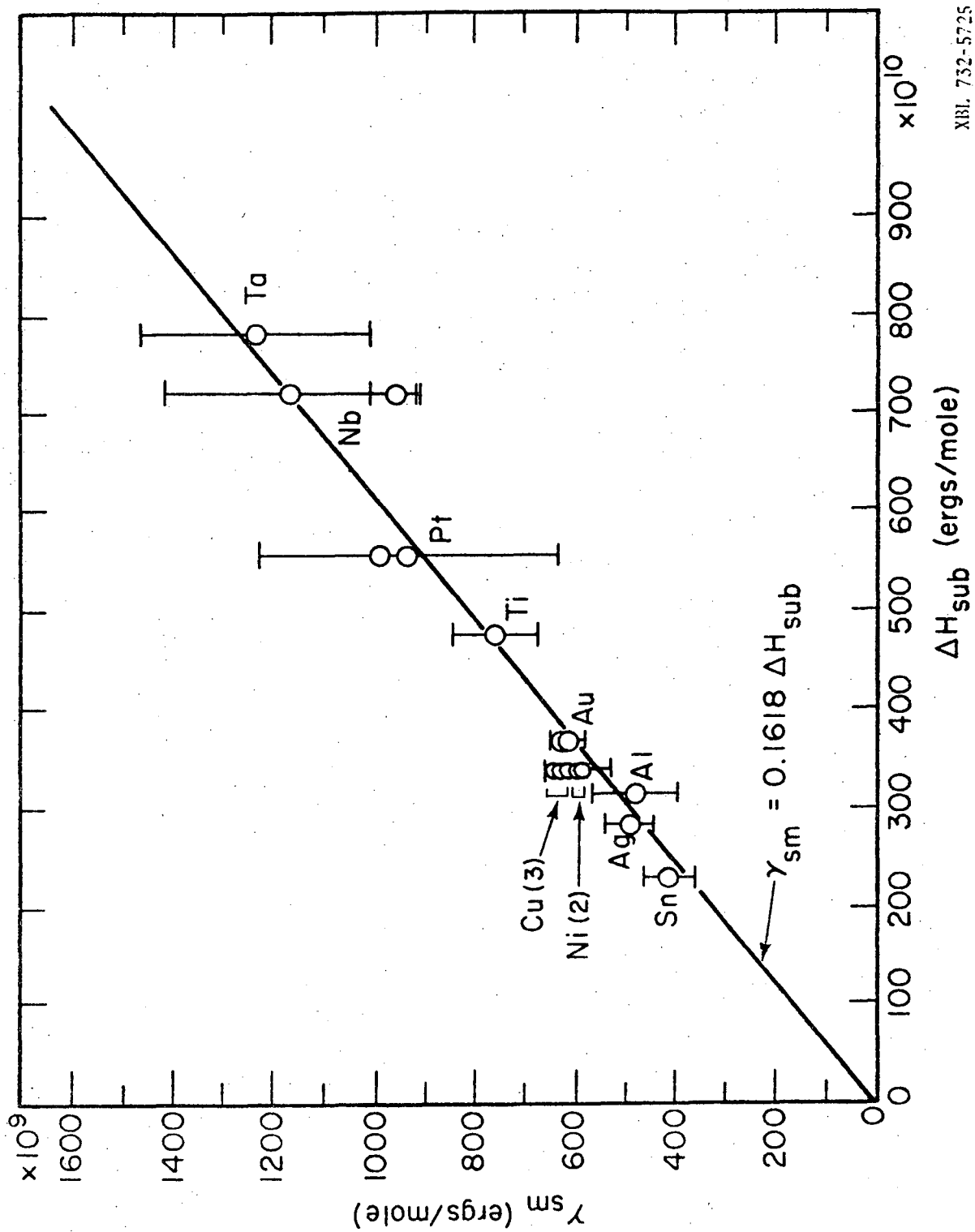
0 0 0 0 3 9 0 2 2 6 9



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