

# The Solubility of Hydrogen in Molten Aluminum Alloys

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The solubility of hydrogen in molten aluminum alloys containing copper, lithium, magnesium, and silicon has been calculated from the solubility of hydrogen in pure metals and binary metal-metal interaction parameters. For the aluminum-copper binary system, where experimental data exist, the agreement between calculated and experimental values is excellent. The solubility of hydrogen in liquid silicon was calculated from the solubility data in aluminum-silicon alloys.

## I. INTRODUCTION

**H**YDROGEN in molten aluminum and aluminum alloys is the main cause of pinhole porosity in solidified aluminum ingots and casting.<sup>[1]</sup> It is the only gas having a measurable solubility in aluminum. The main source of hydrogen comes through the reaction of aluminum with moisture in the melting environment. During solidification, hydrogen bubbles nucleate and grow, resulting from the fact that for aluminum and its alloys, the solubility of hydrogen in solids is normally much smaller than that in the molten state.

While there have been a number of studies to determine the hydrogen solubility in pure aluminum, very few data are available on the solubility of hydrogen in aluminum alloys. In this paper, the solubility of hydrogen in aluminum-based alloys containing copper, lithium, magnesium, and silicon is calculated.

## II. METHOD OF CALCULATION

Recently, Hoch and Arpshofen<sup>[2,3]</sup> developed a thermochemical model to determine the solution behavior of binary, ternary, and larger component systems. This model appears to be suitable in predicting the thermodynamic properties of a ternary solution from the corresponding binary systems. The derivation of this solution model for a multi-component system is summarized in the following.

Let a solution contain components *A, B, C, D* etc. with the corresponding concentrations in mole fractions being *x, y, z, u, etc.* The contribution of the A-B binary system to the multicomponent solution is<sup>[3]</sup>

$$H^M = Wnx[1 - (1 - y)^{(n-1)}] \quad [1]$$

$$H_A^M = Wnx[1 - (1 - y)^{(n-1)} - xy(n - 1)(1 - y)^{(n-2)}] \quad [2]$$

$$H_B^M = Wnx(n - 1)(1 - y)^{(n-1)} \quad [3]$$

$$H_C^M = H_D^M = -Wnxy(n - 1)(1 - y)^{(n-2)} \quad [4]$$

where  $H^M$  is the enthalpy of mixing,  $H_i^M$ 's are the partial quantities of *i*'s,  $W$  is the interaction parameter, and  $n$  is an integer (2, 3, 4, etc.). The values of  $n$  and  $W$  are deduced from the binary system. If one makes

$x + y = 1$ , the following equations for the binary system<sup>[2]</sup> are obtained:

$$H^M = Wn(x - x^n) \quad [5]$$

$$H_A^M = Wn[1 - nx^{(n-1)} + (n - 1)x^n] \quad [6]$$

$$H_B^M = Wnx(n - 1)x^n \quad [7]$$

The quantity  $x$  is the mole fraction of the component, so that the maximum of  $H^M$  (either positive or negative) is at  $x > 0.5$ . The quantity  $n$  is chosen such that  $W$ , determined from thermodynamic data, is independent of composition. For the detailed derivation of Eqs. [1] through [7], one is encouraged to refer to original papers by Hoch and Arpshofen.<sup>[2,3]</sup> From each experimental point, one value of  $W$  is obtained, and all of these can be averaged with a standard deviation obtained through the regression analysis.

$S^{ex}$ , the excess entropy of mixing, has the same form as Eq. [5]. It can have another value for  $n$ , and its maximum may be at  $x < 0.5$ . Therefore, it is possible that

$$S^{ex} = rU(y - y') \quad [8]$$

where  $U$  and  $r$  are constants which can be determined similarly to  $W$  and  $n$ .  $G^{ex}$ , the excess Gibbs energy of mixing, which is a combination of  $H^M$  and  $S^{ex}$ , will have a simple form only if  $S^{ex}$  has the same form as  $H^M$  or is zero.

The partial quantities represented by Eqs. [3] and [4] do not change signs when the composition changes from  $x = 0$  to  $x = 1$ . The signs of  $H_A^M$  and  $H_B^M$  are determined by the sign of  $W$ . In other words, one interaction parameter  $W$  describes one type of interaction or reaction in a system. If two types of solution behavior exist in a binary system, two interaction parameters, an attractive  $W$  and a repulsive  $M$ , are needed. Each has its major effect at different composition ranges with different dependencies on composition ( $n$  and  $x$  in one case,  $m$  and  $y$  in the other). Thus,

$$H^M = nW(x - x^n) + mM(y - y^m) \quad [9]$$

$W$  and  $M$  are determined by least-squares analysis with confidence limit and error in  $W$  and  $M$  also calculated. The values of  $n$  and  $m$  are varied until the confidence limit is high and the error limits on  $W$  and  $M$  reach the smallest ratio of error to value. A relationship seems to exist between  $n$  and  $m$ :  $n = 2m$  or  $m = 2n$ .

The ideal Gibbs energy of mixing is always

$$G_{id}^M = RT(x \ln x + y \ln y + z \ln z + \dots) \quad [10]$$

The other binary systems (A-C, A-D, B-C, etc.) contribute similarly to the thermodynamic properties of the solution.

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The advantage of the present solution model is that in a binary system, a maximum of only four interaction parameters is needed: two for  $H^M$ , the enthalpy of mixing, and two for  $S^{ex}$ , the excess entropy of mixing. No ternary or larger system has yet been found in which ternary or larger parameters are needed.

In the present treatment of binary systems, Hultgren *et al.*'s<sup>[4]</sup> evaluations of the original literature were used. All thermodynamic data are divided by  $R$ , the gas constant.  $H^M$  (the enthalpy of mixing) and  $G^M$  (the Gibbs energy of mixing) are, therefore, expressed in kK (kiloKelvin), and  $S$  (the entropy) and  $C_p$  (the heat capacity) are dimensionless.

The following nomenclature is adopted in this work. If  $W$  was derived from enthalpies of mixing, it is designated by  $W_h$ . If two interaction parameters are needed to describe  $H^M$  in a system, the second value is designated by  $M_h$ . The interaction parameter derived from the excess entropy of mixing is denoted as  $W_s$ . In rare cases where  $S^{ex}$  has the same form as  $G^{ex}$  or is 0,  $W_g$  is used for  $G^{ex}$  (the excess Gibbs free energy). The notation 4,(Cu) indicates that in that system,  $n = 4$  and  $x$  represents the mole fraction of copper.

In all the treatments of binary data, an average value and a standard deviation ( $\pm$ ) are given for each constant calculated. For linear correlations, the least-square analysis is used to determine the slopes and the intercepts; an error ( $\pm$ ) is given for each, plus the line fitting correlation coefficient  $R$ . In addition to these uncertainties, caused by fitting the model to the experimental data, an additional uncertainty exists for the  $\pm$  value given by Hultgren *et al.* where their values are used. This value is converted to this study at  $x = 0.5$  and is given in the last column of Table I. This uncertainty, which is generally larger than that caused by fitting the model, is added to the first value (generally,  $W_h$ ) by  $c = (a^2 + b^2)^{1/2}$  and introduced into the tables describing ternary systems.

Enthalpies of mixing were treated according to Eqs. [5] through [7]. To obtain the excess entropy of mixing ( $S^{ex}$ ), two procedures were applied. First,  $W_h$  was calculated from  $H^M$ . With this value and  $G^{ex}$ ,  $TS^{ex}$  was calculated at every composition:

$$TS^{ex} = W_h(n, x) - G^{ex} \quad [11]$$

From this value,  $W_s$  is obtained using Eqs. [5] through [7]. The other method, which was also applied at every composition, splits  $G^{ex}$  into its components by least-square analysis:

$$G^{ex} = W_h(n, x) - TW_s(r, y) \quad [12]$$

The values  $n$  and  $r$  are varied until the errors in  $W_h$  and  $W_s$  are minimum and the correlation coefficient  $R$  is

maximum. Obviously, if  $S^{ex}$  has the same shape as  $H^M$ , this method is not applicable. In binary systems,  $W_g$  was not calculated, but when calculating ternary systems,  $W_g$  can be obtained from  $-TW_s = W_g$ .

### III. RESULTS

The binary metal-metal interaction parameters were calculated from the thermodynamic data given by Hultgren *et al.*<sup>[4]</sup> and Barin and Knacke<sup>[5]</sup> if available. In other cases, and the hydrogen-metal interaction, parameters (Henry's law constants) were evaluated from the original data. The Al-Cu and Al-Mg systems are treated similarly to the Al-Li system, and only the results are given in Table I.

#### A. Al-Li System

McAlister<sup>[6]</sup> evaluated the available data in this system. However, calculations applying Hoch-Arpschhofen models are needed to obtain an equation for the Li-Al interaction parameter which can be extrapolated into ternary and larger systems. For the interaction parameter, the regular solution model is sufficient: in the present notation,<sup>[2]</sup> 2,(Al) or 2,(Li) for  $W_h$  and  $W_s$ .

Figure 1 plots the interaction parameter  $W_h$  in the Al-Li melts as a function of the composition as calculated from data of Moser<sup>[7]</sup> and Bushmanov and Yatsenko.<sup>[8]</sup> Figure 2 shows the calculated  $W_g$  in the Al-Li melts as a function of the composition from data by Yatsenko and Saltykova<sup>[9]</sup> and Hicter *et al.*<sup>[10]</sup> The data of Yatsenko

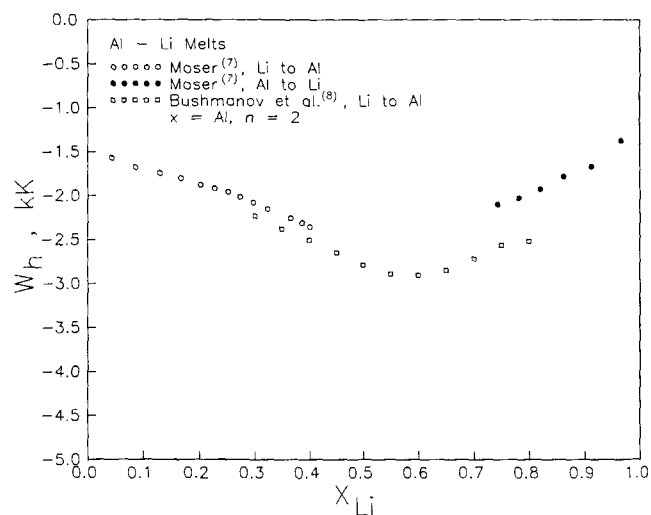


Fig. 1—Interaction parameter,  $W_h$ , in the Al-Li melt as a function of the melt composition.

Table I. Metal-Metal Interaction Parameters

System	$W_h$ , kK	$W_s$ , kK
Al-Cu	4 (Cu) $-0.6868 \pm 0.1072$	2 (Al) $0.5538 \pm 0.00518$
Al-Mg	4 (Al) $-0.1881 \pm 0.0788$	2 (Mg) $-0.0357 \pm 0.0048$
Al-Si	3 (Al) $-0.2099 \pm 0.0837$	3 (Al) $0.3221 \pm 0.0653$
Cu-Li	3 (Li) $0.4888 \pm 0.0787$	—
Cu-Mg	3 (Cu) $-0.7496 \pm 0.0308$	3 (Mg) $-0.0233 \pm 0.0023$
Al-Li	2 (Al) $-2.2830 \pm 0.2910$	2 (Al) $-1.1750 \pm 0.2680$

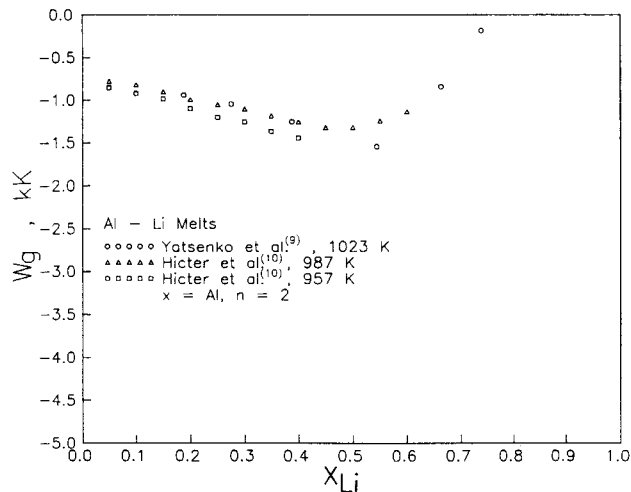


Fig. 2—Interaction parameter  $W_g$ , in the Al-Li melt as a function of the melt composition.

and Saltykova<sup>[9]</sup> had to be read off a small graph. From the Li activity data,  $W_g$  is calculated at each temperature. Table II summarizes the five calculated interaction parameter values (two  $W_h$ , three  $W_g$ ) from individual papers. From these data, the following equation of  $W_g$  as a function of temperature is obtained:

$$W_g = (-2.283 \pm 0.291) + (1.175 \pm 0.268) T \quad \text{in kK} \quad [13]$$

The enthalpy and entropy of formation of solid AlLi (taken as a line compound) from solid aluminum and lithium (Table III) were calculated using the enthalpy and entropy of fusion of the elements given by Kubaschewski and Alcock<sup>[11]</sup> at the three invariant points where AlLi(s) is in equilibrium with liquid (973 K,  $x_{Li} = 0.5$ ; 873 K,  $x_{Li} = 0.14$ ; 793 K,  $x_{Li} = 0.77$ ) and compared to the values of Wen *et al.*,<sup>[12]</sup> Yao *et al.*,<sup>[13]</sup> and Sigli and Sanchez.<sup>[14]</sup> The agreement is very good.

### B. Al-H System

The solubility of hydrogen in aluminum has been measured by a number of research groups. Most of them applied Sieverts' method in their measurements. In this technique, the volume change of hydrogen gas above aluminum due to the hydrogen dissolution is directly detected. Data obtained by Bauklok and Oesterlen,<sup>[16]</sup> Ransley and Neufeld,<sup>[17]</sup> and Opie and Grant<sup>[18]</sup> were relatively newer than others and agree with each other. Ransley and Talbot<sup>[19]</sup> had verified the accuracy of data obtained by Ransley and Neufeld with the hot extraction method.<sup>[17]</sup> Most of the data in earlier investigations were

Table III. Enthalpy and Entropy of Formation of AlLi(s)

	Enthalpy of Formation		Entropy of Formation	
	kcal/g-at	±	cal/g-at * K	±
This research	-5.044	0.029	-2.152	0.225
Wen <i>et al.</i> <sup>[12]</sup>	-5.170	—	-2.460	—
Yao <i>et al.</i> <sup>[13]</sup>	-5.200	—	-2.540	—
Sigli and Sanchez <sup>[14]</sup>	-5.160	—	-2.540	—

in considerable divergence due to experimental difficulty. In this study, results from these earlier studies were not examined further. Meanwhile, more recently, Eichenauer *et al.*<sup>[20,21]</sup> had determined the solubility of hydrogen in both solid and liquid aluminum through a degassing process. Their data were considerably lower than those of the others. However, they have admitted in their second paper<sup>[21]</sup> that data in the first paper<sup>[20]</sup> were too low. Since there are no data in the liquid aluminum region in their second paper, we decided to omit their data in this study.

The solubility of hydrogen in liquid aluminum, therefore, gives the following equation in terms of the activity coefficient of hydrogen:

$$\ln \gamma_H^{o,Al(l)} = 4.371(\pm 0.045) + \frac{6254(\pm 250)}{T} \quad [14]$$

where  $\gamma_H^{o,Al(l)}$  is the activity coefficient of hydrogen in liquid aluminum and  $T$  is the temperature in Kelvin. The standard state of hydrogen is the state of monatomic hydrogen in liquid aluminum, which is in equilibrium with 1 atm  $H_2$  gas.

Figure 3 shows the experimental data from previous investigations converted to the activity coefficient of hydrogen. The straight line is the equation derived by regression analysis.

### C. Cu-H System

Literature data on the solubility of hydrogen in copper have been reviewed by Hansen and Anderko.<sup>[22]</sup> Among available experimental data, those of Sieverts,<sup>[23]</sup> Rontgen and Moller,<sup>[24]</sup> and Bever and Floe<sup>[25]</sup> are in good agreement. Thus, in this study, analyses are focused on their results. Figure 4 shows the calculated hydrogen activity coefficient as a function of temperature. Using linear regression analysis, the following equation has been obtained:

$$\ln \gamma_H^{o,Cu(l)} = 4.3406(\pm 0.0058) + \frac{5127(\pm 260)}{T} \quad [15]$$

Table II. Interaction Parameters for the Al-Li Melt Calculated from Literature Data

Sources	Temperature	$W_h$ , kK	$W_g$ , kK
Moser <sup>[7]</sup>	873 to 973 K	$-1.930 \pm 0.250$	—
Bushmanov and Yatsenko <sup>[8]</sup>	1023 K	$-2.638 \pm 0.209$	—
Yatsenko and Saltykova <sup>[9]</sup>	1023 K	—	$-1.125 \pm 0.251$
Hicter <i>et al.</i> <sup>[10]</sup>	987 K	—	$-1.094 \pm 0.179$
Hicter <i>et al.</i> <sup>[10]</sup>	957 K	—	$-1.140 \pm 0.198$

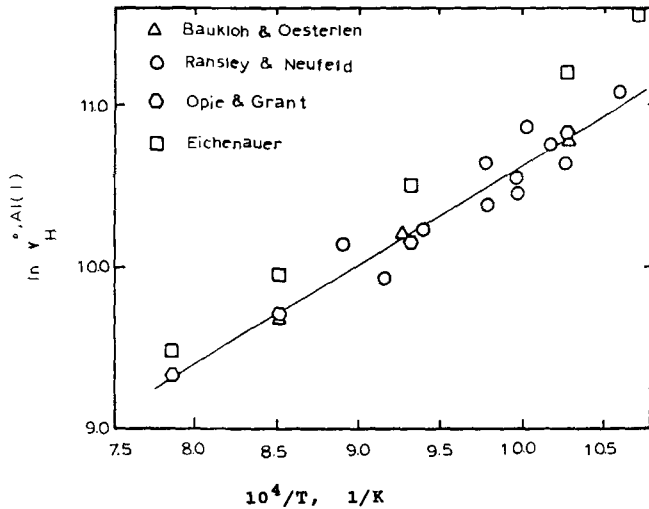


Fig. 3—Activity coefficient of hydrogen in molten aluminum. The solid line is the best fit line obtained in this study.

where  $\gamma_{\text{H}}^{\text{o,Cu(l)}}$  is the activity coefficient of hydrogen in liquid copper.

#### D. Mg-H System

Koeneman and Metcalfe<sup>[26]</sup> have measured the solubility of hydrogen in both liquid and solid magnesium. Their data are used to obtain an equation for the hydrogen activity coefficient in liquid magnesium as a function of temperature (Figure 5). The data of Sharov and Serebryakov<sup>[27]</sup> are not used as no temperature was reported in their paper. The linear regression analysis of the data gives

$$\ln \gamma_{\text{H}}^{\text{o,Mg(l)}} = 3.743(\pm 0.058) + \frac{3040(\pm 260)}{T} \quad [16]$$

where  $\gamma_{\text{H}}^{\text{o,Mg(l)}}$  is the activity coefficient of hydrogen in liquid Mg.

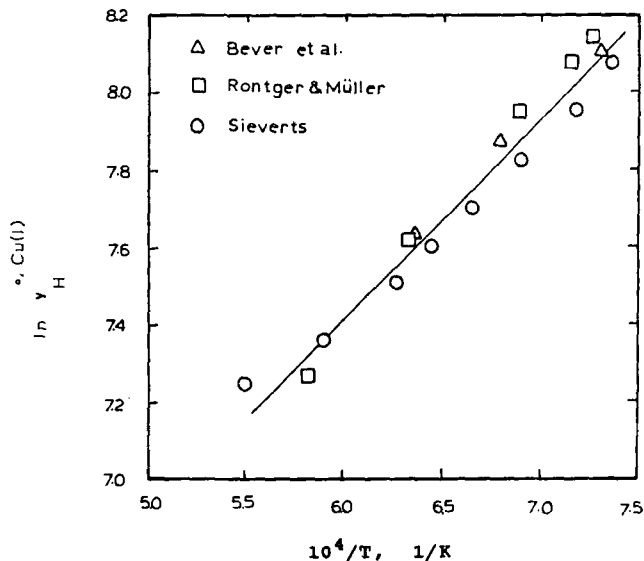


Fig. 4—Activity coefficient of hydrogen in molten copper. The solid line is the best fit line obtained in this study.

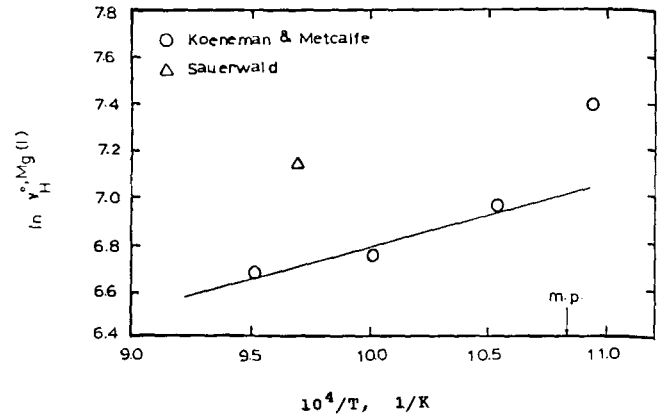


Fig. 5—Activity coefficient of hydrogen in molten magnesium. The solid line is the best fit line obtained in this study.

#### E. Li-H System

The Li-H binary system has been studied by Veleckis *et al.*,<sup>[28]</sup> Heumann and Salmon,<sup>[29]</sup> Amirh *et al.*,<sup>[30]</sup> and Shpilrain *et al.*<sup>[31]</sup> From the data on the hydrogen solubility in lithium, the activity coefficient of hydrogen is calculated and shown in Figure 6. By linear regression analysis, one obtains

$$\ln \gamma_{\text{H}}^{\text{o,Li(l)}} = 5.260(\pm 0.015) - \frac{5318(\pm 250)}{T} \quad [17]$$

where  $\gamma_{\text{H}}^{\text{o,Li(l)}}$  is the activity coefficient of hydrogen in liquid lithium and the standard state of hydrogen is atomic hydrogen in pure liquid lithium in equilibrium with  $\text{H}_2(\text{g})$  at 1 atm.

Table IV summarizes the activity coefficient of the

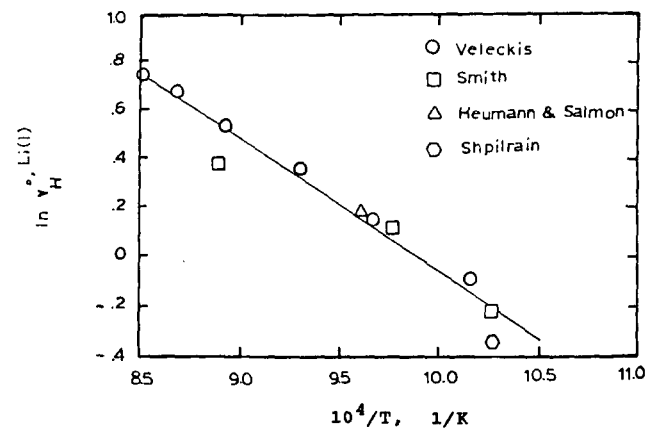


Fig. 6—Activity coefficient of hydrogen in molten lithium. The solid line is the best fit line obtained in this study.

Table IV. Henry's Law Constants for Hydrogen in Me-H Systems

System	$\ln \gamma = A' + A/T$	
	$A'$	$A \times 10^3$
Al-H	$4.371 \pm 0.045$	$6.254 \pm 0.250$
Cu-H	$4.341 \pm 0.058$	$5.127 \pm 0.260$
Mg-H	$3.742 \pm 0.058$	$3.040 \pm 0.260$
Li-H	$5.260 \pm 0.015$	$-5.318 \pm 0.015$
Si-H	$-9.411 \pm 1.521$	$19.832 \pm 0.340$

**Table V. Activity Coefficient  $\ln \gamma$  of Hydrogen in Binary Aluminum Alloys as a Function of Addition Element and Temperature**

Cu Wt Pct						
$T, \text{ kK}$	4	8	12	16	20	50
0.973	10.937	11.065	11.182	11.288	11.381	11.550
1.073	10.326	10.444	10.553	10.651	10.737	10.901
1.173	9.820	9.930	10.031	10.122	10.202	10.364
1.273	9.394	9.496	9.590	9.676	9.751	9.911
Li Wt Pct						
$T, \text{ kK}$	0.5	2	5	10		
0.973	10.632	10.149	9.260	7.972		
1.073	10.046	9.602	8.787	7.616		
1.173	9.560	9.148	8.395	7.320		
1.273	9.150	8.765	8.065	7.070		
Mg Wt Pct						
$T, \text{ kK}$	1	5	10	15	30	
0.973	10.779	10.686	10.540	10.367	9.749	
1.073	10.181	10.094	9.958	9.798	9.227	
1.173	9.685	9.603	9.475	9.326	8.795	
1.273	9.267	9.189	9.068	8.928	8.430	

hydrogen-metal systems obtained above. Since we are interested only in alloys containing less than 10 at. pct of each addition element (Cu, Li, Mg, Si), the Cu-Mg, Cu-Li, *etc.* interactions are assumed to be negligible in the present calculation. There are no data on the solubility of hydrogen in liquid silicon. This value was extracted from the solubility data of hydrogen in aluminum-silicon alloys.

The final expression for  $T \ln \gamma$  in kK for hydrogen is given by

( $X_{\text{Al}}$  indicates atomic fraction of aluminum, *etc.*;  $T$  is in kK everywhere).

$$\begin{aligned}
 T \ln \gamma = & X_{\text{Al}}(E + E'T) + X_{\text{Cu}}(F + F'T) \\
 & + X_{\text{Mg}}(G + G'T) + X_{\text{Li}}(H + H'T) \\
 & + X_{\text{Si}}(I + I'T) - 12AX_{\text{Cu}}X_{\text{Al}}(1 - X_{\text{Al}})^2 \\
 & + 2A'TX_{\text{Cu}}X_{\text{Al}} - 12BX_{\text{Al}}X_{\text{Mg}}(1 - X_{\text{Mg}})^2 \\
 & + 2B'TX_{\text{Mg}}X_{\text{Al}} - 6CX_{\text{Al}}X_{\text{Si}}(1 - X_{\text{Si}}) \\
 & + 6C'TX_{\text{Si}}X_{\text{Al}}(1 - X_{\text{Si}}) - 2DX_{\text{Al}}X_{\text{Li}} \\
 & + 2D'TX_{\text{Al}}X_{\text{Li}}
 \end{aligned}
 \quad [18]$$

where  $A \dots D$  and  $A' \dots D'$  are  $W_h$ 's and  $W_s$ 's for various binary Al-Me systems, as listed in Table I, and  $E \dots I$  and  $E' \dots I'$  are constants in Table IV from the corresponding Me-H systems.

Table V contains the activity coefficients,  $\ln \gamma$ , of hydrogen in aluminum-copper, aluminum-lithium, and aluminum-magnesium alloys as a function of additional element content and temperature. In Figure 7, the values are plotted for 1100 K. Table VI contains the solubility in cc(STP)/100 g alloy of hydrogen in aluminum-copper alloys. These calculated values are compared in Figure 8 with the experimental results of Opie and Grant.<sup>[18]</sup> The agreement is excellent. Figure 9 shows the calculated hydrogen solubility in the Al-Mg melts at various tem-

peratures together with data from Bauklok and Oesterlen.<sup>[16]</sup> Their experimental data scatter rather widely, probably due to the highly oxidizing nature of Mg. The solubility of hydrogen in molten Al-Li alloys is also calculated, as shown in Figure 10. Since no experimental

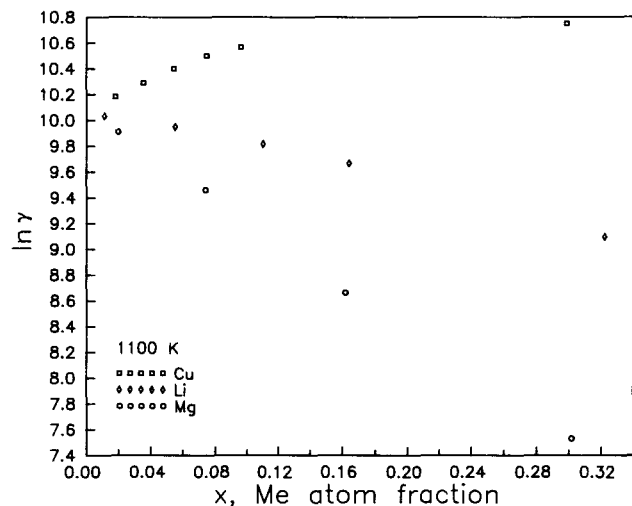


Fig. 7—Estimated activity coefficient of hydrogen in binary Al-Me melts.

**Table VI. Calculated Solubility of Hydrogen [cc(STP)/100 g Alloy] in Aluminum-Copper Alloys**

Wt Pct	$T, \text{ }^\circ\text{C}$			
	700	800	900	1000
Cu				
0	0.89	1.75	2.75	4.11
2	0.60	1.31	2.50	3.90
4	0.50	1.08	2.14	3.47
7	0.54	1.00	2.00	3.19
12	0.44	0.89	1.55	2.50
23	0.31	1.19	1.22	2.00
50	0.36	0.56	1.06	1.55

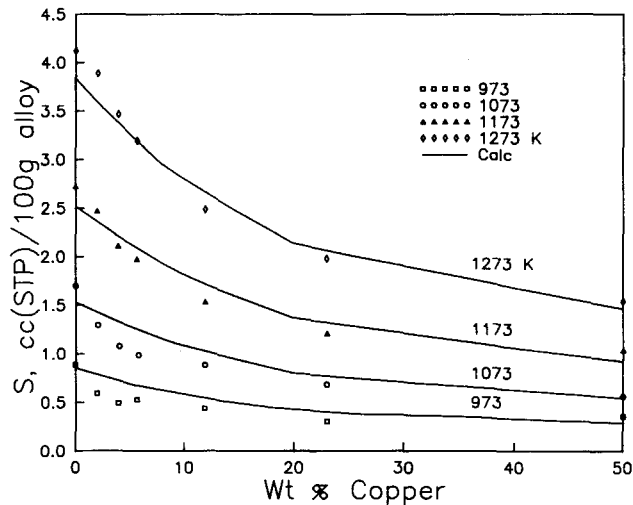


Fig. 8—Solubility of H<sub>2</sub> in molten Al-Cu alloys. Solid lines are the estimated values from this study. Data points are from Opie and Grant.<sup>[18]</sup>

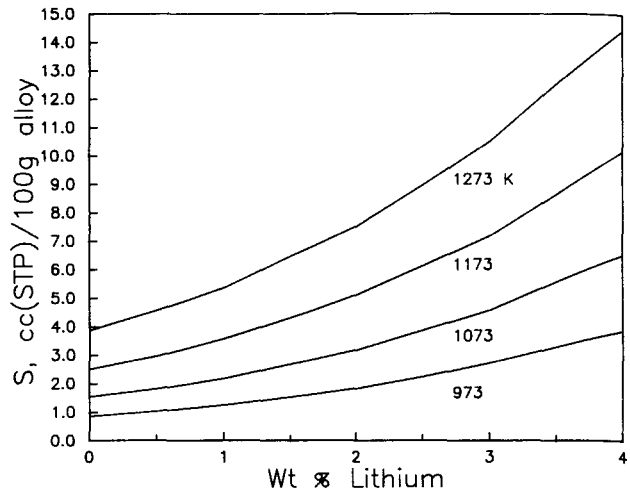


Fig. 10—Solubility of H<sub>2</sub> in molten Al-Li alloys. Solid lines are the estimated values from this study. No experimental data are available in the literature.

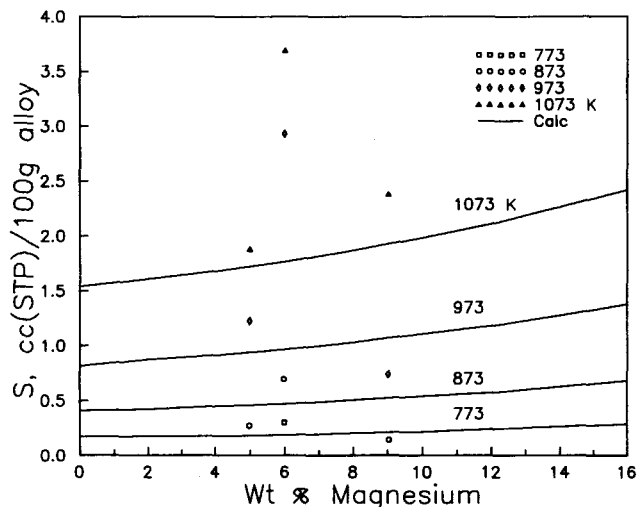


Fig. 9—Solubility of H<sub>2</sub> in molten Al-Mg alloys. Solid lines are the estimated values from this study. Data points are from Bauklok and Oesterlen.<sup>[16]</sup>

data are available from the literature, this figure may provide valuable information in the preparation of the aluminum-lithium alloys.

Table VII contains the solubility of hydrogen in liquid silicon, obtained from the experimental data on aluminum-silicon alloys of Opie and Grant.<sup>[18]</sup> This value is also introduced into Table IV and can be used with other silicon-containing alloys.

Table VII. Solubility of Hydrogen in Liquid Silicon

T, kK	T ln γ, kK Si, Wt Pct							Std. Dev.
	2	4	7.5	10.5	12	19	Avg.	
0.973	12.684	11.065	9.928	11.066	10.617	10.096	10.909	0.904
1.073	8.216	11.303	10.783	9.313	8.675	8.773	9.510	1.139
1.173	5.739	9.085	9.845	9.006	8.821	8.715	8.535	1.302
1.273	5.833	7.436	8.955	8.749	8.668	8.941	8.097	1.137

#### IV. SUMMARY

The solution model developed by Hoch and Arpshofen<sup>[2,3]</sup> appears to be suitable in describing the solution behavior of binary and ternary alloys of aluminum. Applying this model, the thermodynamics of the Al-H, Cu-H, Mg-H, Li-H, Al-Cu, Al-Si, Al-Mg, Al-Li, Al-Li-H, Al-Cu-H, Al-Mg-H, and Al-Si-H systems have been evaluated. The advantage of this model is the possibility of applying the binary parameters in evaluating the thermodynamic properties of the corresponding ternary systems without any measurement in the ternary system. The agreement between the theoretically derived thermodynamic data and the experimental results obtained by Opie and Grant<sup>[18]</sup> on the Al-Cu-H system is good, as shown in Figure 8. Using the ternary Al-Si-H data by Opie and Grant, the solubility of hydrogen in molten silicon is suggested.

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