

Critical Evaluation and Optimization of the Thermodynamic Properties and Phase Diagrams of the Al-Mg, Al-Sr, Mg-Sr, and Al-Mg-Sr Systems

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All available thermodynamic and phase diagram data were critically assessed for all phases in the Al-Mg, Al-Sr, and Mg-Sr systems at 1 bar pressure from room temperature to above the liquidus temperatures. For these systems, all reliable data were simultaneously optimized to obtain a set of model equations for the Gibbs energy of the liquid alloy and all solid phases as functions of composition and temperature. The modified quasi-chemical model was used for the liquid. The Al-Mg-Sr ternary phase diagram was calculated from the optimized thermodynamic properties of the binary systems. Since no reliable ternary data were available, three assumptions were made: no ternary terms were added to the model parameters for the thermodynamic properties of the liquid, no ternary solid solutions are present in the system, and no ternary compound is present in the system. The calculated ternary phase diagram is thus a first approximation, which can be improved by the addition of new experimental data and can be used as a base for the calculation of phase diagrams of multicomponent systems.

Introduction

Strontium is used, like sodium, in aluminum cast alloys containing silicon to modify the acicular structure of the Al-Si eutectic. Timminco Ltd., which produces Al-Sr master alloys for the aluminum industry, has patented master Al-Sr-Mg compositions with an increased Sr content. To control the production of ingots of these master alloys, the Al-Mg-Sr phase diagram is required, but no satisfactory experimental phase diagram is currently available. The prediction of the phase diagram is possible from the thermodynamic optimizations of the three binary systems using appropriate models. [82Mur] reviewed the literature and optimized the Al-Mg system. Other optimizations of this system were performed by [77Sab], [86Lud], [90Sau], and [93Zuo]. [89Alc] studied and optimized the Al-Sr system. [91Sri] also made an optimization of this system. [86Nay] made the only optimization of the Mg-Sr system.

Because more recent thermodynamic data are now available, and in order to obtain more complete and precise results, the present authors decided to reoptimize the three binary systems before calculating the ternary phase diagram.

A ternary diagram calculation requires that the same model be used in all binary systems for each phase present in the ternary field. For the liquid phase, the present authors used the modified quasi-chemical model [86Pel], which is well adapted to describe the ordered liquid in the Al-Sr system. The model considers the Gibbs energy for the formation of two $A-B$ bonds from one $A-A$ bond and one $B-B$ bond (see Eq 1).

$$[A-A] + [B-B] = 2[A-B] \quad (\text{Eq 1})$$

Expressions for enthalpies and entropies of mixing are written in terms of the bond fractions X_{AA} , X_{BB} , and X_{AB} , and in terms of $(\omega - \eta T)$ which is the Gibbs energy change of the bond exchange reaction (Eq 1). The equilibrium bond fractions are obtained by setting:

$$\left(\frac{\partial G}{\partial X_{AB}} \right) = 0 \quad (\text{Eq 2})$$

while taking account of the mass balances. This results in an "equilibrium constant" for the bond exchange reaction (Eq 1). Details of the model were presented by [86Pel]. The fixed parameters of the model in the present evaluation are "coordination numbers" $Z_{Al} = b_{Al}Z$, $Z_{Mg} = b_{Mg}Z$, and $Z_{Sr} = b_{Sr}Z$ where $Z = 6$, $b_{Al} = 1.3774$, $b_{Mg} = 0.9183$, and $b_{Sr} = 2.0661$. These parameters are those used in previous evaluations, which are forming a data base on metallic liquid solutions.

The thermodynamic properties of Al, Mg, and Sr used in this evaluation are given in Table 1 and apply to Eq 3:

$$H = A + \int_{298.15}^T C_p dT \text{ J/g-atom}$$

$$S = B + \int_{298.15}^T (C_p/T) dT \text{ J/K} \cdot \text{g-atom}$$

$$C_p = a + b(10^{-3})T + c(10^5)T^{-2} + d(10^{-6})T^2 \text{ J/K} \cdot \text{g-atom} \quad (\text{Eq 3})$$

Section I: Basic and Applied Research

Table 1 Thermodynamic Properties of Elements and Compounds Relative to Elements at 25 °C

	A	B	a	b	c	d
Elements (from literature)						
Al(L) (298.15 to 933.45 K).....	10 711.00	39.7961	31.3758	-16.3929	-3.6066	20.7526
Al(L) (>933.45 K).....	8 680.87	35.2537	31.7482
Mg(L) (298.15 to 923 K).....	8 477.00	41.8612	21.3886	11.7780
Mg(L) (>923 K).....	5 943.07	36.5117	32.6352
Sr(L) (298.15 to 2000 K).....	10 019.31	61.1563	30.9616
Al(s) (298.15 to 933.45 K).....	0	28.3215	31.3758	-16.3929	-3.6066	20.7526
Mg(s) (298.15 to 923 K).....	0	32.6770	21.3886	11.7780
(αSr) (298.15 to 829.3 K).....	0	52.3000	22.2170	13.8909
(γSr) (829.3 to 1042.15 K).....	2 006.68	56.1759	12.6775	26.7776
Compounds (optimized)						
Al ₄₅ Mg ₂₈ (298.15 to 724.25 K).....	-306.45	34.3321	27.5451	-5.5876	-2.2233	12.7927
Al ₁₂ Mg ₁₇ (298.15 to 733.65 K).....	-250.00	35.7317	25.5212	0.12105	-1.4924	8.5873
R (Al _{0.58} Mg _{0.42}) (576.14 to 674.58 K)...	-153.14	34.8738	27.1812	-4.5611	-2.0918	12.0365
Al ₄ Sr (298.15 to 1298.25 K).....	-31 210.0	24.9642	29.5441	-10.3362	-2.8853	16.6021
Al ₂ Sr (298.15 to 1195.45 K).....	-28 447.5	34.5783	24.8229	-6.2983	-2.4044	13.8350
Al ₇ Sr ₈ (617.97 to 937.1 K).....	-19 279.3	39.8740	28.9911	-0.24156	-1.6831	9.6850
Mg ₁₇ Sr ₂ (298.15 to 879.24 K).....	-1 985.68	35.9122	22.8758	-12.0004
Mg ₃₈ Sr ₉ (298.15 to 871.72 K).....	-3 684.85	38.3116	21.5472	12.1826
Mg ₂₃ Sr ₆ (298.15 to 876.2 K).....	-4 050.00	38.5300	21.5600	12.2151
Mg ₂ Sr (298.15 to 953.23 K).....	-7 106.34	40.0430	21.6648	12.4823

Note: See Eq 3.

For Al, $S^0(298)$ for the solid phase, $\Delta_{\text{fus}}H^0$, $\Delta_{\text{fus}}S^0$, and expressions of C_p for solid and liquid were taken from [77Bar], who also fixed the melting point at 660.3 °C. Below the melting point, $C_p(L)$ was set equal to $C_p(s)$. For Mg, $\Delta_{\text{fus}}H^0$ and $\Delta_{\text{fus}}S^0$ were taken from [85Cha], who also fixed the melting point at 649.85 °C. $S^0(298)$ for the solid phase and expressions of C_p for solid and liquid were taken from [77Bar]. Below the melting point, $C_p(L)$ was set equal to $C_p(s)$. For Sr, $S^0(298)$ for the α -Sr solid phase, $\Delta_{\text{fus}}H^0$, $\Delta_{\text{fus}}S^0$, and expressions of C_p for solid allotropes and liquid were taken from [77Bar], who also fixed the melting point at 660.3 °C. $\Delta_{\text{trs}}H^0$ and $\Delta_{\text{trs}}S^0$ were also taken from [77Bar]. The properties of the compounds listed in Table 1 were obtained from optimizations performed in this work.

Equations 4 through 7 provide optimized parameters of the modified quasi-chemical model for the liquid.

$$Y_m = \frac{b_m X_m}{b_m X_m + b_n X_n} \quad (\text{Eq 4})$$

where $m, n = \text{Al, Mg, and Sr}$ ($Z=6$, $b_{\text{Al}} = 1.3774$, $b_{\text{Mg}} = 0.9183$, and $b_{\text{Sr}} = 2.0661$).

For Al-Mg:

$$\begin{aligned} \omega &= -9813.4 + 6169.4 Y_{\text{Mg}} - 1536 Y_{\text{Mg}}^2 \text{ J/g-atom} \\ \eta &= -4 + 4 Y_{\text{Mg}} \text{ J/K} \cdot \text{g-atom} \end{aligned} \quad (\text{Eq 5})$$

For Al-Sr:

$$\begin{aligned} \omega &= -47 938.5 + 2 863.4 Y_{\text{Sr}} + 20 283 Y_{\text{Sr}}^2 \text{ J/g-atom} \\ \eta &= -8 \text{ J/K} \cdot \text{g-atom} \end{aligned} \quad (\text{Eq 6})$$

For Mg-Sr:

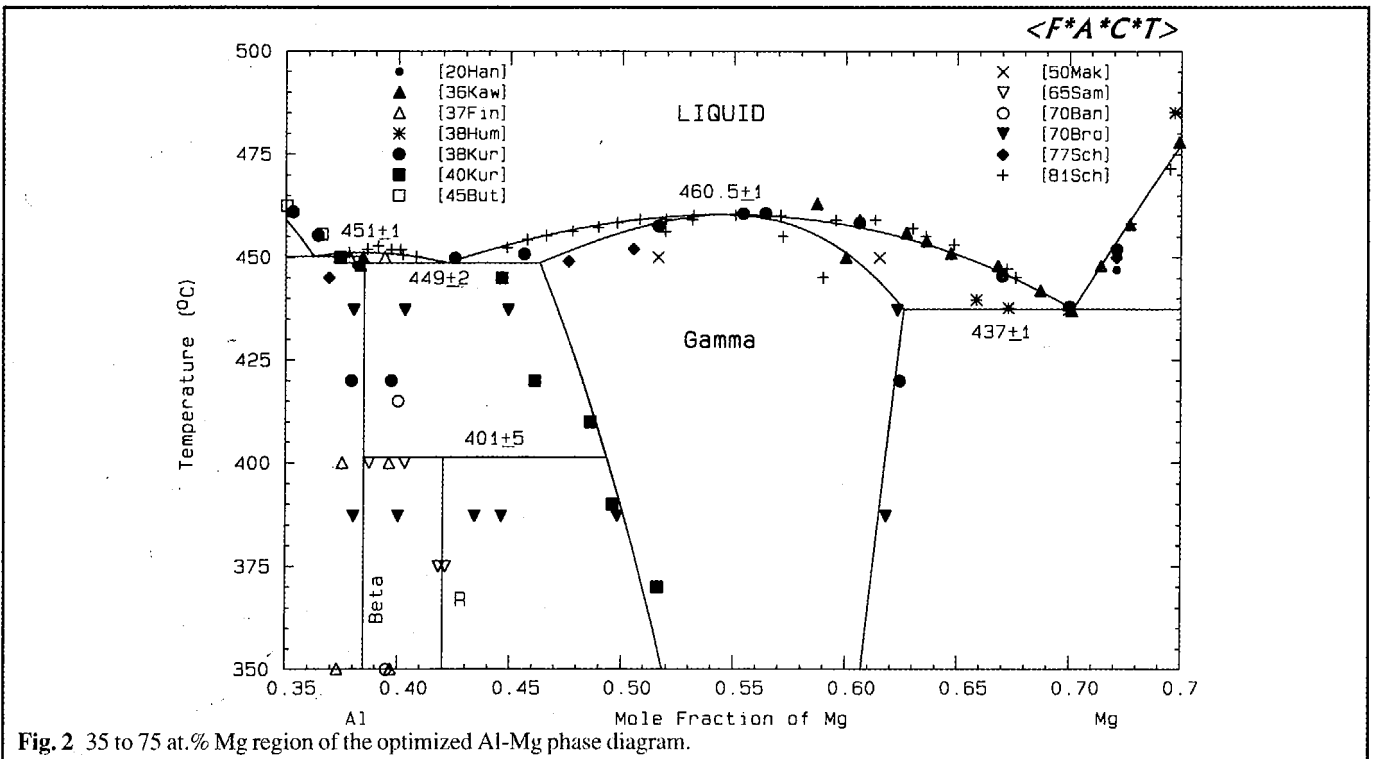
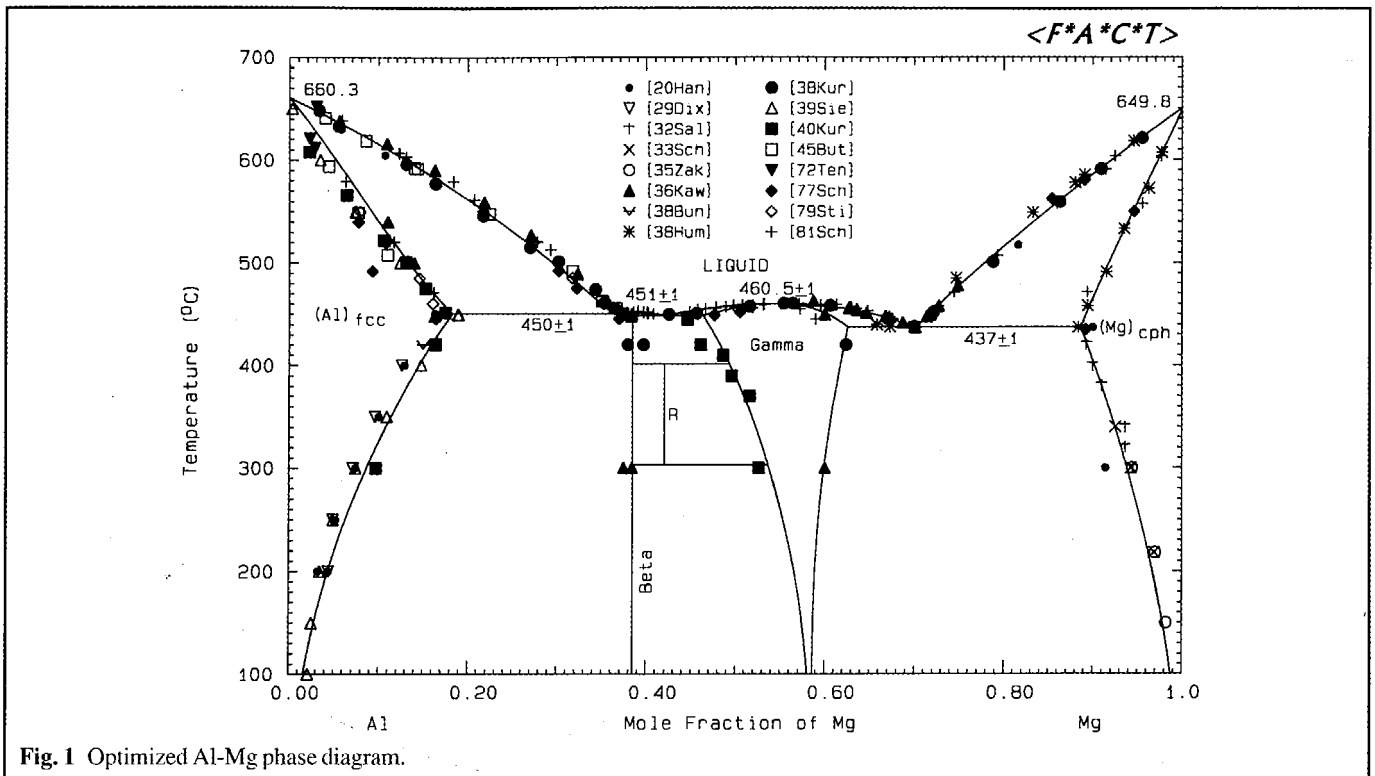
$$\begin{aligned} \omega &= -16 757.8 + 3 395.7 Y_{\text{Sr}} - 5 632.7 Y_{\text{Sr}}^2 \text{ J/g-atom} \\ \eta &= -9 + 4 Y_{\text{Sr}} \text{ J/K} \cdot \text{g-atom} \end{aligned} \quad (\text{Eq 7})$$

The Al-Mg System

Equilibrium Diagram

The Al-Mg system was studied by numerous authors, who reported a great many liquidus, solidus, and solvus points. A review of these data was published by [82Mur]. Most of the present calculated phase diagram (Fig. 1 and 2) agrees with this review. [82Mur] stated that the equilibrium solid phases are: the (Al)fcc solid solution with a maximum solubility of Mg of 18.9 at.% at the eutectic temperature of 450 °C; the (Mg)cph solid solution with a maximum solubility of Al of 11.8 at.% at the eutectic temperature of 437 °C; the β fcc solid solution; the γ solid solution with the α Mn structure; and the R phase at 42 at.% Mg (also designated ϵ). Results of [81Sch] are not considered in the review of [82Mur]. The (Al)fcc solidus points measured by [81Sch] agree in this region with the experimental results of [36Kaw], [39Sie], [40Kur1], [45But], and [79Sti].

In their optimizations, [90Sau] and [93Zuo] included another phase named ζ (from [77Sch]), and [86Lud] considered the ζ phase and not the R phase. The results of [89Goe] supported the conclusion of [82Mur], but stated that the R phase is stable between 305 ± 5 and 405 ± 5 °C instead of 370 ± 5 °C proposed by [82Mur]. [89Goe] studied several diffusion couples of Al, Mg, and their alloys in graphite dies between 280 and 435 °C during 5 to 21 days. The following phases were observed: (Al)fcc, (Mg)cph, β , γ , and R .



Crystal Structures and Lattice Parameters

The crystal structures and lattice parameters were discussed by [82Mur], who mentioned an uncertainty concerning the exact number of atoms per unit cell for the β phase. In the present evaluation, the hypothetical stoichiometry of the β compound

is $Al_{45}Mg_{28}$ as proposed by [65Sam]. Otherwise the evaluation of [82Mur] is accepted.

Thermodynamics

The enthalpy of mixing of the liquid was determined calorimetrically by [30Kaw], [71Bat], and [91Aga]. [76Bha] and

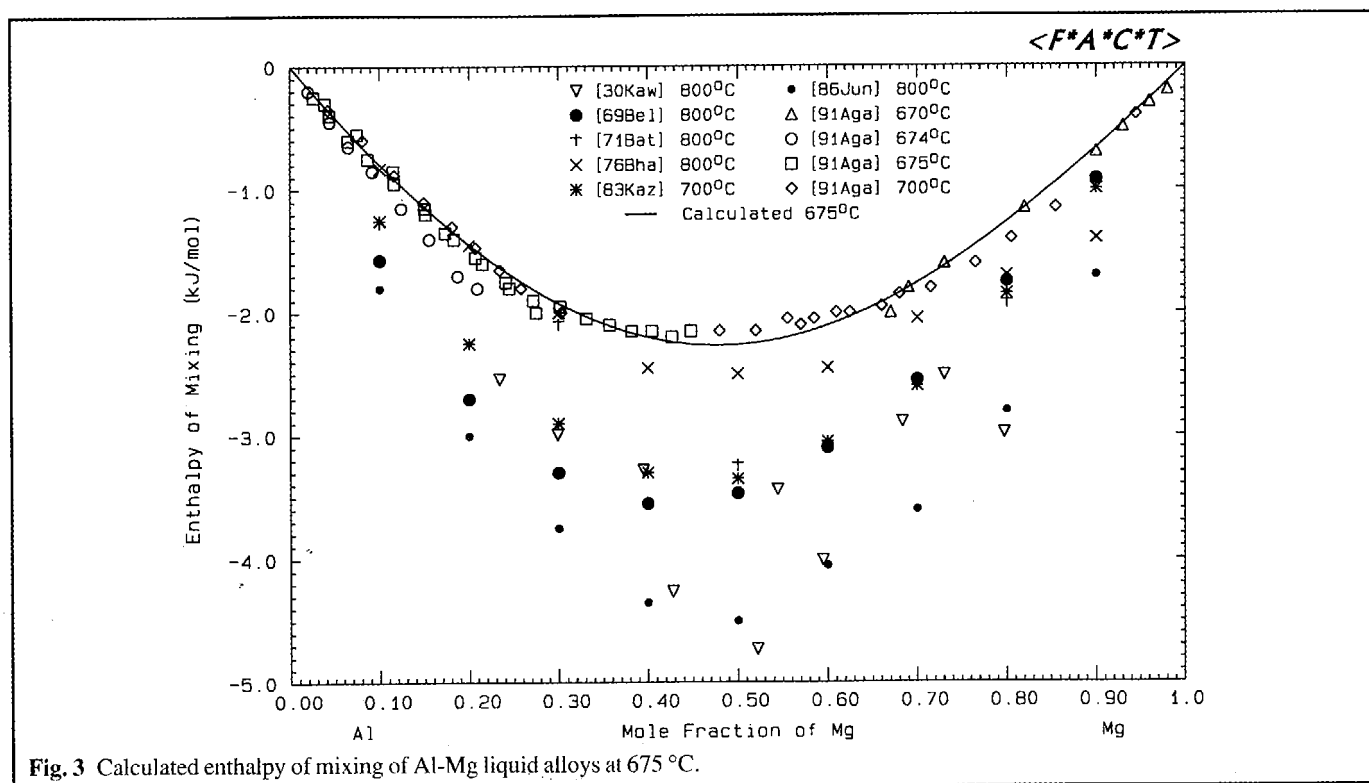


Fig. 3 Calculated enthalpy of mixing of Al-Mg liquid alloys at 675 °C.

[86Jun] derived the enthalpy of mixing from partial pressure measurements, while [69Bel] and [87Tiw] obtained the enthalpy of mixing from emf measurements. The methods used by [83Kaz] are not clear. The results of [91Aga] appear to be reliable. Systematic errors were limited by the use of three different calorimetric methods. From their results, the liquid is a regular solution with a minimum enthalpy of mixing of -2.2 kJ/mol at 50 at.% Mg. Results are shown in Fig. 3.

The activity of Mg in the liquid alloy was determined with emf measurements by [62Ere], [69Bel], [69Tsy], and [87Tiw]. [41Sch], [71Vya], [76Bha], and [86Jun] measured the partial pressure of Mg. All results are scattered but show a small negative deviation from ideality, except for those of [69Tsy]. See Fig. 4.

By the integration of DTA curves, [78Pre1] obtained the enthalpy required to heat the solid phases from a temperature just below the eutectic or the solidus to a temperature just above the liquidus. [80Tim] obtained the enthalpy of fusion of the β phase. Results of [78Pre1] and [80Tim] are shown in Fig. 5. The activity of Mg in solid phases at 387 and 437 °C was determined by [70Bro] using emf techniques. See Fig. 6 and 7.

Optimization of Thermodynamic Properties and Phase Diagram

The calculated optimized Al-Mg phase diagram is shown in Fig. 1. Figure 2 shows the 35 to 75 at.% Mg region. The calculated parameters for the solid phases are presented in Table 1 and Eq 8, 9, and 11. The calculated parameters for the liquid phase are shown in Eq 5. Calculated invariant points are listed in Table 2.

The liquid was modeled by the modified quasi-chemical method using the results of [91Aga] for the modified quasi-chemical and experimental liquidus points of the (Al)fcc and (Mg)cph phases. Measurements of the activity of the (Mg) in the liquid were not used directly for the optimization process because of the high dispersion of data but were used for validation of calculated parameters. Figure 8 shows the calculated entropy of the liquid at 675 °C; the excess entropy, S^{ex} , is less than -1 J/mol · K over the entire composition range. In Fig. 3 and 4, the calculated enthalpy of mixing and Mg activity in the liquid are compared with the experimental data.

All the solid phases were modeled by taking into account the experimental results of [70Bro] and [78Pre1], in concert with the solvus, solidus, and liquidus of the phase diagram. The (Mg)cph solid solution was modeled as a Henrien solution. The calculated parameters are:

$$RT \ln \gamma_{Al}^0 = 12\,740 - 11.2T \text{ J/g-atom} \quad (\text{Eq 8})$$

The (Al)fcc phase was modeled as a Henrien solution with an additional regular mixing term as follows:

$$G^{ex} = X_{Al}X_{Mg}(12\,000 - 10.72T) + 150X_{Mg} \text{ J/g-atom} \quad (\text{Eq 9})$$

For these phases, the mutual solid solubilities considered in the present evaluation are those of [82Mur] (~ 18.9 at.% Mg in Al and ~ 11.5 at.% Al in Mg). The atomic radii ratio of Al and Mg is 1.12, which suggests high mutual solubilities.

For calculation, the β phase was assumed to be stoichiometric. This assumption has little effect on the solvus of the (Al)fcc and γ phases because of the small range of homogeneity of the β phase. See [37Fin], [38Kur2], [65Sam], and [70Bro]. The en-

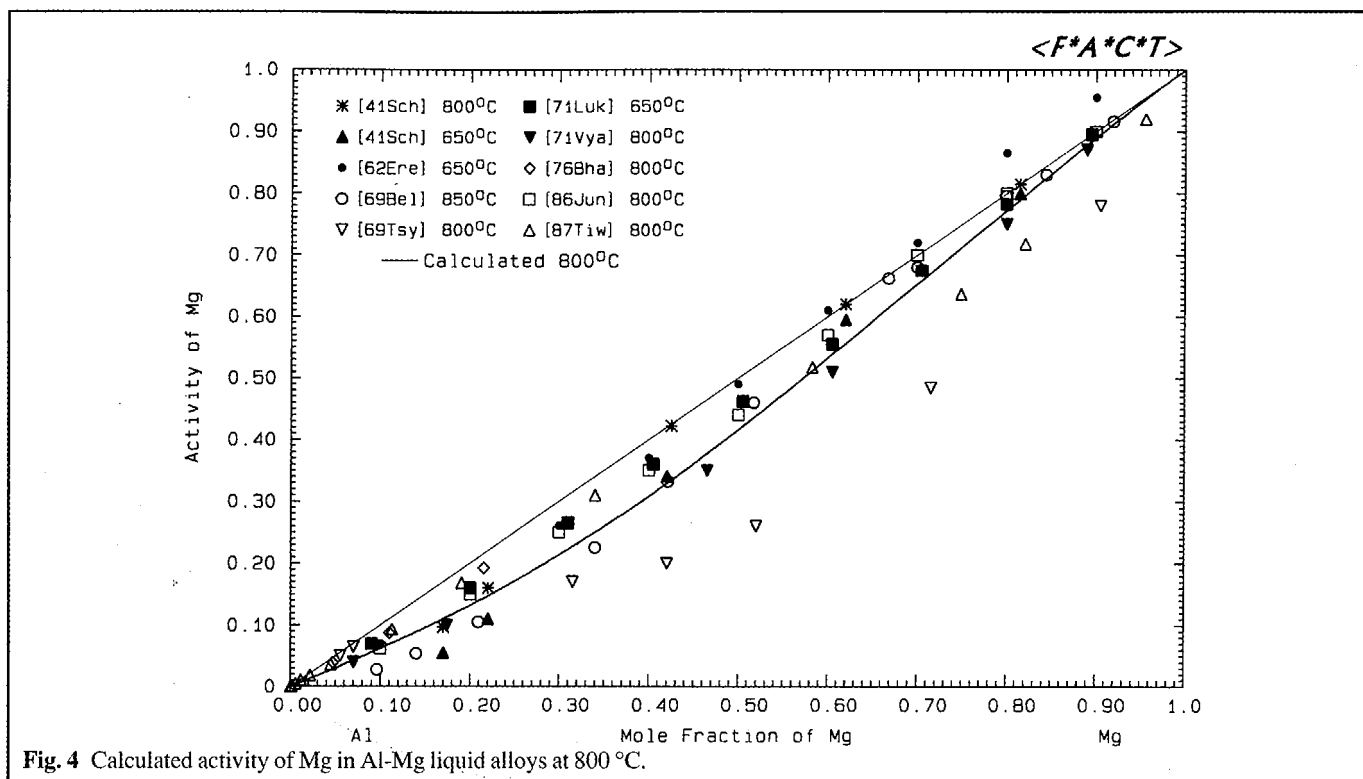


Fig. 4 Calculated activity of Mg in Al-Mg liquid alloys at 800 °C.

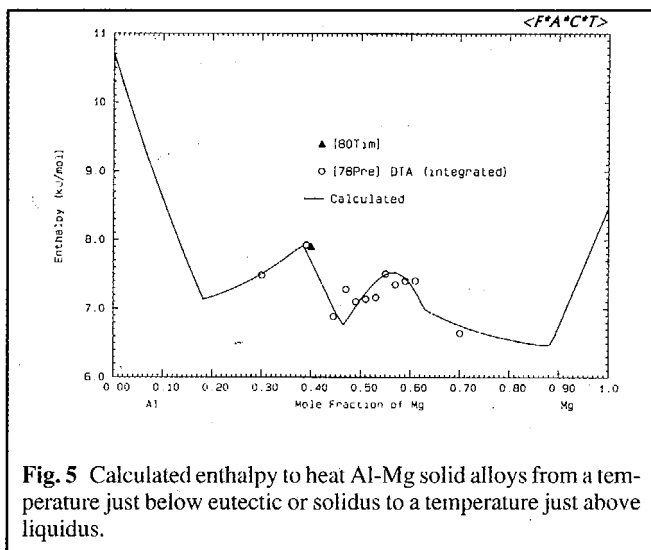


Fig. 5 Calculated enthalpy to heat Al-Mg solid alloys from a temperature just below eutectic or solidus to a temperature just above liquidus.

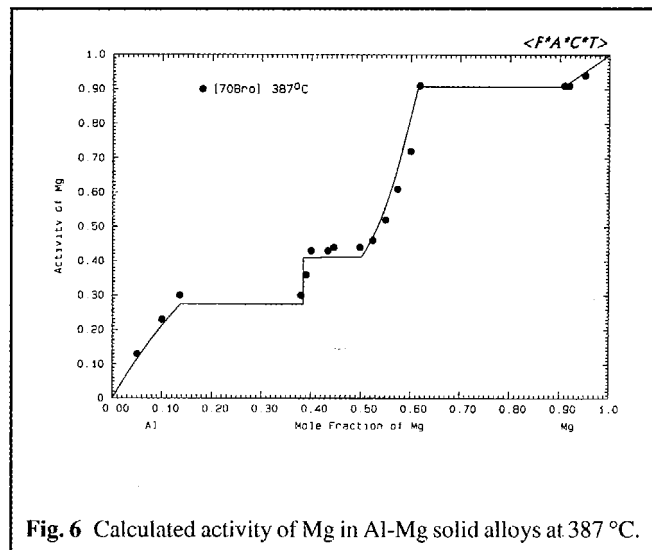


Fig. 6 Calculated activity of Mg in Al-Mg solid alloys at 387 °C.

thalpies of fusion of [78Pre1] and [80Tim] were used with the temperature of fusion (estimated at 451 °C) to calculate the entropy of fusion of the compound. The optimized entropy of fusion for the β phase is 10.94 J/K · g-atom, which is reasonable.

The γ phase was modeled with a defect model [90Li], which is similar to the Wagner-Schottky model. This model incorporates the Gibbs energy associated with the defects on each side of the stoichiometric composition (in this case $Al_{12}Mg_{17}$). The entropic term for the expression of the Gibbs energy of the phase is expressed in terms of the mole fractions of the majority point defects on the Al-rich (X_A) and Mg-rich (X_M) sides of the stoichiometric composition. The Gibbs energy of forma-

tion of the majority defects are added to this entropic term. The Gibbs energy of the phase is then given by:

$$G = RT[X_A \ln X_A + (1 - X_A) \ln(1 - X_A)] + RT[X_M \ln X_M + (1 - X_M) \ln(1 - X_M)] + G_A X_A + G_M X_M \quad (\text{Eq 10})$$

For a fixed deviation from the stoichiometric composition, equilibrium mole fractions of the majority defects X_A and X_M can be calculated to obtain the Gibbs energy of the phase. The optimized expression for the Gibbs energy at the stoichiometric composition $Al_{12}Mg_{17}$ is given in Table 1. The optimized expressions for the Gibbs energies of formation of the majority

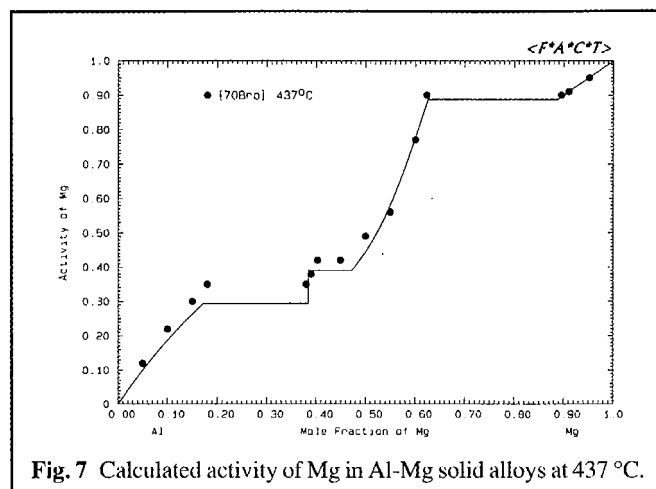


Fig. 7 Calculated activity of Mg in Al-Mg solid alloys at 437 °C.

defects on the Al-rich and Mg-rich sides of $\text{Al}_{12}\text{Mg}_{17}$ are, respectively:

$$\begin{aligned} G_A &= 18\,200 \text{ J/g-atom} \\ G_M &= 30\,250 - 14T \text{ J/g-atom} \end{aligned} \quad (\text{Eq 11})$$

The optimized entropy of fusion for the γ phase (at the composition of the maximum of the azeotrope) is $10.27 \text{ J/K} \cdot \text{g-atom}$. This value is reasonable.

The linear expression for the Gibbs energy of the R phase, which is considered stoichiometric for calculation purposes (42 at.% Mg), is given in Table 1 and was obtained from the formation and decomposition temperatures of [89Goe] ($305 \pm 5 \text{ }^\circ\text{C}$ and $405 \pm 5 \text{ }^\circ\text{C}$). In Fig. 5 to 7, the calculated "enthalpy of fusion" of solid phases and the calculated Mg activities in the solid phases are compared with experimental data.

The principal differences between the present optimization and those of [90Sau] and [93Zuo] are their inclusion of a ζ phase and their use of an optimized enthalpy of mixing, which is an average of the various measurements in Fig. 3, whereas in the present study the results of [91Aga] are given preference.

The Al-Sr System

Equilibrium Diagram

[89Alc] reviewed the Al-Sr system. According to their evaluation, the stable phases of the system are: the liquid, the (Al)fcc solid solution, the (γ Sr)bcc solid solution, the (α Sr)fcc solid solution, and three intermetallic compounds— Al_4Sr , Al_2Sr , and Al_7Sr_8 .

The solid phases considered in the present evaluation (Fig. 9 and 10) are the same, but the interpretation of the phase diagram is somewhat different. The proposed phase diagram of [89Alc] was largely inspired by the experimental points of [75Bru]. The present evaluation is based on the work of [86Clo], which is more consistent with thermodynamic principles. [75Bru], supported by [79Vak], reported that the melting point of Al_4Sr is $\sim 1040 \text{ }^\circ\text{C}$, whereas [86Clo] observed a melting point at $1025 \text{ }^\circ\text{C}$. In a first attempt, [74Vak] proposed a melting point at $1000 \text{ }^\circ\text{C}$. [75Bru] suggested that Al_2Sr is associated with a peritectic reaction at $936 \pm 2 \text{ }^\circ\text{C}$, and their results are supported by [79Vak], who placed the reaction at $940 \text{ }^\circ\text{C}$.

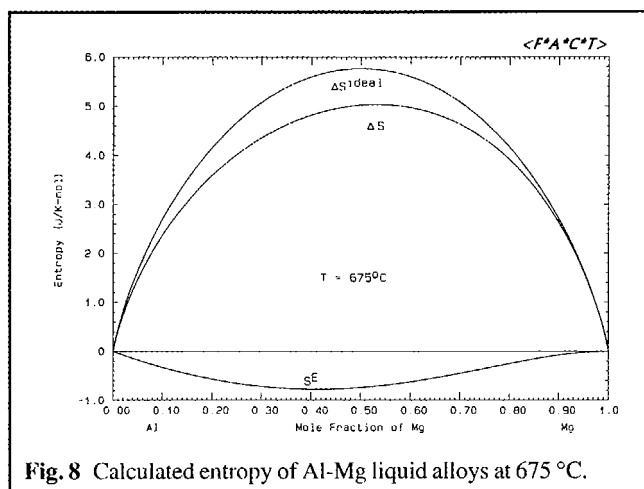


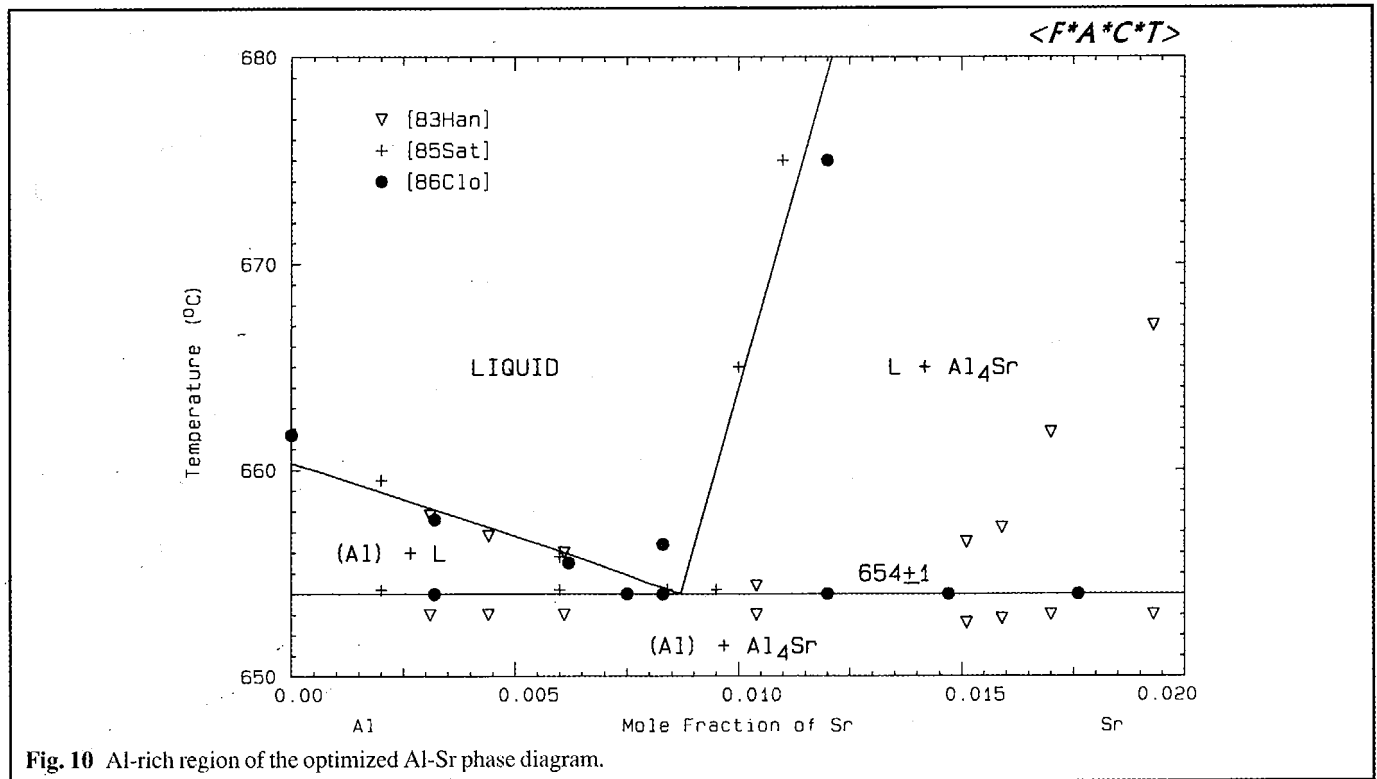
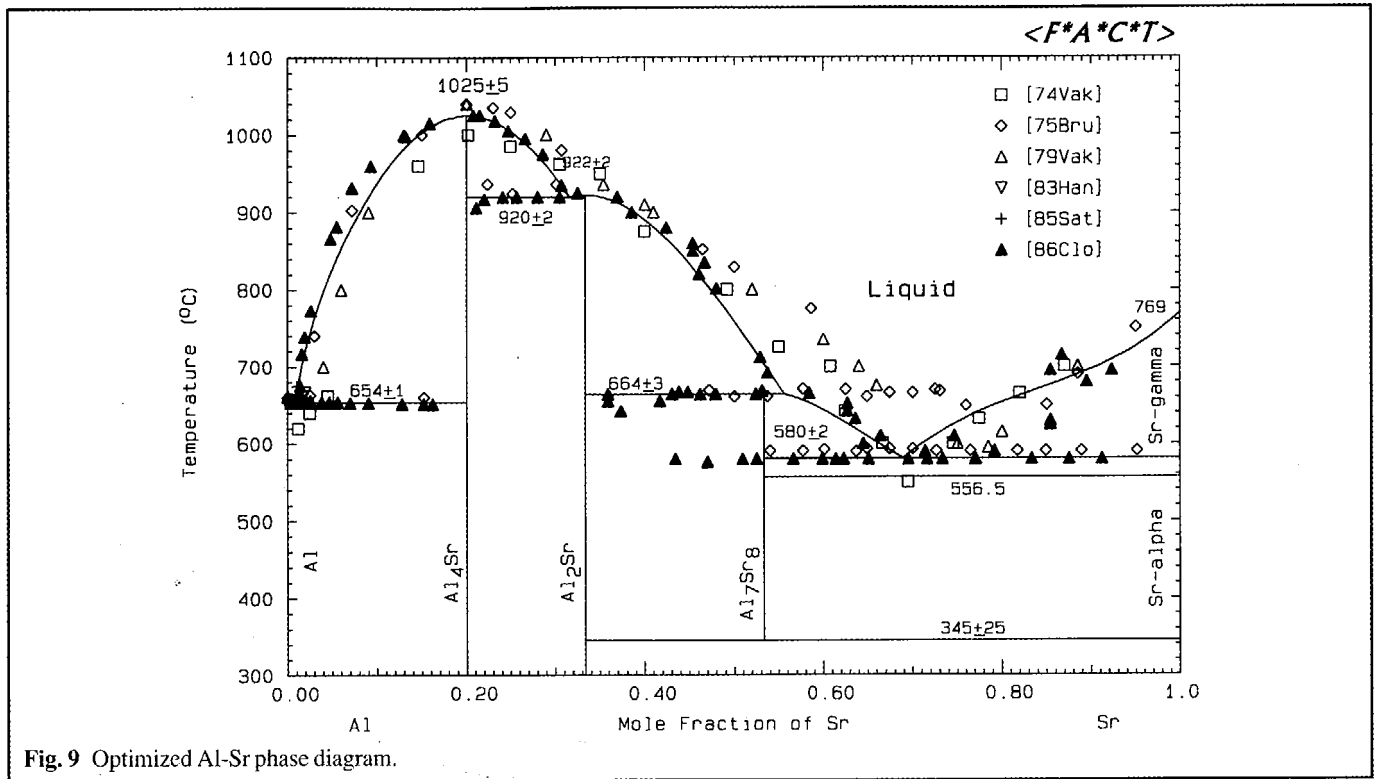
Fig. 8 Calculated entropy of Al-Mg liquid alloys at 675 °C.

[86Clo] observed a thermal arrest at $\sim 920 \pm 1 \text{ }^\circ\text{C}$ and proposed that this was associated with a peritectic reaction. The description of the experiments of [86Clo] is more complete, and their results are self-consistent in the 0 to 70 at.% Sr range of composition. Results of [74Vak], [75Bru], and [79Vak] are not supported by a good description of the experimental methods. The flatness of the liquidus of Al_2Sr as proposed by [75Bru] requires an unreasonable entropy of fusion of this compound ($>25 \text{ J/K} \cdot \text{g-atom}$), while the experimental liquidus points of Al_2Sr of [86Clo] agree with a more reasonable entropy of fusion.

The present evaluation considers that Al_2Sr is a congruent compound and that there is a eutectic reaction $L \rightarrow \text{Al}_4\text{Sr} + \text{Al}_2\text{Sr}$ at $920 \text{ }^\circ\text{C}$ with a eutectic liquid composition at $\sim 32 \text{ at.}\%$ Sr. This interpretation agrees better with the experimental points of [86Clo] and is more probable than the proposed peritectic reaction by reason of symmetry of the liquidus of Al_4Sr . Interpretations of previous authors show a liquidus of Al_4Sr very asymmetric on either side of the stoichiometric composition. The peritectic reaction $L + \text{Al}_2\text{Sr} \rightarrow \text{Al}_7\text{Sr}_8$ at $\sim 664 \text{ }^\circ\text{C}$ is placed at 56 at.% Sr as proposed by [86Clo] (who reported AlSr instead of Al_7Sr_8). Another difference between the present evaluation and the conclusions of [89Alc] is the liquidus of γ Sr. As demonstrated by [89Alc] and as discussed below, the solubility of Al in γ Sr should be very small, so the limiting slope of the liquidus of γ Sr at $X_{\text{Sr}} \rightarrow 1$ (which is related to the enthalpy of fusion and the temperature of fusion of Sr by Eq 12) must be more negative than the proposed slope of [89Alc].

$$\left(\frac{dX_i}{dT} \right)_{X_i=1}^L - \left(\frac{dX_i}{dT} \right)_{X_i=1}^S = \frac{\Delta_{\text{fus}} H(i)}{RT_{\text{fus}}^2(i)} \quad (\text{Eq 12})$$

In the present evaluation, the experimental liquidus of γ Sr was not used in the optimization of the liquid, but was calculated afterwards from the optimized thermodynamic properties of the liquid (and the properties of γ Sr from the literature). The calculated temperature of the eutectic $L \rightarrow \text{Al}_7\text{Sr}_8 + \gamma\text{Sr}$ is that of [86Clo] at $580 \text{ }^\circ\text{C}$; the eutectic liquid composition is more uncertain and was set at $\sim 70 \text{ at.}\%$ Sr from the experimental points of [86Clo].



Thermodynamics

The enthalpy of mixing of the liquid was determined calorimetrically by [83Som] and [85Esi] (see Fig. 11), and as

shown in [89Alc], the results are in good agreement for the composition of the minimum of the curve (35 to 40 at.% Sr). The activity of Sr in the liquid alloy was measured by [74Bur],

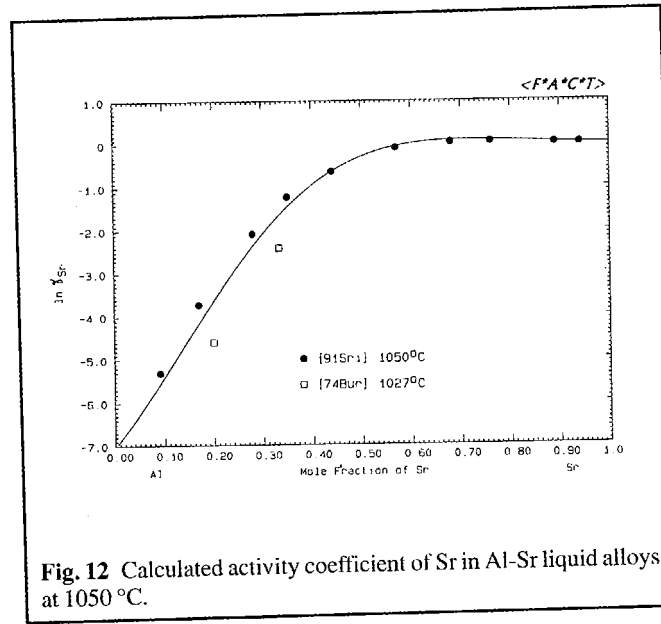
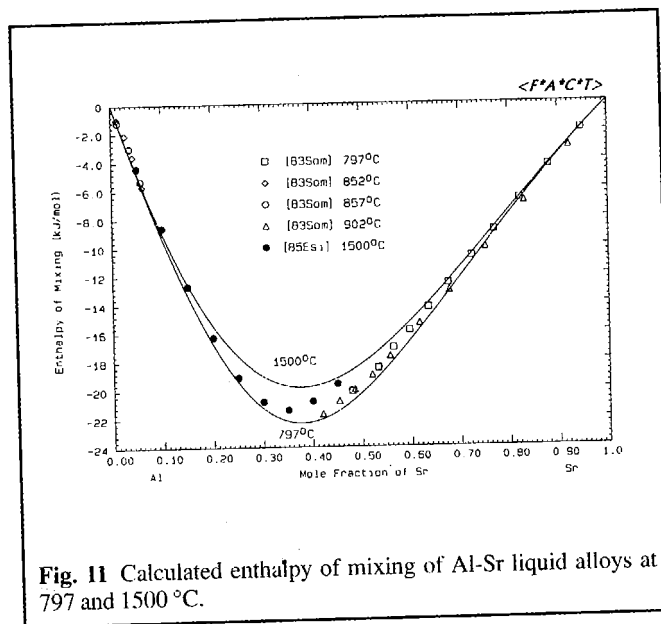


Fig. 12 Calculated activity coefficient of Sr in Al-Sr liquid alloys at 1050 °C.

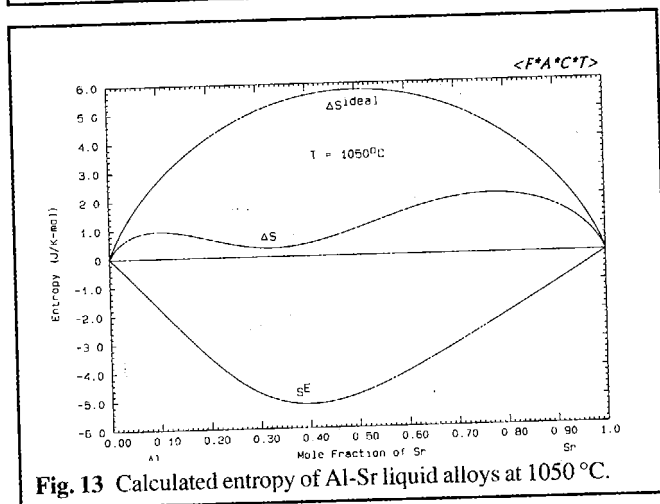


Fig. 13 Calculated entropy of Al-Sr liquid alloys at 1050 °C.

shows the calculated entropy of the liquid at 1050 °C. In Fig. 11 and 12, the calculated enthalpy of mixing and $\ln \gamma_{Sr}$ are compared with the experimental points.

For calculation, all solid phases were presumed stoichiometric, although a very small solubility of Sr in Al was observed (~0.0077 at.% Sr in Al at 600 °C) as discussed by [89Alc]. [89Alc] rejected the ~5.5 at.% solubility of Al in Sr reported by [79Vak]; this conclusion is accepted in the present work. The calculated eutectic in Fig. 10 is in good agreement with experimental results of [83Han], [85Sat], and [86Clo], so the assumption of only a very small solubility of Sr in Al is justified. The atomic radii ratio is ~1.5 also suggesting a very small solubility of Al in Sr. More precise measurements should be made for the determination of the solubility of Al in Sr. [39Now] reported that the "AlSr" phase (Al_7Sr_8 as demonstrated by [83For]) decomposed below 300 °C. The calculated entropies of fusion of the intermetallic compounds are 17.7 J/K · g-atom for Al_4Sr , 16.1 J/K · g-atom for Al_2Sr , and 9.0 J/K · g-atom for Al_7Sr_8 . These are reasonable values.

The activity of Sr in the liquid alloy was measured by [74Bur], [79Vak], and [91Sri]. See Fig. 12 for $\ln \gamma_{Sr}$. Experiments of [91Sri], who used Knudsen and pseudoisopiestic methods at 1050 °C, were more complete and detailed. [74Bur] also measured activities with the Knudsen weight loss method between 850 and 1100 °C. Estimations of the thermodynamic properties of solids were given by [84Kha] but were judged incomplete and obsolete by [89Alc]; this conclusion is accepted in the present evaluation.

Optimization of Thermodynamic Properties and Phase Diagram

The calculated optimized Al-Sr phase diagram is shown in Fig. 9 and 10. The optimized properties of compounds are listed in Table 1. The optimized parameters for the liquid phase are listed in Eq 6, and calculated invariant points are presented in Table 2.

The liquid phase was modeled using the results of [83Som] and [85Esi] for the enthalpy of mixing and the activity of Sr given by [91Sri]. As discussed earlier, no experimental liquidus points of γ_{Sr} were used in the optimization process. Figure 13

The Mg-Sr System

Equilibrium Diagram

[86Nay] reviewed the Mg-Sr system. From their conclusions, the stable phases are: the liquid, the (Mg)cph solid solution, the (γ_{Sr})bcc solid solution, the (α_{Sr})fcc solid solution, and four intermetallic compounds— $Mg_{17}Sr_2$, $Mg_{38}Sr_9$, $Mg_{23}Sr_6$, and Mg_2Sr .

In the present evaluation (Fig. 14 and 15), the region between 10 and 20 at.% Sr and the liquidus of the Sr allotropes differ from the phase diagram proposed by [86Nay]. [86Nay] suggested that the liquidus points of α_{Sr} and γ_{Sr} correspond to the experimental points of [47Ray]. In the present evaluation, experimental points of [73Bro] are considered because they respect the theoretical limiting slope of the liquidus of Sr at $X_{Sr} \rightarrow 1$ (Eq 12) if a negligible solubility of Mg in Sr allotropes is present; moreover [73Bro] and [47Ray] are both Ph.D. theses produced

Table 2 Calculated Special Points of the Al-Mg-Sr System

Reaction	°C	Phase	Temperature,			Reaction type
			Al	Mg	Sr	
Pure components						
L ↔ (Al)fcc.....	660.3	...	100	0	0	Melting
L ↔ (Mg)cph.....	649.8	...	0	100	0	Melting
L ↔ γSr.....	769.0	...	0	0	100	Melting
γSr ↔ αSr.....	556.5	...	0	0	100	Allotropic
Al-Mg system						
L ↔ (Al)fcc + β.....	450 ± 1	L	63.8	36.2	0	Eutectic
		(Al)fcc	81.8	18.2	0	Eutectic
L ↔ β.....	451 ± 1	L	Congruent
L ↔ β + (γ).....	449 ± 2	L	57.8	42.2	0	Eutectic
		(γ)	53.7	46.3	0	Eutectic
L ↔ (γ).....	460 ± 1	...	45.4	54.6	0	Congruent
L ↔ (γ) + (Mg)cph.....	437 ± 1	L	29.9	70.1	0	Eutectic
		(γ)	37.4	62.6	0	Eutectic
		(Mg)cph	11.3	88.7	0	Eutectic
β + (γ) ↔ R.....	401 ± 5	(γ)	50.7	49.3	0	Peritectoid
R ↔ β + (γ).....	303 ± 10	(γ)	46.3	53.7	0	Eutectoid
Al-Sr system						
L ↔ (Al)fcc + Al ₄ Sr.....	654 ± 1	L	99.13	0	0.87	Eutectic
		(Al)fcc	100	0	0	Eutectic
L ↔ Al ₄ Sr.....	1025 ± 5	Congruent
L ↔ Al ₄ Sr + Al ₂ Sr.....	920 ± 2	L	68.4	0	31.6	Eutectic
L ↔ Al ₂ Sr.....	922 ± 2	Congruent
L + Al ₂ Sr ↔ Al ₇ Sr ₈	664 ± 3	L	44.4	0	55.6	Peritectic
L ↔ Al ₇ Sr ₈ + γSr.....	580 ± 2	L	31.1	0	68.9	Eutectic
Al ₇ Sr ₈ ↔ Al ₂ Sr + αSr.....	345 ± 25	Eutectoid
Mg-Sr system						
L ↔ (Mg)cph + Mg ₁₇ Sr ₂	587 ± 3	L	0	93.8	6.2	Eutectic
		(Mg)cph	0	100	0	Eutectic
L ↔ Mg ₁₇ Sr ₂	606 ± 2	Congruent
L ↔ Mg ₁₇ Sr ₂ + Mg ₃₈ Sr ₉	591 ± 2	L	0	85.0	15.0	Eutectic
L + Mg ₂₃ Sr ₆ ↔ Mg ₃₈ Sr ₉	599 ± 3	L	0	82.9	17.1	Peritectic
L + Mg ₂ Sr ↔ Mg ₂₃ Sr ₆	603 ± 3	L	0	81.3	18.7	Peritectic
L ↔ Mg ₂ Sr.....	680 ± 3	Congruent
L ↔ Mg ₂ Sr + αSr.....	426 ± 2	L	0	30.1	69.9	Eutectic
Al-Mg-Sr system						
L ↔ Mg ₂ Sr + Al ₂ Sr.....	565	L	26.1	40.6	33.3	Saddle
L ↔ (Mg)cph + Al ₄ Sr.....	538	L	17.0	79.6	3.4	Saddle
		(Mg)cph	4.6	95.4	0	Saddle
L + Mg ₂ Sr ↔ Al ₂ Sr + Mg ₂₃ Sr ₆	521	L	17.2	62.5	20.3	Peritectic
L + Mg ₂₃ Sr ₆ ↔ Mg ₃₈ Sr ₉ + Al ₂ Sr.....	517	L	16.7	64.3	19.0	Peritectic
L ↔ Mg ₁₇ Sr ₂ + Al ₂ Sr.....	509	L	15.7	68.3	15.9	Saddle
L ↔ Mg ₃₈ Sr ₉ + Mg ₁₇ Sr ₂ + Al ₂ Sr.....	509	L	15.6	67.6	16.8	Eutectic
L + Al ₄ Sr ↔ (Mg)cph + Al ₂ Sr.....	501	L	16.4	73.4	10.2	Peritectic
		(Mg)cph	1.5	98.5	0	Peritectic
L ↔ (Mg)cph + Mg ₁₇ Sr ₂ + Al ₂ Sr.....	496	L	15.1	73.9	11.0	Eutectic
		(Mg)cph	1.2	98.8	0	Eutectic
L ↔ (γ) + Al ₄ Sr.....	460	L	45.4	54.6	0.0306	Saddle
		(γ)	45.4	54.6	0	Saddle
L ↔ β + Al ₄ Sr.....	451	L	61.5	38.4	0.0156	Saddle
L ↔ (Al)fcc + Al ₄ Sr + β.....	450	L	63.8	36.2	0.0147	Eutectic
		(Al)fcc	81.8	18.2	0	Eutectic
L + Al ₂ Sr ↔ Al ₇ Sr ₈ + Mg ₂ Sr.....	450	L	17.8	23.8	58.4	Peritectic
L ↔ Al ₄ Sr + (γ) + β.....	449	L	57.8	42.1	0.0156	Eutectic
		(γ)	53.7	46.3	0	Eutectic
L ↔ (Mg)cph + (γ) + Al ₄ Sr.....	437	L	29.9	70.1	0.0354	Eutectic
		(Mg)cph	11.3	88.7	0	Eutectic
		(γ)	37.4	62.6	0	Eutectic
L ↔ Al ₇ Sr ₈ + Mg ₂ Sr + αSr.....	412	L	4.8	27.3	67.9	Eutectic

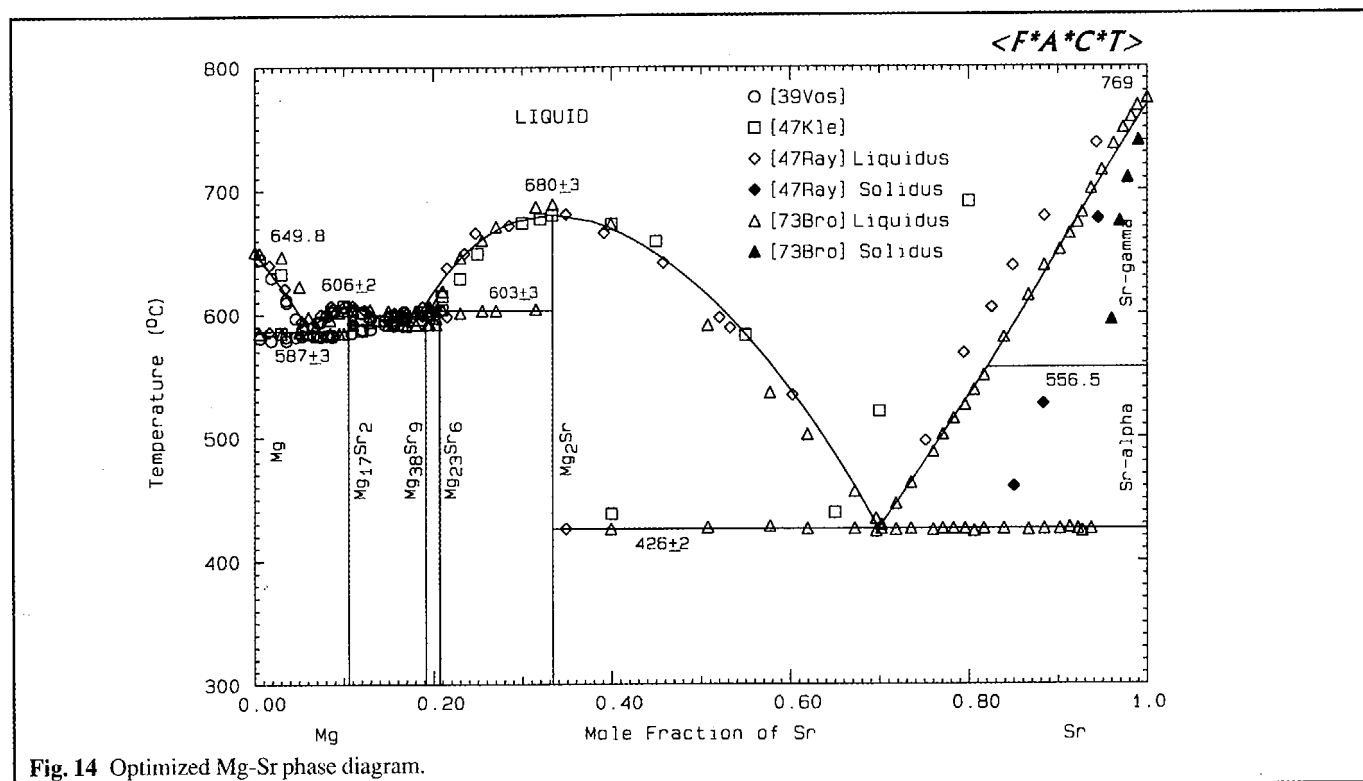


Fig. 14 Optimized Mg-Sr phase diagram.

in the same laboratory under the supervision of F.A. Kanda. Hence, it may be inferred that [73Bro] is an improvement of the work of [47Ray].

The 10 to 20 at.% Sr region of the phase diagram is subject to more than one interpretation as stated by [86Nay]. The interpretation of [86Nay] shows a large asymmetry for the liquidus of Mg₁₇Sr₂ on either side of the stoichiometric compound, which is thermodynamically very unlikely. In addition, since all four compounds of this system are close in composition and no decomposition at lower temperature is observed, then their entropies of formation must be approximately equal. The interpretation of [86Nay] requires that the entropy of formation of Mg₁₇Sr₂ and Mg₂₃Sr₆ be high in order to produce flat liquidus curves. However, this results in a calculated eutectoid decomposition at relatively high temperature of one or more of the four compounds. In our interpretation of this region, the assumption was made that all four intermetallic compounds are stable at room temperature because no decomposition was observed experimentally. For these reasons, then, Mg₃₈Sr₉ is associated with a peritectic reaction (L+Mg₂₃Sr₆→Mg₃₈Sr₉) at 599 °C and with a eutectic reaction L→Mg₁₇Sr₂+Mg₃₈Sr₉ at 591 °C. [86Nay] proposed this interpretation as an alternative.

Crystal Structures and Lattice Parameters

All necessary information is described in the review of [86Nay].

Thermodynamics

The available thermodynamic data include the enthalpy of mixing of the liquid measured by [77Som] at 807 °C (Fig. 16). The minimum of the enthalpy of mixing is between 30 and 35 at.% Sr. [80Som] measured the activity of Mg in liquid at 757

°C using a modified Ruff boiling point method (Fig. 17) and estimated that the minimum of the excess entropy of the liquid is approximately $-2 \text{ J/K} \cdot \text{mol}$ at 30 at.% Sr. [64Kin] measured the enthalpy of formation of Mg₂Sr from solid Mg and Sr. A tin solution calorimeter was used to obtain a value of -21.35 kJ/mol .

Optimization of Thermodynamic Properties and Phase Diagram

The calculated optimized Mg-Sr phase diagram is shown in Fig. 14 and 15. The optimized Gibbs energies of the compounds are shown in Table 1. The optimized parameters for the liquid phase are listed in Eq 7, and calculated invariant points are presented in Table 2.

The liquid was modeled by using the experimental values of enthalpy of mixing of [77Som], the activity of Mg in the liquid of [80Som], and the liquidus curves of Mg and Sr allotropic phases of [73Bro]. Figure 16 shows the calculated enthalpy of mixing at 807 °C along with the experimental results of [77Som]. The calculated entropy of the liquid at 807 °C is presented in Fig. 18; the excess entropy curve has a minimum of -2.45 at 32 at.% Sr in agreement with the evaluation of [80Som]. Figure 18 shows the calculated activity of Mg in liquid alloys at 757 °C in comparison with results of [80Som].

All solid phases were assumed to be stoichiometric for the calculations. This is valid for intermetallic compounds and for the Mg solid phase where the solubility of Sr is stated to be negligible by [39Vos], [47K1e], and [47Ray]. [47Ray] and [73Bro] observed a nonnegligible solubility of Mg in Sr (14.5 at.% and 6.5 at.% at the eutectic temperature, respectively). Only a very small solubility of Mg in Sr is predicted by the difference in

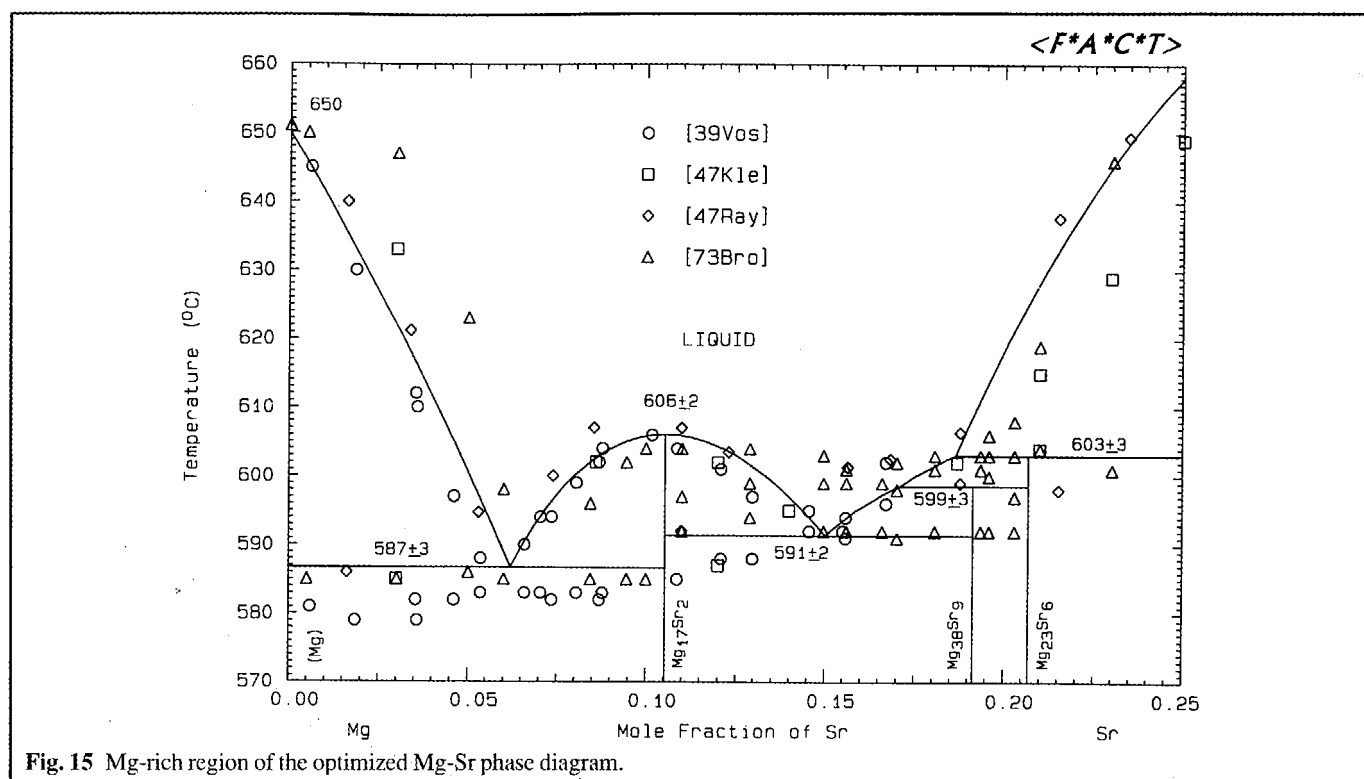


Fig. 15 Mg-rich region of the optimized Mg-Sr phase diagram.

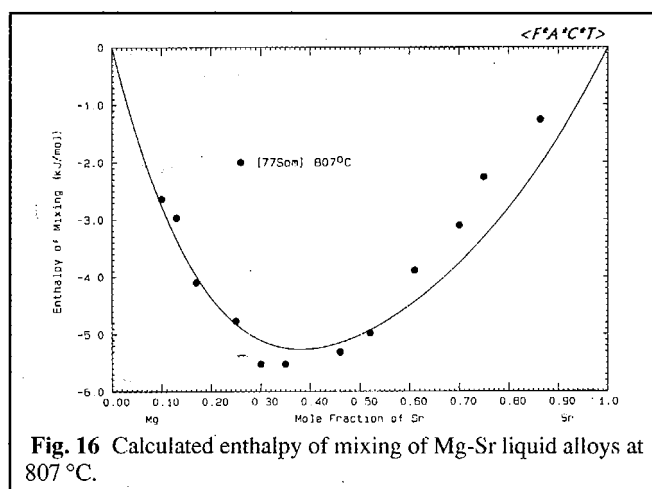


Fig. 16 Calculated enthalpy of mixing of Mg-Sr liquid alloys at 807 °C.

atomic radii (more than 34%) and by observation of Mg solubility in similar systems (Mg-Ca and Mg-Ba). The calculated parameters for the solid intermetallic phases give no decomposition at room temperature, and the calculated entropies of fusion of these phases are 7.87 J/K · g-atom for $Mg_{17}Sr_2$, 9.31 J/K · g-atom for $Mg_{38}Sr_9$, 9.51 J/K · g-atom for $Mg_{23}Sr_6$, and 11.34 J/K · g-atom for Mg_2Sr , which are reasonable values.

The Al-Mg-Sr System

Equilibrium Diagram

[80Mak] studied solid solubility at 400 °C in the Mg-rich region of the system by X-ray analysis, micrography, and microhardness. A ternary compound was reported and named "X,"

of unknown stoichiometry, in equilibrium with (Mg)cph, "Mg₉Sr" ($Mg_{17}Sr_2$), and $Al_{12}Mg_{17}$ (γ). [81Mak1] performed micrography, X-ray analysis, and microhardness tests on 200 alloys of Al, Mg, and Sr quenched from 400 °C. Large solubilities were observed in the solid phases. Another ternary compound was reported (different from "X") with a stoichiometry of " $Al_{34}Mg_6Sr_{60}$ " (Al_6MgSr_{10}). [80Mak2] studied the liquidus surfaces in Al-rich and Mg-rich regions of the Al- Al_4Sr - $Al_3Mg_2(\beta)$ and Mg- $Mg_{17}Sr_2$ - $Al_{12}Mg_{17}(\gamma)$ systems. The authors considered $Mg_{17}Sr_2$ - $Al_{12}Mg_{17}(\gamma)$ and Al_4Sr - $Al_3Mg_2(\beta)$ as simple eutectic quasi-binary systems. A ternary eutectic was reported at 71 at.% Mg, 27 at.% Al, and 2 at.% Sr at 430 °C, and another at 35 at.% Mg, 63.6 at.% Al, and 1.4 at.% Sr at 445 °C. [82Mak1] studied pseudobinary sections in the ternary system. The system $Al_{12}Mg_{17}(\gamma)$ - $Mg_{17}Sr_2$ was considered quasi-binary with a eutectic at 32 mol% $Mg_{17}Sr_2$ at 438 °C, with a solubility at T_E of $Mg_{17}Sr_2$ in $Al_{12}Mg_{17}$ of ~3 mol% and of $Al_{12}Mg_{17}$ in $Mg_{17}Sr_2$ of 46 mol%. The system $Al_3Mg_2(\beta)$ - $Mg_{17}Sr_2$ was reported to be a simple eutectic type with eutectic at 38 mol% $Mg_{17}Sr_2$ at 439 °C, with a solubility at T_E of $Mg_{17}Sr_2$ in Al_3Mg_2 of ~5 mol%, and of Al_3Mg_2 in $Mg_{17}Sr_2$ of 36 mol%. The system Al_4Sr - Mg_2Sr was considered to be a simple eutectic type with eutectic at 40 mol% Al_4Sr at 610 °C, with a solubility at T_E of Mg_2Sr in Al_4Sr of 48 mol%, and of Al_4Sr in Mg_2Sr of 25 mol%. The system Al_4Sr - $Mg_{17}Sr_2$ was considered quasi-binary with a eutectic at 46 mol% Al_4Sr at 571 °C, with a solubility at T_E of Al_4Sr in $Mg_{17}Sr_2$ of 26 mol%, and of $Mg_{17}Sr_2$ in Al_4Sr of 35 mol%. The system Al_4Sr - $Al_3Mg_2(\beta)$ was reported to have one eutectic at 85 mol% Al_3Mg_2 at 446 °C, with a solubility at T_E of Al_3Mg_2 in Al_4Sr of 18 mol%, and of Al_4Sr in Al_3Mg_2 of 3 mol%. There is no mention of the "S" phase of [81Mak1] in this study. [82Mak2] used

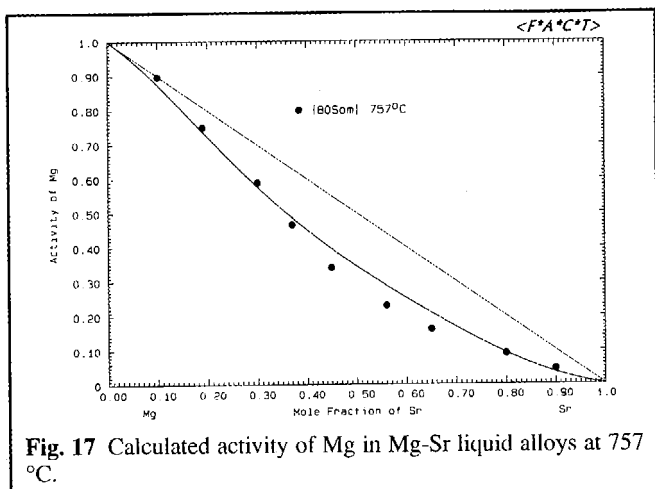


Fig. 17 Calculated activity of Mg in Mg-Sr liquid alloys at 757 °C.

their previous results to approximate the liquidus surface of the Sr-Al₄Sr-Mg₂Sr system by a simplex method from 19 judiciously chosen liquidus points. The ternary "S" phase was considered in this study.

Thermodynamics

No data are available for the liquid or the possible ternary phases.

Calculated Phase Diagram

The calculated Al-Mg-Sr phase diagram is shown in Fig. 19 and 20 considered are: the liquid, the (Al)fcc binary solid solution, the (Mg)cph binary solid solution, α Sr and γ Sr allotropic solid phases, the binary γ solid solution, and the stoichiometric intermetallic compounds— β , R, Al₄Sr, Al₂Sr, Al₇Sr₈, Mg₁₇Sr₂, Mg₃₈Sr₉, Mg₂₃Sr₆, and Mg₂Sr. Calculated ternary univariant points are listed in Table 2.

The thermodynamic properties of the liquid were estimated from the optimized binary parameters by the modified quasi-chemical model for the ternary liquid phase with the symmetric approximation [86Pel, 93Eri] with no adjustable ternary terms added since no ternary thermodynamic data are available.

No ternary solid phase was considered in the present evaluation because of the uncertainties related to them (existence, stability and homogeneity range, melting or decomposition temperature, etc.). The extensive solubilities between solid phases reported by [80Mak], [81Mak1], [81Mak2], and [82Mak1] seem unlikely for solids of such different crystal structures and stoichiometry. The three solid solutions in the Al-Mg system—(Al)fcc, (Mg)cph, and γ —were not extended into the ternary field due to a lack of pertinent information. Because of these uncertainties and because no measured ternary phase equilibrium data are available for comparison, no error estimates for the calculated ternary invariant temperatures are given in Table 2.

The liquidus surface of the calculated phase diagram is divided into thirteen primary crystallization fields: (Al)fcc, (Mg)cph, α Sr, γ Sr, (γ), β , Al₄Sr, Al₂Sr, Al₇Sr₈, Mg₁₇Sr₂, Mg₃₈Sr₉, Mg₂₃Sr₆, and Mg₂Sr. Liquidus surfaces of Al₄Sr, Al₂Sr, and

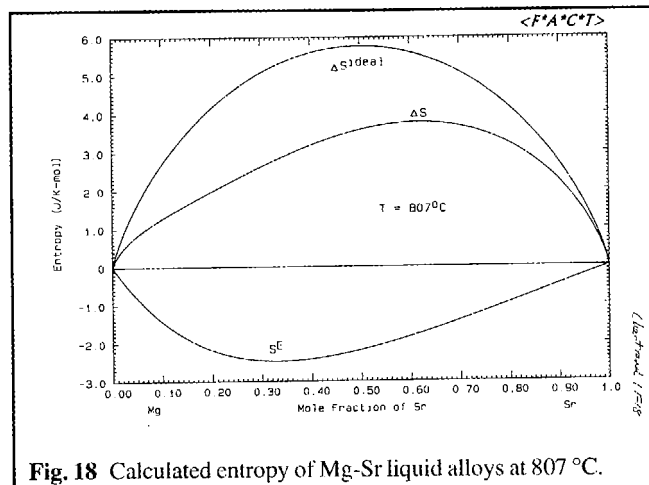


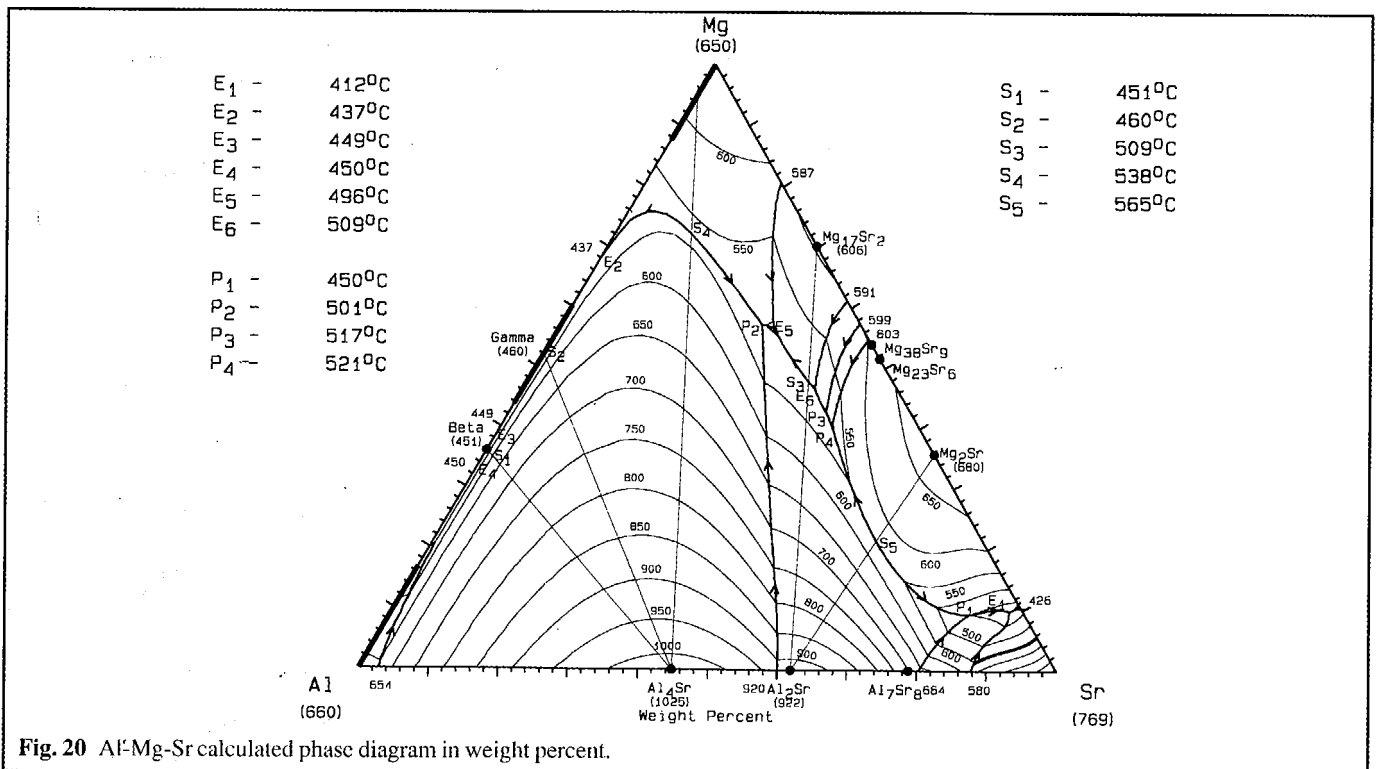
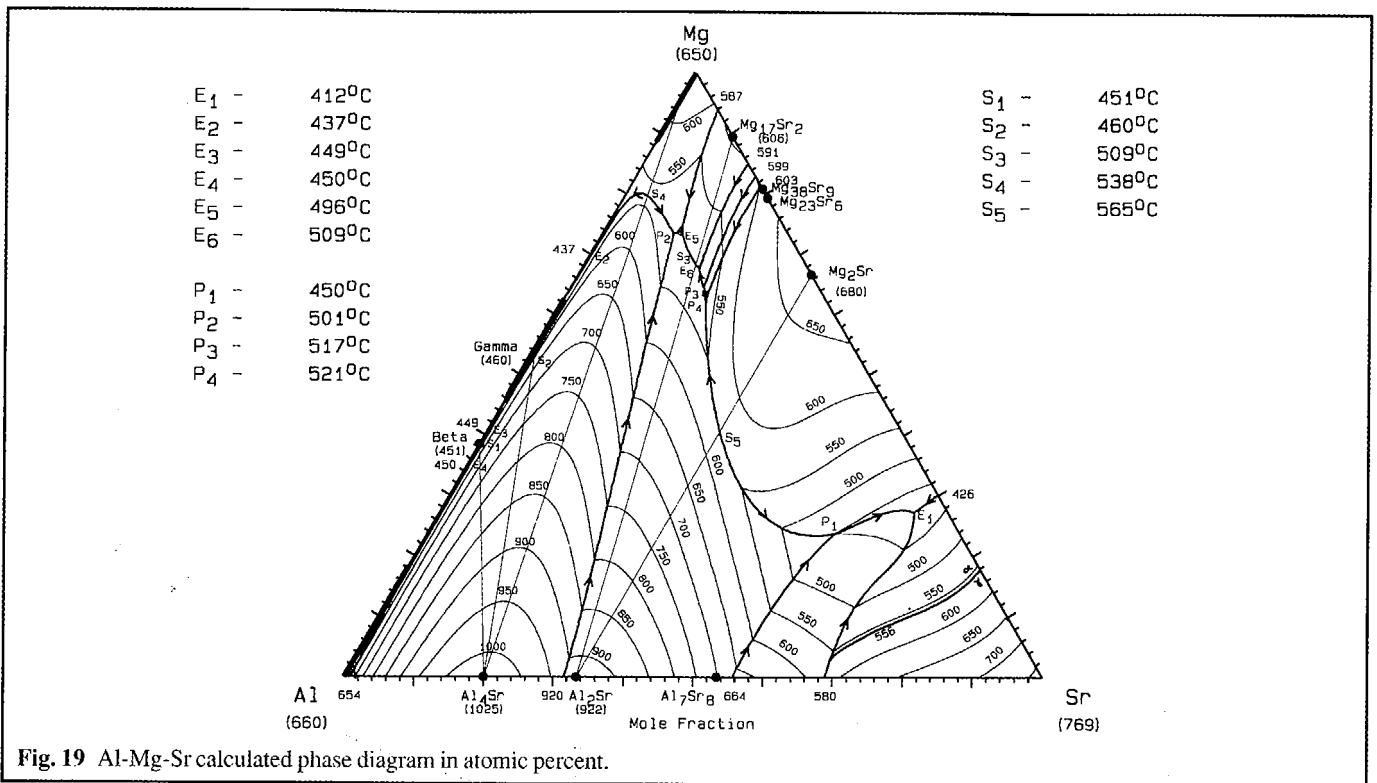
Fig. 18 Calculated entropy of Mg-Sr liquid alloys at 807 °C.

Mg₂Sr dominate the liquidus surface. Three quasi-binary systems are observed: Al₄Sr- β (it must be noted that the β phase was supposed stoichiometric; thus this system might actually not be quasi-binary), Al₂Sr-Mg₂Sr, and Al₂Sr-Mg₁₇Sr₂. Two systems, Al₄Sr-(γ) and Al₄Sr-(Mg)cph, are not quasi-binary but have a maximum on their common univariant line. Six ternary eutectic invariants and four ternary peritectic invariants are observed on the calculated liquidus surface.

The 400 °C isothermal section of [81Mak1] shows a triangulation involving Mg, Mg₁₇Sr₂, and the γ -phase. Given the low thermodynamic stabilities of these compounds relative to Al₄Sr and Al₂Sr, this seems unlikely. The 400 °C section of [81Mak1] also shows triangulations involving Al₄Sr in equilibrium with Mg₁₇Sr₂, Mg₂₃Sr₆, and Mg₂Sr. In the present optimization, these compounds are calculated to be in triangulations with Al₂Sr. This seemingly large difference actually only requires a moderate change to the ternary phase diagram. If the univariant line on Fig. 19 and 20, which starts in the Al-Sr binary and descends to point P₂, were to descend instead to a point on the univariant line between the Mg₂Sr and Al₂Sr fields just to the right of point P₄, then the triangulations of [81Mak1] would be reproduced. However, given the questions raised above regarding the results of [81Mak1] and the inconsistencies among the various publications of the same group [80Mak, 81Mak1, 81Mak2, 82Mak1, 82Mak2], it was decided not to attempt to reproduce the reported ternary sections by introducing ternary parameters to the liquid model or ternary solid solubilities. Rather, the calculated ternary phase diagram is presented as a starting point for future experimental verification and refinement.

Conclusions

A self-consistent set of model equations for the Gibbs energy of the phases is required to permit the calculation of the ternary Al-Mg-Sr phase diagram. Hence, the present evaluators critically evaluated and reoptimized the thermodynamic properties of the binary Al-Mg, Al-Sr, and Mg-Sr systems with all available data in order to calculate the Al-Mg-Sr phase diagram



from these binary parameters. Available thermodynamic data in the binary systems were reproduced within experimental errors, with a minimum number of coefficients. For the ternary system, this work is a first step to a more complete evaluation that could be extended by the availability of new experimental results and by the addition of other components like silicon,

manganese, etc. (for example, the Al-Mg-Si-C has already been optimized [94Ber]).

Acknowledgment

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