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*Indicates key paper.
#Indicates presence of a phase diagram.

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The S-Sn (Sulfur-Tin) System

32.06 118.69

By R.C. Sharma and Y.A. Chang
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Equilibrium Diagram

Three intermediate phases, SnS, Sn_2S_3, and SnS_2, are formed in the Sn-S system. SnS and SnS_2 melt congruently, whereas Sn_2S_3 melts peritectically to a liquid and SnS_2. There are two liquid miscibility gaps, one in the Sn-SnS region, and one in the SnS_2-S region. The three eutectics are in the Sn-rich corner, between SnS and Sn_2S_3, and in the S-rich corner. All three intermediate phases undergo several phase transformations in the solid state. Figure 1 gives the assessed Sn-S phase diagram, and Fig. 2 gives the Sn-S system from 45 to 75 at.% S, showing the transformations in the intermediate phases in more detail.

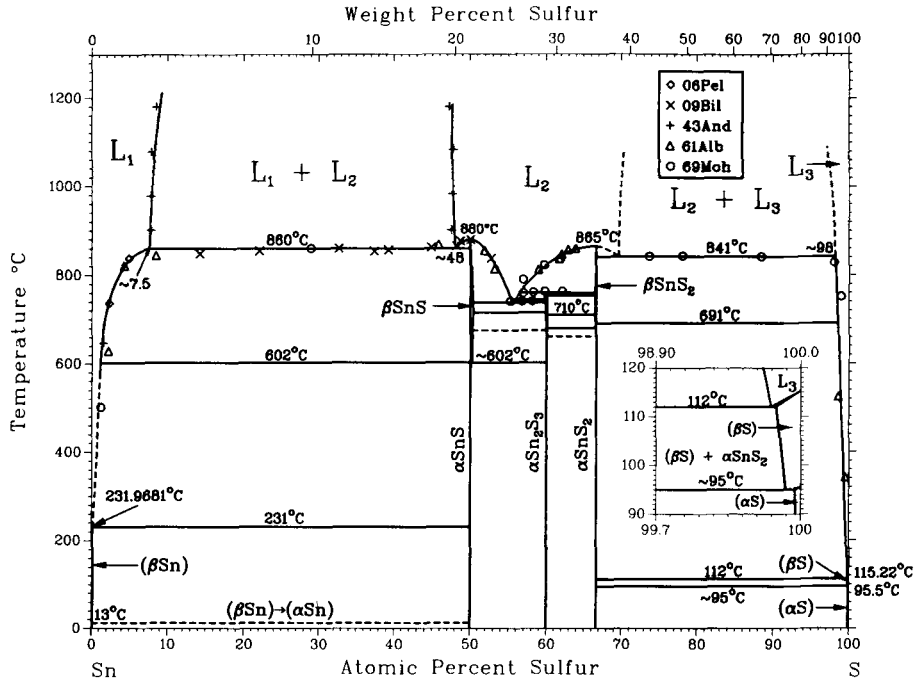
Liquidus. The melting points of Sn and S are 231.9681 and 115.22 °C, respectively [Melt]. The Sn-S liquidus was determined by [06Pel], [09Bil], [43And], [61Alb1],

Table 1 Invariant Reactions in the Sn-S System

Reaction	Compositions of the respective phases, at.% S			Temperature, °C	Reaction type	Reference
$L_1 \rightleftarrows (Sn) + \alpha SnS$	···	···	50	~231	Eutectic	[Assessed]
$L_2 \rightleftarrows L_1 + \beta SnS$	~48	~7.5	50	858	Monotectic	[43And]
	···	···	50	860		[61Alb1, 69Moh]
	~48	···	50	855		[66Kar]
	~48	~7.5	50	860		[Assessed]
$L_2 \rightleftarrows \beta SnS + \gamma Sn_2S_3$	55	50	60	740	Eutectic	[61Alb1]
	55	50	57(a)	705		[66Kar]
	55.6	≥50	≤60	738		[69Moh, Assessed]
$L_2 + \beta SnS_2 \rightleftarrows \delta Sn_2S_3$	~59	66.67	60	745	Peritectic	[66Kar]
	~56.5	66.67	60	760		[69Moh]
	56.5	66.67	60	760		[Assessed]
$L_2 \rightleftarrows \beta SnS_2 + L_3$	~70	66.67	~98	841	Monotectic	[69Moh, Assessed]
$L_3 \rightleftarrows \alpha SnS_2 + (\beta S)$	≤100	66.67	<100	112	Eutectic	[69Moh, Assessed]

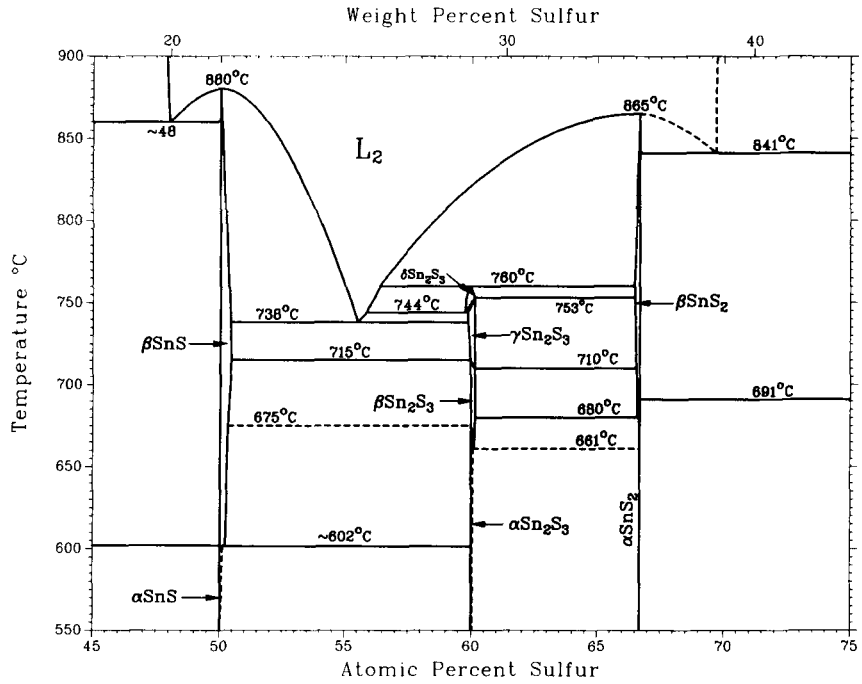
(a) [66Kar] found Sn_3S_4 to be a stable phase in the Sn-S system and suggested a eutectic reaction, $L_2 \rightleftarrows SnS + Sn_3S_4$ (also see text).

Fig. 1 Assessed Sn-S Phase Diagram



R.C. Sharma and Y.A. Chang, 1986.

Fig. 2 Sn-S Phase Diagram Between 45 and 75 at.% S



R.C. Sharma and Y.A. Chang, 1986.

Table 2 Sn-S Crystal Structure Data

Phase	Homogeneity, at.% S	Pearson symbol	Space group	Strukturbericht designation	Prototype	Reference
βSn	0	*tI*4	$I4_1/amd$	*A*5	βSn	[Pearson2]
αSnS	50	*oP*8	*Pnma*	*B*16	GeS	[Pearson2]
αSn_2S_3	60	*oP*20	*Pnam*	···	···	[Pearson2]
αSnS_2	66.67	*hP*3	$P\bar{3}m1$	*C*6	CdI_2	[Pearson2]
βS	~100	*mP*48	$P2_1/a$	···	···	[Pearson2]
αS	~100	*oP*128	*Fddd*	···	···	[Pearson2]
Metastable phases						
SnS (thin film)	50	*cF*8	$F\bar{4}3m$	*B*3	ZnS (sphalerite)	[Pearson2]
Sn_3S_4	57.14	Tetragonal	···	···	···	[57Bok, 66Kar]

Table 3 Sn-S Lattice Parameter Data

Phase	Composition, at.% S	Lattice parameters, nm			Reference
		a	*b*	*c*	
βSn	0	0.58315	···	0.31814	[Pearson2]
αSnS	50	1.120	0.399	0.434	[Pearson2]
αSn_2S_3	60	0.8864	1.4020	0.3747	[62Moo, Pearson2]
		0.8881	4.2145	0.3762	[66Kar]
αSnS_2	66.67	0.3639	···	0.5868	[28Oft]
		0.3646	···	0.5880	[Pearson2]
βS	100	1.092	1.098	1.104(a)	[Pearson2]
αS	100	1.0465	1.2866	2.4486	[Pearson2]
Metastable phases					
SnS (thin film)	50	0.5445	···	···	[Pearson2]
Sn_3S_4	57.14	0.7553	···	0.8383	[57Bok]

(a) $\beta = 83° 16'$.

[66Kar], and [69Moh]. [43And] also determined the liquid miscibility gap in the Sn-SnS region. The data are shown in Fig. 1.

Invariant Reactions. Data for the invariant reactions, along with the assessed values, are given in Table 1.

SnS, Sn_2S_3, and SnS_2 Phases. SnS exists in two allotropic forms. The low-temperature form, αSnS, transforms to the high temperature form, βSnS at ~602 °C [58Orr, 66Kar, 69Moh]. αSnS is essentially stoichiometric, whereas βSnS can dissolve excess S. It has a maximum S content of 50.5 at.% S at about 700 °C in equilibrium with Sn_2S_3 [69Moh]. βSnS melts congruently, and its melting point was reported as: 880 °C [06Pel, 09Pel], 870 °C [08Bil], 882 °C [09Bil], 880 °C [58Orr], 881.5 °C [61Alb1], 875 °C [66Kar], and 880 °C [69Moh]. The melting point of βSnS as 880 °C is preferred.

[69Moh] found four forms of Sn_2S_3 between room temperature and its peritectic melting point. These are designated α, β, γ, and δSn_2S_3 in Fig. 2. The low-temperature form, αSn_2S_3, is essentially stoichiometric, whereas the high-temperature forms, β, γ, and δSn_2S_3, were found to dissolve some S as well as Sn [69Moh]. The peritectic melting temperature of Sn_2S_3 is given as 745 °C by [66Kar] and 760 °C by [69Moh]. A peritectic temperature of 760 °C is preferred.

According to [69Moh], the SnS_2 phase exists in two allotropic forms, αSnS_2 (low-temperature form) and βSnS_2 (high-temperature form). αSnS_2 is essentially stoichiometric, whereas βSnS_2 is slightly Sn-rich at high temperatures. βSnS_2 melts congruently at 870 °C, according to [61Alb1], and at 865 °C, according to [69Moh]. The melting point of 865 °C is preferred.

Figure 2 gives the phase relationships and transformation temperatures for the different forms of the intermediate phases in the Sn-S system [69Moh].

Apart from the three intermediate phases, SnS, Sn_2S_3, and SnS_2, [37Ger] obtained Sn_4S_5 as an intermediate product during the thermal decomposition of SnS_2 ($SnS_2 \rightarrow Sn_2S_3 \rightarrow Sn_4S_5 \rightarrow SnS$), and [57Bok], [61Alb1], [66Kar], and [67Kar] reported the existence of Sn_3S_4. [71Bar], from their Mössbauer effect studies, concluded that only SnS and SnS_2 are formed. [69Moh], in their extensive investigation of the Sn-S system by X-ray diffraction, optical microscopy, and differential thermal analysis, found SnS, Sn_2S_3, and SnS_2 to be the only stable intermediate phases. Based on the work of [69Moh], SnS, Sn_2S_3, and SnS_2 are assessed as the stable intermediate phases in the Sn-S system; however, Sn_4S_5 and Sn_3S_4 may exist as metastable phases.

Solid Solubilities. The mutual solid solubilities of Sn and S are negligible and have not been determined accurately. S in the solid state exists in two allotropic forms. αS is stable up to 95.5 °C, where it transforms to βS. βS melts at 115.22 °C. The presence of Sn in S lowers the melting point of βS, as well as the $\beta S \rightleftarrows \alpha S$ transition temperature [69Moh] (see insert in Fig. 1). βSn transforms to αSn at 13 °C.

Crystal Structures and Lattice Parameters

Crystal structures and lattice parameters for the phases in the Sn-S system are given in Tables 2 and 3, respec-

tively. In addition, [61Alb1] listed powder diffraction data for Sn_2S_3 and Sn_3S_4 from the literature, and [69Moh] reported powder diffraction data for the various high-temperature modifications of SnS, Sn_2S_3, and SnS_2.

Thermodynamics

Liquid Alloys. [61Che] measured the P_{H_2S}/P_{H_2} ratio in equilibrium with dilute solutions of S in liquid Sn in the temperature range 500 to 700 °C. They expressed their results as:

$$k' = \frac{P_{H_2S}}{P_{H_2} \times \text{at.\%S}} \qquad \text{(Eq 1)}$$

where P_{H_2S} and P_{H_2} are the partial pressures of H_2S(g) and H_2(g) in the gas, respectively, in equilibrium with the liquid solution. They obtained:

$$\log k' = -\frac{735}{T} - 1.461 \qquad \text{(Eq 2)}$$

where T is in K.

SnS, Sn_2S_3, and SnS_2 Phases. The Gibbs energy of formation of SnS was measured by [25Jel], [39Kap], [51Sud], [55Ric], [65Rau], and [66Esp]. [74Mil] assessed these data and suggested $\Delta_f H^0$(SnS, 298 K) to be −108 kJ/mol. [58Orr] and [74Bla] measured the heat content of SnS as a function of temperature. [58Orr], based on their heat content measurements, found the enthalpy of transformation for the αSnS to βSnS transition to be 670 J/mol at 602 °C, and the enthalpy of fusion of βSnS to be 31.6 kJ/mol at 880 °C. Table 4 summarizes thermodynamic data for SnS. The Gibbs energy of formation for the SnS phase, given in Table 4, is obtained from $\Delta_f H^0$(SnS, 298 K) selected by [74Mil] and $[G^0(T) - H^0(298)]/T$ values from [74Mil] for the SnS phase, from [71Stu] for S and from [Hultgren, E] for Sn.

Table 4 Thermodynamic Data for the SnS(cd) Phase

$Sn(L) + \frac{1}{2}S_2(g) \rightleftarrows \alpha SnS(cd)$

$\Delta G^0 = -173\,170 + 92.222\,T$ J/mol
$500 < T < 875$ K

$\alpha SnS(cd) \rightleftarrows \beta SnS(cd)$

$\Delta_{tr}H = 670$ J/mol
$T_{tr} = 875$ K

$Sn(L) + \frac{1}{2}S_2(g) \rightleftarrows \beta SnS(cd)$

$\Delta G^0 = -172\,500 + 91.456\,T$ J/mol
$875 < T < 1153$ K

$\beta SnS(cd) \rightleftarrows SnS(L)$

$\Delta_{fus}H = 31\,600$ J/mol
$T_{fus} = 1153$ K

[70Gla2] measured the volume change during the melting of SnS and [70Gla1] determined the viscosity of SnS liquid. [61Alb2] investigated the electrical properties of the SnS phase.

The Gibbs energy of formation of Sn_2S_3 was determined by [37Ger], [65Rau], and [67Kar]. [65Rau] obtained $\Delta_f H^0(Sn_2S_3, 298\text{ K})$ for the formation of Sn_2S_3 to be −297.5 kJ/mol; however, [74Mil] assessed $\Delta_f H^0(Sn_2S_3, 298\text{ K})$ to be −263.5 kJ/mol, from the data of [37Ger] and [67Kar]. $S^0(Sn_2S_3, 298\text{ K})$ was estimated as 164 J/mol·K by [74Mil].

The Gibbs energy of formation of SnS_2 was determined by [37Ger] and [67Kar]. From these measurements, [74Mil assessed $\Delta_f H^0(SnS_2, 298\text{ K})$ for the formation SnS_2 as −153.6 kJ/mol. $S^0(SnS_2, 298\text{ K})$ for SnS_2 is 87.5 J/mol·K [74Mil].

[37Ger] and [67Kar] also measured the dissociation pressure of metastable Sn_3S_4. From these data, [74Mil] obtained $\Delta_f H^0(Sn_3S_4, 298\text{ K})$ for the formation of Sn_3S_4 as −370 kJ/mol. $S^0(Sn_3S_4, 298\text{ K})$ for Sn_3S_4 was estimated as 243.5 J/mol·K by [74Mil].

[74Mil] listed $[G^0(T) - H^0(298)]/T$ values for SnS, Sn_3S_4, Sn_2S_3, and SnS_2.

Vapor Pressure. SnS vaporizes with very little decomposition. The vapor pressure of SnS(g) over SnS(cd) was measured by [52Hsi], [55Ric], [60Klu], [62Col], [65Rau], and [79Gal]. All these data are summarized in Table 5, along with the assessed values. The assessed values are based on thermodynamic data selected by [74Mil].

[62Col] reported the presence of Sn_2S_2(g) species, in addition to SnS(g), on vaporization of SnS(cd). They obtained $\Delta_r H^0(Sn_2S_2, g \rightarrow 2\text{ SnS, g}, 298\text{ K})$ as 203.8 kJ/mol. $S^0(Sn_2S_2, g, 298\text{ K}) = 314$ J/mol·K was assumed.

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Table 5 Partial Pressure of SnS(g) in Equilibrium with SnS(cd)

Reference	Method	$\log(p, \text{bar}) = -A/T + B + C \log T$ A	B	C	Temperature range, K
[52Hsi]	Gravimetric	8380	3.853	...	...
[55Ric]	Transpiration	10470	7.094	...	...
[60Klu]	Boiling point	9980	6.676	...	...
[62Col]	Mass spectrometry	10625	7.356	...	...
[65Rau]	Transpiration, Knudsen effusion	11194	14.518	−2.19	700 to 875
		11160	14.728	−2.19	875 to 1154
[79Gal]	...	9977	6.628	...	
[Assessed]	...	10829	7.496	...	<875
		10794	7.456	...	875 to 1153
		9144	6.025	...	>1153

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*Indicates key paper.
#Indicates presence of a phase diagram.

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