

On Phase Equilibria of Sn-Sr and Mn-Sn-Sr Systems

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Abstract

Sn-Sr system is critically evaluated and the most reliable experimental data are incorporated in thermodynamic modeling. Thermodynamic properties of the binary liquid solution are estimated using the modified quasichemical model (MQM). The optimized Sn-Sr phase diagram and the corresponding thermodynamic properties are found to be in fair agreement with the experimental data in the previous related literature. For all the compounds in Sn-Sr system, a comparison between the calculated enthalpy of formation and that calculated by Miedema and first-principles has been reported. Combining Sn-Sr with the self-established thermodynamic database of Mn-Sr and Mn-Sn was used to calculate liquidus projection of Mn-Sn-Sr ternary system.

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1. Introduction

Magnesium alloys have a great potential for structural applications because of their significant weight savings, superior damping capacity and good castability [1]. Therefore, the automotive industry leads the way in the growing interest in Mg alloys in order to decrease fuel consumption and reduce emissions [1-3]. Recently, research activities have been carried out to improve the elevated temperature properties of the magnesium alloys through precipitation hardening and/or solid solution hardening [4-8]. The most common magnesium alloys for automotive industry contain aluminum as a major alloying element (AZ91 and AM60). Aluminum improves the mechanical properties at room temperature; however the loss of the mechanical strength at service temperature (above 120°C) of Mg alloys is reported due to the discontinuous precipitation of γ -Mg₁₇Al₁₂ [9-15]. Alloying Mg with Ca, Zn, Sr, Mn and Sn are found to improve the elevated temperature properties of Al containing Mg alloys by delaying or inhibiting the formation of γ and/or precipitating of thermally stable compounds such as Al₂Ca or Al₄Sr intermetallic phase [9-19]. Strontium is an important element in multi-component magnesium and aluminum alloys. Alloying magnesium with strontium refines the microstructure and improves the mechanical properties. Moreover, Sr enhances the corrosion resistance of Mg and Al-based alloys [20-23]. On the other hand, tin

addition to magnesium based alloys has a potential to improve creep and corrosion resistance of these alloys. Tin is also a reasonably cheap element as compared to RE elements and has a low melting point and is known to improve castability [24-26]. Mn addition to Mg-Sr alloys enhances creep resistance by precipitation of α -Mn at the interdendritic regions and Mn dissolves in the Mg matrix and Mg₁₇Sr₂ phase [27, 28].

An understanding of thermodynamic characteristics and equilibrium phase diagram for magnesium and its alloys will allow a better control in developing and designing a new Mg-alloy with desired properties. The present study deals with thermodynamic modeling of the Mn-Sn-Sr system which is one of the important systems in Mg-Mn multi-component systems. Sn-Sr binary system will be critically evaluated and optimized using the available phase equilibrium and thermodynamic data. The latest published journal article on thermodynamic modeling of the Sn-Sr system used Bragg-Williams model [29]. In their work [29], the enthalpies of formation for the stoichiometric compounds at 0 K were computed by first-principles calculations. In the present study, this system will be optimized using the modified quasi-chemical model (MQM). The MQM is considered more physically sound than other models such as the associate solution model [30]. Sn-Sr and Mn-Sr binary systems have been optimized using most up-to-date experimental data. Each of the phases in any binary system has been critically assessed based on the thermodynamic properties.

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In this research article, the experimental phase diagram and thermodynamic properties of Sn-Sr binary system will be critically evaluated and the most reliable data will be used in optimization. Sn-Sr phase diagram will be optimized and combined with the self-established thermodynamic database of the Mn-Sn and Mn-Sr systems. The optimized parameters of the constituent binary sub-systems will be used to draw liquidus projections of the Mn-Sn-Sr ternary phase diagram.

2. Experimental data

2.1. Sn-Sr phase diagram

Ray [31] tracks the thermal history of twelve tin-strontium alloys within the composition range of 0-29.65 at.% Sr using thermal analysis and optical microscopy. In Ray's study, alloys were prepared by electrolysis of a mixture of sodium and strontium chlorides over molten tin in an iron crucible. During electrolysis, strontium reacted with iron crucible and the fused salts were rapidly volatilized. Therefore, Ray's [31] thermal data were in error and will not be used in the current optimization. Phase equilibria of thirteen tin-strontium alloys were investigated by Marshall and Chang [32] using DTA, XRD and optical microscopy from pure tin up to 65 at.% Sn. They reported that Sn_3Sr forms peritectically at $598 \pm 1^\circ\text{C}$ which is in contrast with the work of Ray who showed that Sn_3Sr melted congruently at 607°C and forming a eutectic at 26.5 at.% Sr and 580°C . Marshall and Chang also reported that Sn_4Sr is in equilibrium with Sn instead of Sn_5Sr as reported by Ray [31] and Sn_4Sr decomposes peritectically at $334 \pm 4^\circ\text{C}$. They [32] found that Sn_4Sr and Sn form an eutectic at $230 \pm 2^\circ\text{C}$ and 1 at.% Sr. Widra and Schafer [33] studied Sn-Sr phase diagram in the composition range of 35-100 at.% Sr using DTA and XRD. Five intermetallic compounds, which are Sr_2Sn , Sr_5Sn_3 , SrSn , SrSn_3 and SrSn_5 , were reported in their phase diagram, of which only the first four phases were melted congruently. The assessment proposed by Massalski *et al.* [34] is the summary of the work of Marshall and Chang [32], and Widra and Schafer [33]. In the composition range of 11.6 to 43 at.% Sr, Hoffmann *et al.* [35,36] tracked phase transformation temperatures during cooling and heating for seven Sn-Sr alloys using DTA. Phase stability of Sr_3Sn_5 intermetallic compound were shown from the works of Hoffmann *et al.* [35,36] and Zürcher *et al.* [37]. The phase equilibria over the whole composition range in the Sn-Sr system were investigated by Palenzona and Pani [38] using XRD, DTA and optical composition. According to their work, five intermetallic compounds were confirmed namely; Sr_2Sn , SrSn , Sr_5Sn_3 , SrSn_3 and SrSn_4 , where, Sr_2Sn and SrSn melt congruently. In their work, three eutectic reactions were detected in the Sr-Sn system and SrSn_4 was confirmed instead of SrSn_5 as reported by Ray [31] and Widra and Schafer [33]. Experimental phase diagram data of Palenzona and Pani [38] and Hoffmann and Fassler [37] were in agreement. Those experimental data were the most recent and in good agreement; therefore, they will be used in the current optimization. Crystal structures of the compounds in the Sn-Sr system were reported by [35-37, 39-45].

Limited thermodynamic data for the Sn-Sr system could be found in previous literature. Strontium is highly reactive and, hence, it is very difficult to handle the alloys during high temperature experiments. However, enthalpy of mixing of the Sn-Sr liquid was measured by Esin *et al.* [46] using high temperature calorimeter at 1773 K within the composition range from 0 – 50 at.% Sr. Enthalpy of mixing from Esin *et al.* [46] will be used in the current optimization. Morozova *et al.* [47] estimated the enthalpy of formation of SrSn_2 compound to be -82.7 ± 1 kJ/mole.atom at 25°C . Zhao *et al.* [29] calculated the enthalpy of formation for all compounds in the Sn-Sr system using first-principles method and Miedema model and these data will be used only for comparison. Activity of Sr in Sn-Sr system was measured by Klebanov *et al.* [48] at 900 K.

2.2. Mn-Sr System

Based on the experimental data of Obinata *et al.* [49], Peng *et al.* [50] modeled all phases in the Mn-Sr binary system as completely disordered solutions. In order to construct a self-consistent thermodynamic database of the Mn-Sn-Sr ternary system, the optimized parameters of Mn-Sr binary system by Aljarrah *et al.* [56] will be adopted in the current calculations.

2.3. Mn-Sn-Sr Ternary System

There is no experimental data for Mn-Sn-Sr system could be found in the literature.

3. Thermodynamic Modeling

3.1. Pure Elements

The Gibbs free energy of a pure element with a certain structure ϕ is described as a function of temperature as:

$${}^\circ G_A^\phi(T) = a + bT + cT \ln T + dT^2 + eT^3 + fT^{-1} + gT^7 + hT^{-9}$$

The parameters a through h are taken from the SGTE compilation by Dinsdale [51].

3.2. Stoichiometric Compounds

The intermetallic compounds in the Sn-Sr phase diagram are considered stoichiometric and the Gibbs free energy of these compounds is described by the following equation:

$$G^{phase, \phi} = x_i {}^\circ G_i^\phi + x_j {}^\circ G_j^\phi + \Delta G_f$$

Where ${}^\circ G_i^\phi$ and ${}^\circ G_j^\phi$ denote Gibbs free energy of element i and j in their standard state and $\Delta G_f = a + bT$ is the Gibbs energy of formation of the stoichiometric compound. Where a and b are the model parameters to be optimized based on experimental data of phase equilibria and thermodynamic properties.

3.3. Liquid Phase

In the present work, the liquid phase is modeled using the MQM where the pair approximation is utilized to describe the short range ordering in the liquid. A detailed description of the MQM for binary and multi-components solutions is available elsewhere [52-54]. Only a brief description will be presented here. The molar Gibbs energy of the liquid phase, derived from the modified quasichemical theory [52], is described by the following equation:

$$G^{liq} = n_i \circ G_i^{liq} + n_j \circ G_j^{liq} - T\Delta\Delta^{config} + \frac{n_{ij}}{2} \Delta^{exs} G^{liq}$$

Where n_i and n_j are the number of moles of the component i and j , n_{ij} is the number of $(i-j)$ pairs, ΔS^{config} is the configurational entropy of mixing given for randomly distributing the $(i-i)$, $(j-j)$, and $(i-j)$ pairs.

$$\Delta S^{config} = -R[n_i \ln(x_i) + n_j \ln(x_j)] - R[n_{ii} \ln(\frac{x_{ii}}{2y_i^2}) + n_{jj} \ln(\frac{x_{jj}}{2y_j^2}) + n_{ij} \ln(\frac{x_{ij}}{2y_i y_j})]$$

Where x_i and x_j are the overall mole fractions of the components i and j , respectively.

4. Results and Discussions

4.1. Sn-Sr Phase Diagram

Figure 1 shows the calculated Sn-Sr phase diagram in relation with the experimental data from the previous literature. A good agreement with the liquidus line between the calculated values in this work and the experimental values of Palenzona and Pani [38], Marshall and Chang [32], and Hoffman [35] is found in Figure 1.

The calculated invariant points of Sn-Sr phase diagram in comparison with the experimental data are listed in Table 1. The activity of Sr in Sn-Sr liquid reported by Klebanov *et al.* [48] was used in the optimization of the Sn-Sr system. Comparison between the calculated activity of Sr from Klebanov *et al.* [48] is shown in Figure 2. The calculated activity shows a good agreement with the experimental data. The optimized model parameters of the stable phases in the Sn-Sr binary system are summarized in Table 2.

The data of enthalpy of mixing by Esin *et al.* [46] was used in the optimization and matched the calculated enthalpy of mixing curve seen in Figure 3. As can be seen from this figure, the calculated enthalpy of mixing of liquid Sn-Sr is in fair agreement with the experimental data. The lowest point of the curve is located where thermally high stable compounds are formed. This indicates stronger atomic interactions in the liquid at the composition of the intermetallic compounds.

Figure 4 shows a comparison between the calculated enthalpy of formation for all phases in Sn-Sr system and that reported by Zhao *et al.* [29]. The current enthalpies of formation for intermetallic compounds are in fair agreement with that obtained by Miedema model.

4.2. Mn-Sn-Sr Ternary System

The Gibbs energy of the liquid phase was calculated using the symmetric Kohler-like approximation [55] with no ternary interaction parameters for the liquid phase. The invariant reactions for the Mn-Sn-Sr system are listed in Table 3. The liquidus projection of the Mn-Sn-Sr ternary system is shown in Figure 5. The miscibility gap covers most of the composition triangle, as shown in Figure 5. Since there is no ternary experimental data available for the entire Mn-Sn-Sr system, it is possible that the size of the miscibility gap is over or underestimated by the extrapolation.

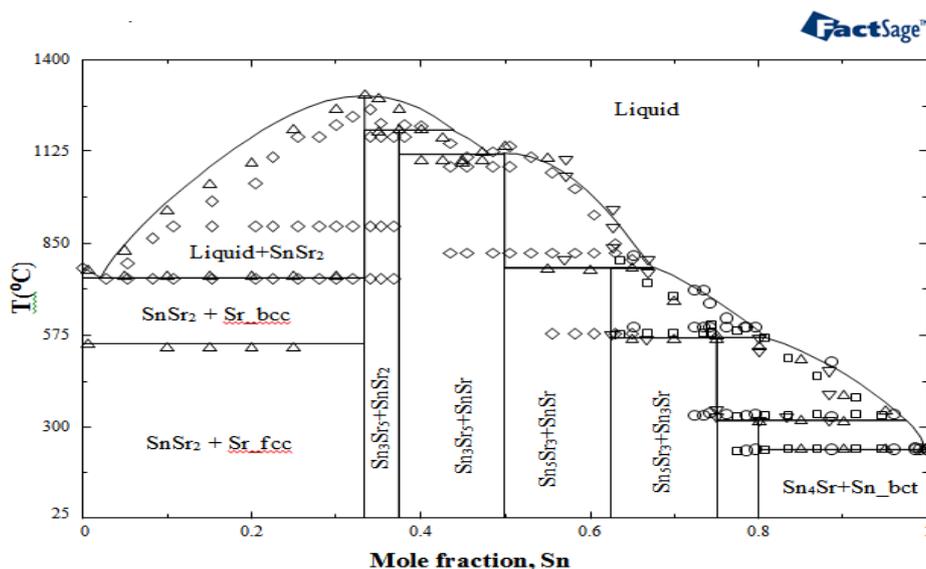


Figure 1. Re-optimized Sn-Sr phase diagram in comparison with experimental data from literature: Palenzona and Pani Δ , Marshall and Chang \circ , Ray \square , Widra and Schafer \diamond and Hoffman ∇

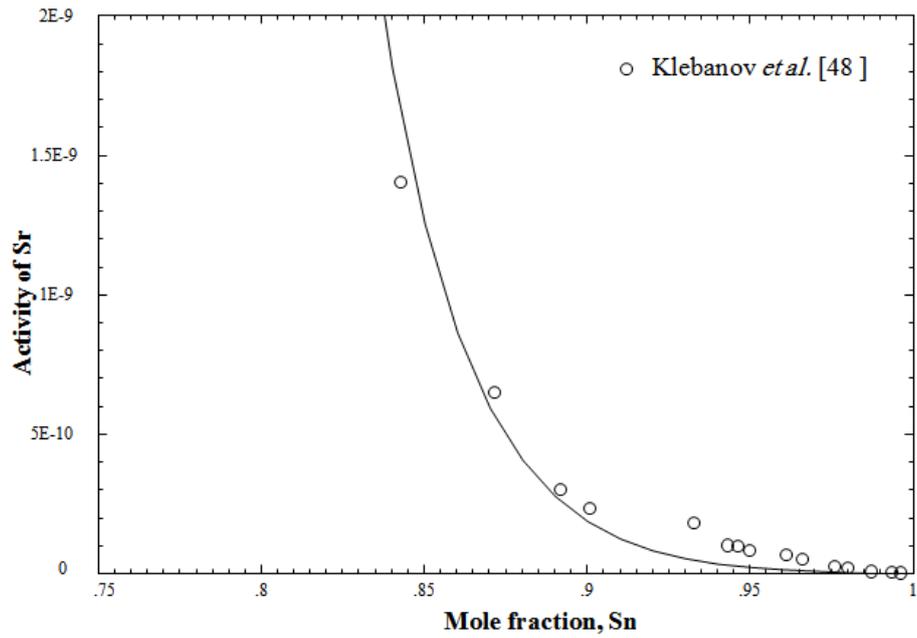


Figure 2. Plot of calculated activity of Sr against mole fraction Sn for the Sn-Sr liquid at 900 K in comparison with experimental data of Klebanov et al. [48]

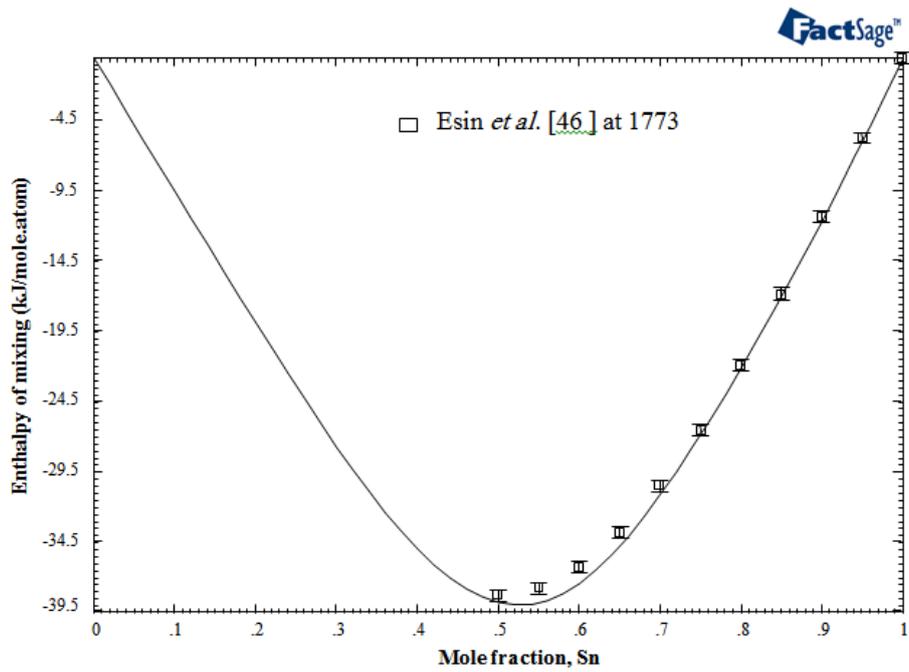


Figure 3. Calculated enthalpy of mixing against mole fraction Sn for the Sn-Sr liquid at 1773 K in comparison with the experimental data of Esin et al. [46]

Table 1: Comparison between calculated and experimental values of the invariant reactions in the Sn-Sr system.

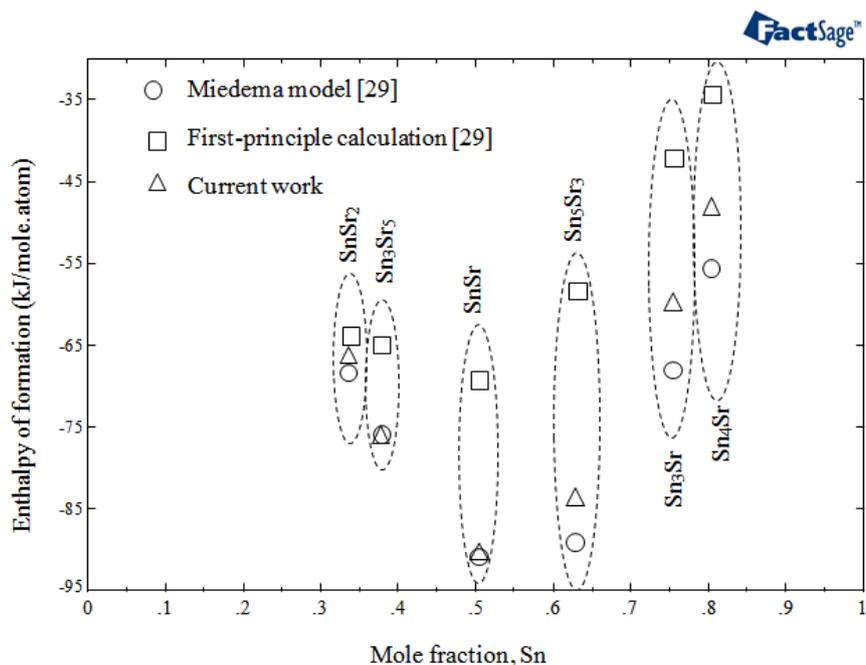
Reaction type	Reaction	Composition (at.%Sn)	T(°C)	Reference
		2.22	741	This work
Eutectic	Liquid \leftrightarrow Sr_BCC + Sr ₂ Sn	2.7	745	[33]
		2.5	752 ± 5	[38]
		33.3	1288	This work
Congruent	L1 \leftrightarrow Sr ₂ Sn		1295 ± 5	[38]
			1255	[33]
Perectitic	Liquid + Sr ₂ Sn \leftrightarrow Sr ₅ Sn ₃	37.5	1185	This work
		39.9	1190 ± 5	[38]
		36.7	1170	[33]
Congruent	L1 \leftrightarrow SrSn	50	1141	This work
		50	1140	[33]
		50	1140 ± 5	[38]
		62.5	776	This work
Perectitic	Liquid + SrSn \leftrightarrow Sr ₃ Sn ₅	65	775 ± 5	[38]
			780	[36]
			810	[37]
		75	557	This work
Perectitic	Liquid + Sr ₃ Sn ₅ \leftrightarrow SrSn ₃	79.7	598 ± 1	[32]
			563	[36]
			560 ± 5	[38]
		73.5	580	[31]
Perectitic	Liquid + SrSn ₃ \leftrightarrow SrSn ₄	80	320	This work
		95.1	338	Ray
		96	334 ± 4	[32]
		95.8	315 ± 5	[38]
			316	[36]
Eutectic	Liquid \leftrightarrow Sn_BCC + SrSn ₄	99.8	231	This work
		99.7	232	[31]
		99	230 ± 2	[32]
		99	230 ± 5	[38]
			212	[32]
Eutectic	Liquid \leftrightarrow SrSn + Sr ₅ Sn ₃	48.7	1144	This work
		45.6	1100 ± 5	[38]
		46.1	1080	[33]

Table 2: Optimized parameters of all phases in Sn-Sr and Mn-Sr systems.

Phase	Thermodynamic parameters	
Liquid	$Z_{SnSr}^{Sn} = 5; Z_{SnSr}^{Sr} = 5$	
Sn-Sr Liquid	$\Delta g_{SnSr}^0 = -2127.2 - 1.55T; \Delta g_{SrSn}^{10} = -478 - 2.39T$ $\Delta g_{SrSn}^{01} = -1.43T$	
SrSn	${}^\circ G_{298.15K}^{SrSn} = -93500 + 8.13T$	
Sr ₂ Sn	${}^\circ G_{298.15K}^{Sr_2Sn} = -80666.7 + 17.4T$	
SrSn ₃	${}^\circ G_{298.15K}^{SrSn_3} = -59500 + 0.78T$	
Sn-Sr intermetallic phases	Sr ₅ Sn ₃	${}^\circ G_{298.15K}^{Sr_5Sn_3} = -90875 + 8T$
	SrSn ₄	${}^\circ G_{298.15K}^{SrSn_4} = -47880 + 0.61T$
	Sr ₃ Sn ₅	${}^\circ G_{298.15K}^{Sr_3Sn_5} = -83375 + 1.05T$

Table 3. The calculated enthalpies of formation for all the stable compounds in the Sn–Sr system in comparison with literature.

Compound	Miedema Model	First-principle method	MQM
SnSr ₂	-68.02	-63.40	-66.00
Sn ₃ Sr ₅	-75.47	-64.47	-75.75
SnSr	-90.55	-68.88	-90.00
Sn ₅ Sr ₃	-88.83	-57.93	-83.38
Sn ₃ Sr	-67.84	-41.74	-59.5
Sn ₄ Sr	-55.32	-33.96	-47.88

**Figure 4.** The calculated enthalpy of formation of all intermetallic compounds in comparison with work of Zhao *et al.* [29].

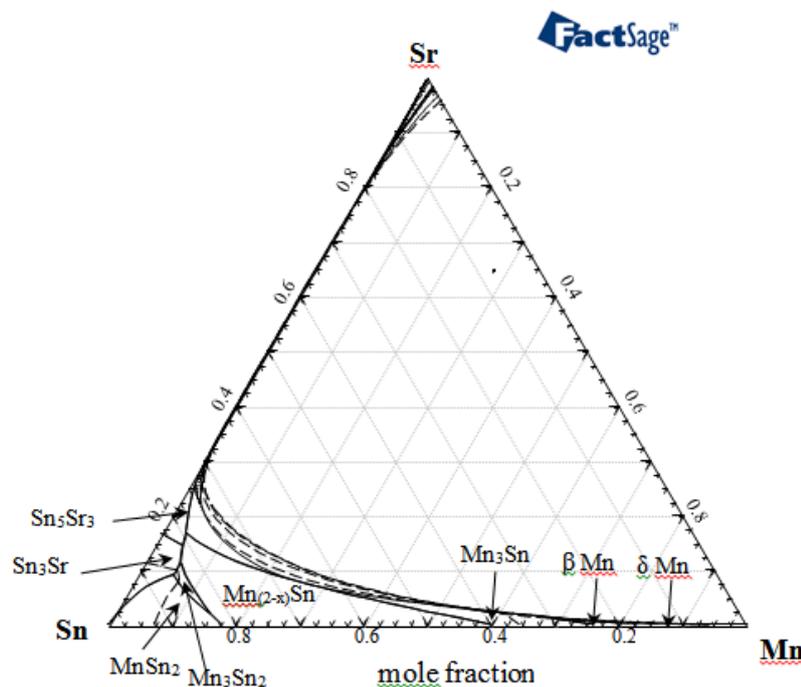


Figure 5. Liquidus projection of Mn-Sn-Sr ternary phase diagram

5. Conclusions

The experimental phase diagram data and thermodynamic properties of the Sn-Sr binary system was evaluated and optimized to obtain self-consistent model parameters for the Gibbs energy of all phases. The calculated liquidus line of Sn-Sr phase diagram was compared with the experimental data and found to be in a good agreement. The calculated thermodynamic properties such as activity and enthalpy of mixing of the Sn-Sr system were in accord with the experimental data in the literature. By combining self-consistent set of optimized parameters of the phases of the constituent binary sub-systems, liquidus projection of the ternary Mn-Sn-Sr phase diagram was obtained.

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