



Experimental and thermodynamic study of the Mg-Sn-Ca-Sr quaternary system: Part I-Mg-Sn-Ca ternary system



Jian Wang^{a,*}, Jiajian Han^b, Beining Du^c, Yixiong Huang^b, Liyuan Sheng^c, Weifeng Rao^{a,*},
Cuiping Wang^b, Xingjun Liu^b

^a Department of Materials Physics, Nanjing University of Information Science & Technology, Nanjing, Jiangsu Province 210044, China

^b Fujian Key Laboratory of Materials Genome, College of Materials, Xiamen University, Xiamen 361005, China

^c Shenzhen Institute, Peking University, Shenzhen, Guangdong Province 518057, China

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ABSTRACT

Four key samples of the Mg-Ca-Sn ternary system in the Mg-rich region at 415 and 350 °C have been determined using the Scanning Electron Microscopy (SEM) equipped with Energy Dispersive X-Ray Spectrometry (EDS). The existence of the CaMgSn ternary compound was confirmed in these two isothermal sections. Thermodynamic optimizations of the Ca-Sn binary system and Mg-Ca-Sn ternary system were carried out using the CALPHAD (Calculation of Phase Diagrams) technique. The Modified Quasichemical Model (MQM) was used for the liquid solution which exhibits a high degree of short-range ordering behavior in the liquid solution. The solid phases are modeled with the Compound Energy Formalism (CEF). Finally, a self-consistent thermodynamic database of Mg-Ca-Sn ternary system has been constructed in the present work, which would provide an efficient and convenient way to study and develop the new Mg-Sn based alloys.

1. Introduction

Magnesium alloys, with a density around 1.74 g/cm³ which is nearly 1.6 and 4.5 times less dense than aluminum alloys and steel, is an exceptionally lightweight structural materials. The low density of magnesium alloys is a strong driving force for their applications in the automotive and aeronautical industries with the reducing weight of vehicles and fuel consumption. The magnesium and its alloys have some advantageous properties as high thermal conductivity, high dimensional stability, high damping characteristics, high machinability, completely recyclable ability *etc.* [1], which make them be applied into the automobile and computer parts, aerospace components, and household equipment parts successfully.

Mg-Sn based alloys have stable microstructures and good mechanical properties at high temperatures due to the high solubility of Sn in hcp (Mg) and to the potential to precipitate a cubic second phase (Mg₂Sn) in the magnesium-rich matrix. Previous investigations indicate that Mg-Sn alloys with additional alloying elements have comparable or even better creep properties than AE42 alloys [2–4]. In addition, it is known that Sn can improve the corrosion resistance [5]. Unfortunately, it is required a long aging time for the Mg-Sn alloys to reach the peak hardness, which is not practical for industrial applications [6]. Hence, it is necessary to improve the age hardening

response and creep resistant behavior of Mg-Sn alloys.

Significant improvements of the binary Mg-Sn and ternary Mg-Sn-Ca systems, in as-cast conditions, have been recently proposed to achieve the best combination of castability, creep and corrosion properties [7–10]. Addition of Ca has been found to improve creep resistance [11]. Consequently, the Mg₂Sn, Mg₂Ca, and Ca_{2–x}Mg_xSn phases have been investigated more [9]. Hirai et al. [12] reported that cast AZ91 magnesium alloys having excellent mechanical properties, such as high strength and high creep resistance, can be made by the addition of Ca. As mentioned above, superior mechanical properties have been obtained for Mg-Sn based alloys by addition of Ca. However, in order to systematic study the mechanisms behind the properties improvements; a better understanding of phase equilibria relationships of Mg-Sn-Ca ternary system is required.

The phase equilibria of the three binary sub-systems Mg-Sn [13–16], Mg-Ca [17,18] and Ca-Sn [19] of the Mg-Sn-Ca ternary system have been well investigated. The final accepted version of each binary phase diagram is shown in Fig. 1. The solid solubility and crystal structure of the ternary compound CaMgSn (orthorhombic Co₂Si-type) in the Mg-Sn-Ca ternary system was studied by Axel et al. [20] in the first time, which was expected to has the same crystal structure of binary compound Ca₂Sn. Later, Ganguli et al. [21] confirmed the existence of the CaMgSn compound and pointed out that the CaMgSn

* Corresponding authors.

E-mail addresses: Jian.wang@nuist.edu.cn (J. Wang), wfraonust@163.com (W. Rao).

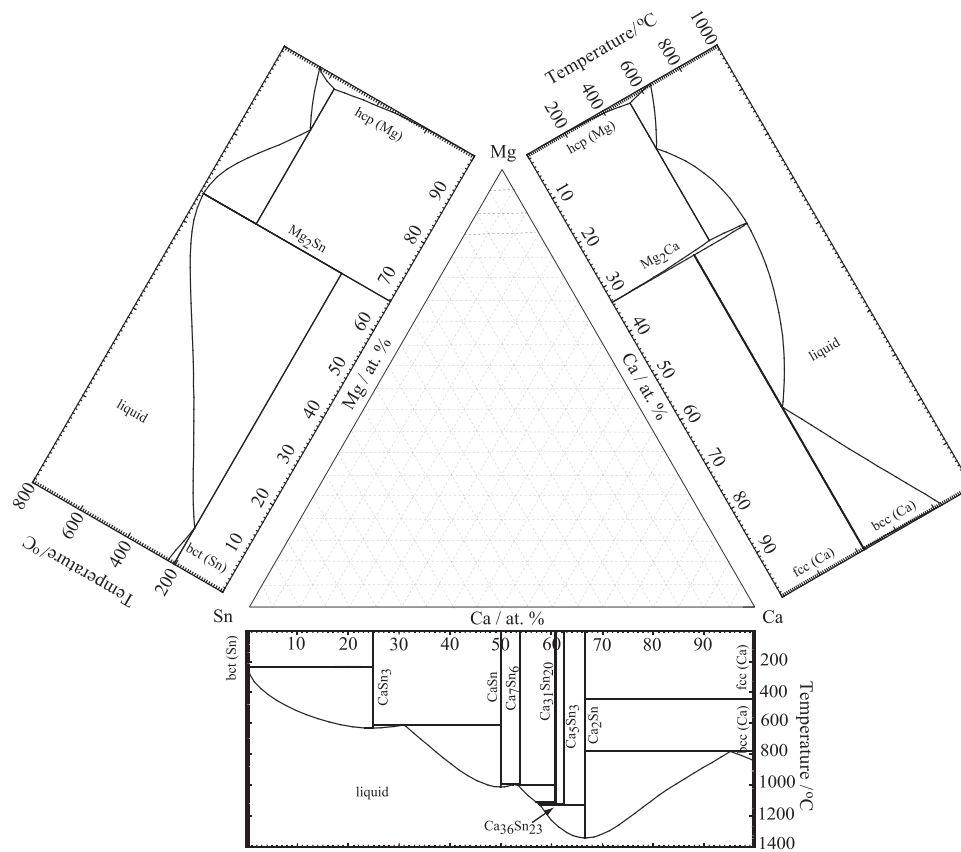


Fig. 1. The final accepted version of each binary phase diagram of Mg-Ca-Sn ternary system.

Table 1
Optimized phases and thermodynamic models used in the present work.

Phase	Pearson symbol	Strukturvricht designation	Space group	Prototype	Model ^a
Liquid	—	—	—	—	MQM
fcc	cF4	A1	$Fm\bar{3}m$	Cu	CEF
bct	tI4	A5	$I4_1/mmm$	Sn	CEF
bcc	cI2	A2	$Im\bar{3}m$	W	CEF
hcp	hP2	A3	$P6_3/mmc$	Mg	CEF
Mg ₂ Sn	cF12	C1	$Fm\bar{3}m$	CaF ₂	ST
Ca ₂ Sn	oP12	C23	$Pnma$	Co ₂ Si	CEF
Ca ₅ Sn ₃	tI32	D8 ₁	$I4_1/mcm$	Cr ₅ B ₃	ST
Ca ₃₆ Sn ₂₃	tP118	—	$P4/mbm$	Y ₃₆ Sn ₂₃	ST
Ca ₃₁ Sn ₂₀	tI204	—	$I4/mcm$	Pu ₃₁ Ph ₂₀	ST
Ca ₇ Sn ₆	oP52	—	$Pnma$	Ca ₇ Sn ₆	ST
CaSn	oC8	Bf	$Cmcm$	CrB	ST
CaSn ₃	cP4	L1 ₂	$Pm\bar{3}m$	AuCu ₃	ST
Mg ₂ Ca	hP12	C14	$P6_3/mmc$	MgZn ₂	CEF
Ca _{6.2} Mg _{3.8} Sn ₇	—	—	—	—	ST
CaMgSn	oP12	C23	$Pnma$	Co ₂ Si	CEF

^a MQM: modified quasichemical model; CEF: compound energy formalism; ST: stoichiometric compound.

ternary phase is actually the ternary solid solution of Ca₂Sn with Mg substituting of Ca. Also, a new ternary compound Mg_{3.8}Ca_{6.2}Sn₇ was synthesized by Ganguli et al. [22]. Recently, phase equilibria of the Mg-Sn-Ca ternary system were studied by Kozlov et al. [23] by DSC/DTA, X-ray, and metallurgical analysis techniques. The isoplethal sections of the Mg₂Sn-Mg₂Ca, Mg-CaSn, Mg₂Sn-Ca₂Sn, and Ca₄₇Sn₅₃-Mg (at%) were reported. All the solid phases and their crystal structure information in the Mg-Sn-Ca ternary system are summarized in Table 1.

Therefore, in the present work, in order to have a comprehensive understanding of the phase equilibria of Mg-Ca-Sn ternary system, the experimental measurements and thermodynamic optimizations on the

phase equilibria of Mg-Sn-Ca ternary system were carried out. It is a part of development a self-consistent thermodynamic database of Mg-X (X: Ag, Ca, In, Li, Na, Sn, Sr, Zn) multi-component system for Mg-based alloys design purpose [15,24–30].

2. Experimental procedure

The Mg-Ca-Sn ternary alloy samples were prepared using Mg (99.8%), Sn (99.9%), and Ca (99.9%) supplied by Alfa Aesar company, in an induction furnace under flowing argon with a pressure of 0.2 MPa. In order to make sure obtaining the completely melted samples, the melting temperatures of samples were set to be 1000–1200 °C in the present work. A cube shaped crucible made with Ta foil (99.5% purity, 0.15 mm thickness) was prepared for sample melting. Samples were washed with methanol to remove the oil, and then were melted three times in the crucible in order to obtain the homogeneity sample. The overall mass losses were less than 3 wt% for each sample. The melting procedure involved the fusion of Mg and Sn in the first step, before adding Ca, in order to reduce the exothermic effect of Ca with Sn. Then the Mg-Ca-Sn key alloys were sealed into quartz capsules under argon (with a pressure of 0.05 MPa) and annealed at 415 °C for 20 days and at 350 °C for 35 days, respectively.

The microstructure and equilibrium composition of the constituted phases in each sample were investigated using SEM/EDS technique within 2 min after the final polishing. Five points in different areas of each target phase were measured using EDS to obtain an average value of each phase composition. The standard deviation of measured values for each phase is less than 1.9. The nominal compositions of the samples and of their constituent phases are summarized in Table 2.

3. Thermodynamic modeling

In the present work, all the thermodynamic optimizations of the

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