Supporting Information

Influence of phase separation and spinodal decomposition on microstructure of $Mg_2Si_{1-x}Sn_x$ alloys

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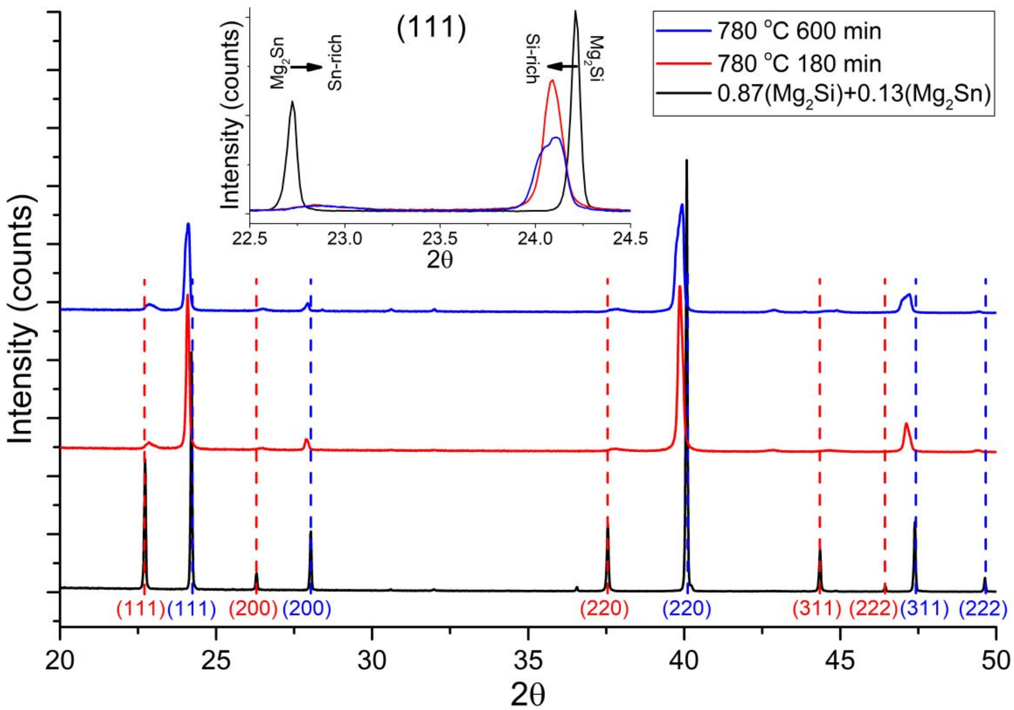

FIG. S1. Homogenization of $Mg_2Si_{0.87}Sn_{0.13}$ at 780 °C (red and blue vertical dashed lines represent positions of the reflections of pure Mg_2Sn and Mg_2Si, respectively).

To determine the time needed to homogenize the mixtures at 780 °C, the initial mixture (black pattern) of Mg_2Si and Mg_2Sn in stoichiometric amounts (to form $Mg_2Si_{0.87}Sn_{0.13}$) was sintered at 700 °C for 10 min in SPS and brought to 780 °C for homogenization. After dwelling for 180 and 600 min (red and blue patterns, respectively), the samples were quickly cooled in SPS (~140 K/min). From the shift in reflection positions indicated by the arrows in the magnification of the reflection (111), the unit cell of Mg_2Si becomes larger, while the Mg_2Sn unit cell becomes smaller. This indicates that the initial mixture was treated in the single-phase region at 780 °C. Rietveld refinement shows that the Si-rich phase greatly dominates over the Sn-rich phase in both samples. The Sn-rich phase does not decrease with dwelling, which provides evidence to suggest that homogenization is accomplished after 3h.

Both samples have wide Si- and Sn-rich reflections, which represents strained structures. The sample treated for 600 min decomposed spinodally, since the peaks of the Si-rich phase split into two sets of peaks, which indicates the formation of two Si-rich phases with similar compositions. A stronger pinning effect due to a smaller size of grains could explain the absence of the peak split in the sample treated for 180 min. A minor amount of the Sn-rich phase in both samples, most likely, is also formed according to the mechanism of spinodal decomposition. Further demixing of the samples was suppressed due to quick cooling.

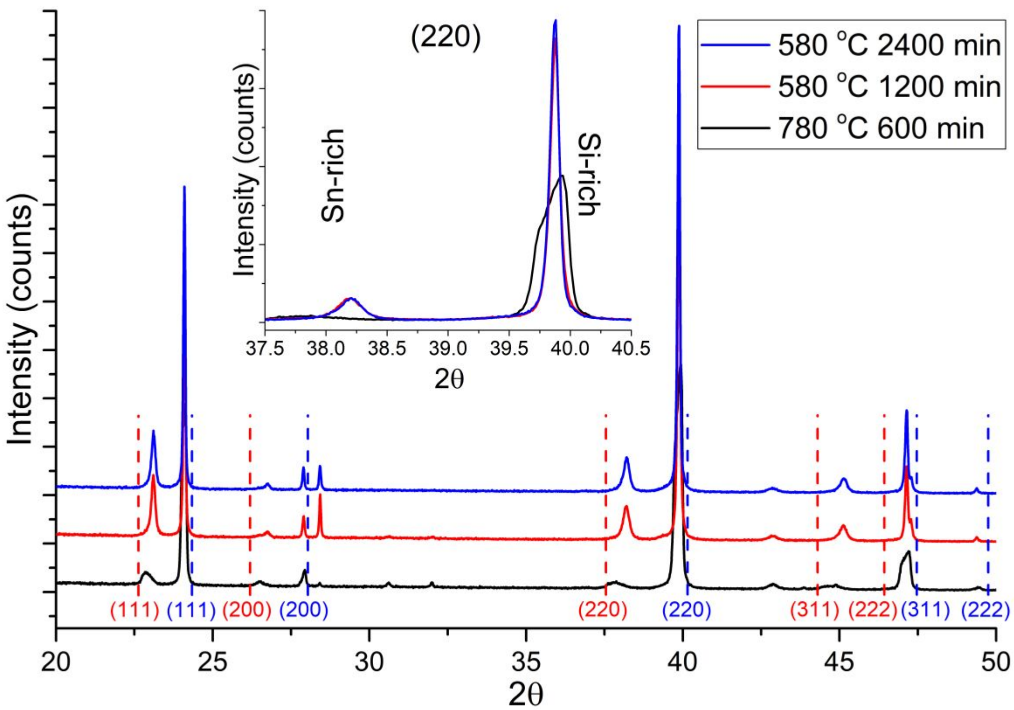

FIG. S2. Phase separation in $Mg_2Si_{0.87}Sn_{0.13}$ at 580 °C (red and blue vertical dashed lines represent positions of the reflections of pure Mg_2Sn and Mg_2Si, respectively).

The sample, which was treated in SPS at 780 °C for 600 min and quickly cooled (black pattern), was also treated in the tube furnace at 780 °C for 600 min (to ensure homogenization) and then annealed at 580 °C for 1200 and 2400 min, respectively (red and blue patterns, respectively). Since compositions and amounts of the formed phases are the same in both cases, we conclude that the sample achieved thermodynamic equilibrium after 1200 min at 580 °C.

Table S1. Extended summary of results from Rietveld refinement of $Mg_2Si_{1-x}Sn_x$ alloys treated at 680 °C. [a)]The second Si-rich phase of sample $Mg_2Si_{0.92}Sn_{0.08}$ was not possible to refine.

Nominal composition	Phases composition	Phase amount, mol%	Lattice parameter, Å	Overall Sn amount, at%	R_{wp}	Zero error
$Mg_2Si_{0.97}Sn_{0.03}$	$Mg_2Si_{0.987}Sn_{0.013}$	29.6	6.360697	4.1	8.6	0.00013
	$Mg_2Si_{0.944}Sn_{0.056}$	66.0	6.378280			
	Si	4.4				
$Mg_2Si_{0.94}Sn_{0.06}$	$Mg_2Si_{0.977}Sn_{0.023}$	11.2	6.364752	6.5	9.8	-0.05692
	$Mg_2Si_{0.926}Sn_{0.074}$	84.9	6.385404			
	Si	3.9				
$Mg_2Si_{0.92}Sn_{0.08}$	[a)] $Mg_2Si_{0.911}Sn_{0.089}$	93.1	6.391747	8.5	9.3	-0.08385
	Si	6.9				
$Mg_2Si_{0.87}Sn_{0.13}$	$Mg_2Si_{0.888}Sn_{0.112}$	87.0	6.401046	16.4	10.0	0.04697
	$Mg_2Si_{0.150}Sn_{0.850}$	6.9	6.703796			
	Si	5.2				
	Sn	0.8				
$Mg_2Si_{0.82}Sn_{0.18}$	$Mg_2Si_{0.880}Sn_{0.120}$	80.7	6.404433	19.4	9.1	-0.00778
	$Mg_2Si_{0.095}Sn_{0.905}$	8.8	6.726414			
	$Mg_2Si_{0.227}Sn_{0.763}$	0.8	6.668349			
	Si	8.5				
	Sn	1.1				

Table S2. Extended summary of results from Rietveld refinement of $Mg_2Si_{1-x}Sn_x$ alloys treated at 580 °C.

Nominal composition	Phase composition	Phase amount, mol%	Lattice parameter, Å	Overall Sn amount, at%	R_{wp}	Zero error
$Mg_2Si_{0.97}Sn_{0.03}$	$Mg_2Si_{0.988}Sn_{0.012}$	41.8	6.360071	3.5	10.4	-0.05108
	$Mg_2Si_{0.958}Sn_{0.062}$	48.9	6.380694			
	Si	9.2				
$Mg_2Si_{0.94}Sn_{0.06}$	$Mg_2Si_{0.975}Sn_{0.025}$	8.7	6.365322	6.6	9.3	-0.04698
	$Mg_2Si_{0.927}Sn_{0.073}$	87.6	6.385321			
	Si	3.7				
$Mg_2Si_{0.92}Sn_{0.08}$	$Mg_2Si_{0.917}Sn_{0.083}$	93.2	6.389428	8.6	9.4	-0.01282
	$Mg_2Si_{0.331}Sn_{0.669}$	0.8	6.629579			
	$Mg_2Si_{0.169}Sn_{0.831}$	0.4	6.695948			
	Si	5.6				
$Mg_2Si_{0.87}Sn_{0.13}$	$Mg_2Si_{0.922}Sn_{0.078}$	85.0	6.387167	13.1	10.0	-0.04698
	$Mg_2Si_{0.328}Sn_{0.672}$	9.1	6.630702			
	$Mg_2Si_{0.110}Sn_{0.890}$	0.4	6.720196			
	Si	5.5				
$Mg_2Si_{0.82}Sn_{0.18}$	$Mg_2Si_{0.901}Sn_{0.099}$	81.4	6.396000	18.2	9.8	-0.01415
	$Mg_2Si_{0.331}Sn_{0.669}$	14.9	6.629608			
	$Mg_2Si_{0.108}Sn_{0.892}$	0.2	6.720899			
	Si	3.6				