Modelling and experimental validation of microstructure evolution during the cooling stage of homogenization heat treatment of Al-Mg-Si alloys

Qiang Du , Lina Jia , Kai Tang , Bjørn Holmedal

PII: S2589-1529(18)30124-8
DOI: https://doi.org/10.1016/j.mtla.2018.09.012
Reference: MTLA 105

To appear in: *Materialia*

Received date: 29 May 2018
Revised date: 12 September 2018
Accepted date: 12 September 2018

Please cite this article as: Qiang Du , Lina Jia , Kai Tang , Bjørn Holmedal , Modelling and experimental validation of microstructure evolution during the cooling stage of homogenization heat treatment of Al-Mg-Si alloys, *Materialia* (2018), doi: https://doi.org/10.1016/j.mtla.2018.09.012

Modelling and experimental validation of microstructure evolution during the cooling stage of homogenization heat treatment of Al-Mg-Si alloys

Qiang Du[1], Lina Jia[1,2,3], Kai Tang[1], Bjørn Holmedal[3]

[1]SINTEF Materials and Chemistry, 7465, Trondheim, Norway

[2]Beihang University, 100191, Beijing, China

[3]Norwegian University of Science and Technology, 7491 Trondheim, Norway

Abstract

A CALPHAD-coupled multi-component multi-phase Kampmann-Wagner Numerical modelling framework has been adapted and coupled with a homogenization model to predict the competitive nucleation and growth of multi-sized metastable/stable phase particles during the cooling stage of a homogenization heat treatment. The reported model is the continuation of our previous work on the homogenization soaking modelling [Du et al, Acta Materialia 61 (2013) 4961ï4973]. It takes the experimentally verified soaking modelling prediction results as the initial microstructural status and predicts the microstructural evolution during the very last step of the homogenization treatment, i.e. the final cooling process. The model has been applied to two Al-Mg-Si alloys: AA6061 and AA6082. The simulation shows a multi-modal MgSi particle size distribution forms due to multiple nucleation events. For the AA6082 alloy the multiple nucleation events occur mainly due to the local micro-chemistry variations along a dendrite while for AA6061 it is due to the two opposite contributions to supersaturation: the positive contribution by the cooling and the negative contribution by the particles' growth. The interaction of intergranular and intragranular MgSi containing particles have been captured, and the partitioning of Mg and Si solutes among the two different sized MgSi particles has been predicted. The model predictions are in reasonable agreement with the microstructure characterization results obtained with Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM) and Electrical Resistivity Measurement on the quenched samples from some dedicated laboratory-scale homogenization heat treatment experiments. It is concluded that the extended KWN modelling framework is applicable to predict the microstructure evolution during the non-isothermal heat treatment of

multicomponent aluminium alloys.

1. Introduction

Each fabrication step of major metallic structural materials, i.e., aluminium alloys and steels, includes an isothermal process and non-isothermal process such as cooling or heating with or without deformation. It has been well documented that many interesting and industrially critical microstructure evolution phenomena occur during the isothermal processes. For example, during the soaking step of homogenization process of Al-Mg-Si, or AA6xxx alloys, the interdendritic network of the plate-like ß-AlFeSi particles transform to the more rounded discrete Ŭ-AlFeSi particle, and micorsegration is alleviated making the billets easier to deform plastically [1, 2]. Another example is the precipitation sequence during the ageing such as the one revealed in 6xxx alloys, i.e., starting with Super Saturated Solid Solution (SSSS) and passing several metastable phases (atomic clusters, Guinier-Preston (GP) zones, ß" and ß'/B') and terminating with Mg_2Si under over-aged conditions [3-8]. On the other side, it also has been recognized that the non-isothermal processes such as cooling and heating have an equally important impact on microstructure evolutions and the final product properties [9-12].

It should be noted that the studies of a non-isothermal precipitation process are also relevant to quenching. Quenching is a critical step before ageing treatment. It is also a non-isothermal process with an extremely high cooling rate. Taking aluminium alloys as examples, quenching involves not only the generation of the quenched-in vacancies, but also precipitation of particles. In their study of quench sensitivity of Al-Mg-Si alloys over a wide range of cooling rate of $3\text{-}120\times10^4$ ɜ/h, Milkereit and Starink have found from their Differential Scanning Calorimetry (DSC) measurement both of high temperature (typically 500 xC down to 380 xC) and a low temperature precipitation (380 xC down to 250 xC) [13]. Nonetheless, the research efforts dedicated to the non-isothermal process is relatively less than those to the isothermal process.

It is the goal of this paper to revisit this important topic and exploit how a modelling tool, combined with lab-scale experiments, could help in predicting the microstructure evolution

during a non-isothermal process. We will take the cooling process of a homogenization treatment of Al-Mg-Si alloys as an example due to the following two reasons. The first reason is that for the selected alloy system rich and unique microstructure features could be generated by tuning of cooling rate, alloy chemistry and even the initial as-cast microstructure. One can mention the multi metastable phases precipitation and its gradual transformation to more stable phases within the precipitation reaction in Al-Mg-Si alloys. This is unique in reference to the well-studied non-isothermal phase transformation in steels [14] [15] [16]. The co-existence of different sized particles is worthy of noting. The particles' sizes range from several nanometres to several microns, and they often interact with each other leading to the formation of microstructural features such as Precipitate Free Zone (PFZ) [17]. Therefore, this competitive precipitation of various metastable/stable and different sized particles in the Al-Mg-Si is a good case for testing the predictive power of the proposed microstructure model. The second reason is its industrial relevance. As pointed out in [18, 19] [20], for AA6xxx alloys, the cooling has to be designed to re-precipitate as many MgSi particles as possible, in a form and size easily re-dissolvable during subsequent processing for high formability and surface quality. This delicate requirement on microstructure could only be met in a timely manner by the modeling aided alloy design and processing parameter optimization.

Some valuable experimental studies on this interesting non-isothermal precipitation process have been reported. Zajac and Stanislaw have investigated the influence of cooling after homogenization on extrudability and final properties of AA6063 and AA6082 alloys [1]. They have shown that Mg_2Si particles precipitate readily during cooling after homogenization. Above 400 xC, cubic Mg_2Si phase (ϐ phase) precipitates, and below 400 xC only metastable ϐ' phase nucleates predominately on Mn-rich dispersoids. The proportion of ϐ and ϐ' particles depends on the cooling processing parameters. They did not characterize the particle size distribution, but presumably the ϐ and ϐ' particles would exhibit different sizes. They concluded that billets with the ϐ' particles formed at the lower temperature give low flow stress at extrusion temperature, allowing ease deformation and quick particle re-dissolution. Birol has carried out a series of experimental work to study the effects of cooling rates on the

precipitation during the cooling step in 6xxx aluminum alloys [20-24]. His results showed that different density, type and sized precipitates are produced for an alloy cooled at different rates. He also demonstrated that the final microstructure is alloy composition dependent. In general, the amount of both ϐ and ϐ̂ particles increase with decreasing cooling rates and the content of alloying components [22]. He recommended a homogenization practice with a 6 hours soak at 580 xC followed by step-cooling at 250-300 xC for an AA6063 alloy [20]. He concluded that step-cooling gives a more complete depletion of the aluminum solid solution with the formation of fine, metastable ϐ' particles instead of those coarse and stable Mg_2Si particles [20].

There are also some dedicated microstructure modeling activities towards the homogenization and/or cooling for aluminum alloys. Myhr and Grong have developed a valuable framework for modelling concurrent nucleation, growth and coarsening process and applied it to predict the microstructure evolution during up-quenching and linear heating of age hardening Al-Mg-Si alloys [10]. Myhr and Grong's modeling framework is inspiring and seminal while the model can still be improved toward industrial application by extending it to multi-component alloys. Milkereit and Starink proposed a model for predicting quench sensitivity of heat-treatable Al alloys within the age hardening heat treatment procedure, in which the importance of incorporating reliable thermodynamic and kinetic models have been emphasized [13]. Their model is concise but requires tuning of some non-directly-measurable input parameters. Sun et al [25] have employed a binary Kampamann Wagner Numerical model to predict the precipitation kinetics of Mg_2Si particles, and predicted a bimodal particle size distribution formed during the cooling of homogenization heat treatment. However, their model only considered the precipitation of the stable phase (ϐ phase) and ignored the formation of the metastable ϐ' phase. The most recent study on this interesting topic is due to Priya, Johnson and Krane [26], and they have presented numerical study of microstructural evolution during homogenization heat treatment of Al-Si-Mg-Fe-Mn alloys. However, their model again is only for binary alloys, and they did not consider the precipitation of metastable phases.

It is worthy of noting that the bimodal or multimodal size distribution of single-phase particles has been well investigated in Ni-based alloys due to its important property consequences. Phase field [27] and mean field approach [28] have been adopted to model this interesting microstructural evolution. Whether this bimodal particle size distribution exists in Al alloys is an interesting question that remains to be confirmed. In this paper, we will investigate the microstructure evolution during the cooling stage of homogenization heat treatment of Al-Mg-Si alloys by a combination of modeling and experimental approaches. The employed modeling approach is the CALculation PHAse Diagram (CALPHA)-coupled multi-component multi-phase Kampmann-Wagner numerical (KWN) approach reported by Du and his co-authors in a series of publications [17, 29-33]. The model predictions on the isothermal homogenization soaking and ageing heat treatment of Al alloys had been validated by the experimental microstructure characterizations. In this paper the KWN model will be integrated with the Scheil additivity rule to extend its application to a cooling (non-isothermal) process.

Our justification to select the KWN approach for the homogenization cooling modeling is that the approach is amenable to couple with phase diagrams, numerically robust and efficient, which is advantageous in comparison to phase field method for the applications involved industrial alloys and processes. Although there are more assumptions in the KWN approach than in phase field method such as local equilibrium at the precipitate-matrix interface and the growth is purely diffusion controlled, the KWN approach, when tuned with some key experimental microstructure characterization results, is considered as a suitable modeling tool in investigating industrial alloys' microstructure evolution.

This paper starts with a brief description of the employed modeling and experimental approaches. Then the predictions of the competitive nucleation and growth of ϐ particles and ϐ' particles during a homogenization cooling process are presented. The model predictions are examined in the light of microstructure characterization results carried out with Scanning Electron Microscopy, Transmission Electron Microscopy and Electrical Resistivity Measurement on the samples from some dedicated laboratory-scale homogenization heat

treatment experiments.

2. The microstructural model for the homogenization cooling

The industrial Al-Mg-Si alloy homogenization treatment is often conducted at a temperature as high as possible without causing incipient melting for a couple of hours. This high temperature soaking results in that Mg and Si solutes are taken back into the Al solid solution phase and ready to re-precipitate intra-granularly and inter-granularly upon cooling. Given insufficient soaking time or low soaking temperature, the spatial distributions of the alloying components might remain non-uniform across a secondary dendrite arm, i.e., the local micro-chemistry varies leading to different kinetic response during the cooling. In addition, constituent particles, which initially were formed at the end of solidification (casting process) in the interdendritic region, would also grow and drain some solute atoms reducing the total solute supersaturation available for the intragranular MgSi particles formation during the cooling. These are the unique features which have motivated us to perform the research activities reported in this paper.

To describe the complex homogenization, precipitation and their interactions process, some adaptions to the modeling framework reported recently by the authors in [17, 29, 30, 33] need to be made. These adaptions are described in the following three subsections.

2.1 Modeling precipitation during a non-isothermal process

The precipitation process is modeled by adapting the multi-component multi-phase Kampmann-Wagner Numerical framework described in [33]. Below only a summary of the KWN approach is given, and the focus is on its adaption.

The KWN approach is built on the classical nucleation, growth and coarsening laws, and the particles interactions are handled via the mean filed concept. The essence of this framework is that precipitate size distribution curve could be subdivided into size classes, each of which is associated with some identical precipitates. The temporal evolution of the size distribution is

then tracked by following the size evolution of each discrete size class.

The extension to the KWN framework toward the cooling process is the calculation of nucleation incubation time during a non-isothermal process. The Scheil additivity rule [34] is adopted for this extension. The rule states that the fractional nucleation time are additive, and that nucleation begins when the sum of such fractional nucleation time attains the value of unity. The criteria for nucleation can be expressed as

$$\sum_{i=1}^{n} \frac{\Delta t_i}{\tau_i} = 1 \qquad \text{(Eq.1)}$$

Where Δt_i is the time of isothermal held at temperature T_i, and τ_i the incubation period. There is a re-visit to this rule very recently owing to its important role in bridging the gap between isothermal and non-isothermal transformations [35]. It has been accepted that the reactions for which the additive rule is justified are called isokinetic, implying that the fraction transform at any temperature depends only on time and a single function of temperature. This rule fits well within the context of the KWN modelling framework.

2.2 The non-homogenization process

Even soaking can homogenize the solute spatial distribution across a dendrite arm, the subsequent cooling would cause variations in local microchemistry, and this cooling-induced "non-homogenization" process needs to be tracked to predict the final microstructure.

The first microstructural response to the cooling is growth of constituent particles, which would drain solutes gradually from their adjacent dendrite arms. The extent of this draining at the dendrite centre is least due to their longest diffusion distance to these constituent particles. The second response to the cooling is precipitation, and the precipitation model is described in the previous subsection. However, it is worthy of noting that precipitation kinetics might vary from the centre to the periphery of a dendrite arm due to its local microchemistry variation. As to be shown later, this local microchemistry variation is responsible for the multi-modality in particles size distribution curves. On the other hand, precipitation would reduce the local solid solution solute level, therefore affect the solute diffusion along the

dendrite arm.

This non-homogenization process is modelled by adapting the framework we reported earlier in [17]. The original model consists of a 1D Finite Volume Method for solving the diffusion along a half "average" dendrite arm, and the applications of the KWN model to each volume element for predicting precipitates nucleation and growth. On the base of the original framework two adaptions have been made to enable modelling the cooling process. The first adaption is to extend the original assumption that thermodynamic equilibrium is held in interdendritic region from the soaking stage to the cooling process. This extension is justifiable as the same assumption has been validated in the heating stage of a homogenization heat treatment [17]. The second adaption is to turn on the nucleation model for MgSi containing particles during the cooling.

2.3 Initial microstructural status and some thermo-physical input parameters of the cooling simulations

The initial microstructural status, to be subjected to cooling, results from alloy chemistry, casting conditions and homogenization soaking treatment parameters. This initial microstructural status is as important as the cooling rate in determining the final microstructure status. In our simulation, the initial microstructure state is predicted with the comprehensive and experimentally validated homogenization soaking modeling framework for 6xxx alloys [36, 37]. Some experimental data have been used confirming the model predictions while the focus of this paper is on the cooling process.

The application of the KWN model to the cooling process involves the tuning of the nucleation model input parameters: interfacial energies and number of heterogeneous nucleation sites. The interfacial energies have been evaluated in [33] by employing the solution enthalpy method proposed in [38] and their Al-Mg-Si thermodynamic CALPHAD database [39]. They are listed in Table 1 for all the involved metastable and stable phases, i.e. β' and β. However, these calculated values can only be used as guidance in tuning the values.

Some key experimental measurement data on incubation times must be used for the final tuning. The number of heterogeneous nucleation sites is chosen as $0.03\times10^{18}m^{-3}$ and $2.6\times10^{18}m^{-3}$ for the AA6061 and AA6082 alloys respectively according to the experimental TEM characterization performed in our research (to be reported in Section 4). It should be acknowledged that the tuning of these nucleation model input parameters is restricting the applications of the precipitation kinetics modeling approach. Nevertheless, numerical simulation is still considered to be valuable due to the reduction of the times of experimenting.

The other thermo-physical input parameters include thermodynamic databases and diffusivities. The thermodynamic description of the metastable phases in the Al-Mg-Si alloys, established with first-principles total energies and frozen phonon calculations and verified with experimental measurements by Povoden-Karadeniz et al in [39], is adopted to calculate solvus temperature and partition coefficient used in all the simulations performed in this paper. The diffusivity of the alloying component Mg and Si are taken from the work of Du et al [40]. These parameters are listed in Table 2.

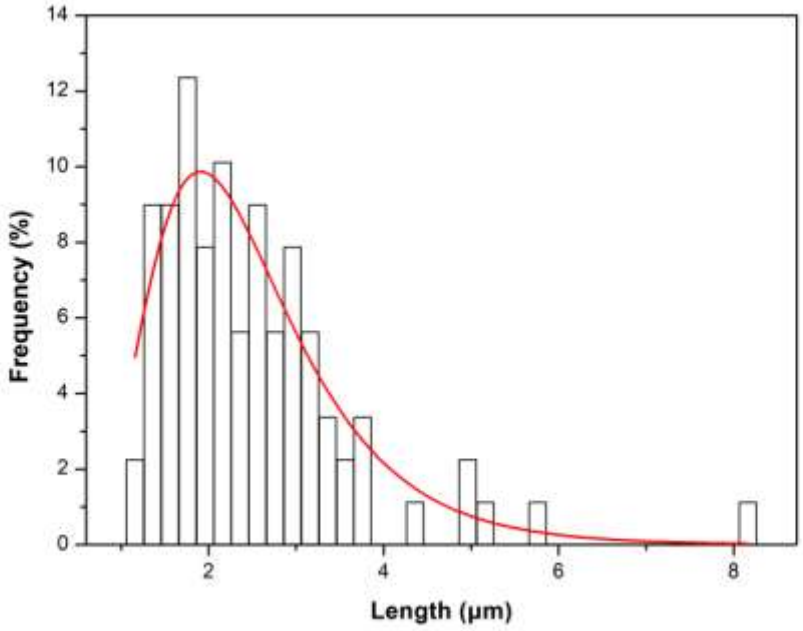
3. Experiments

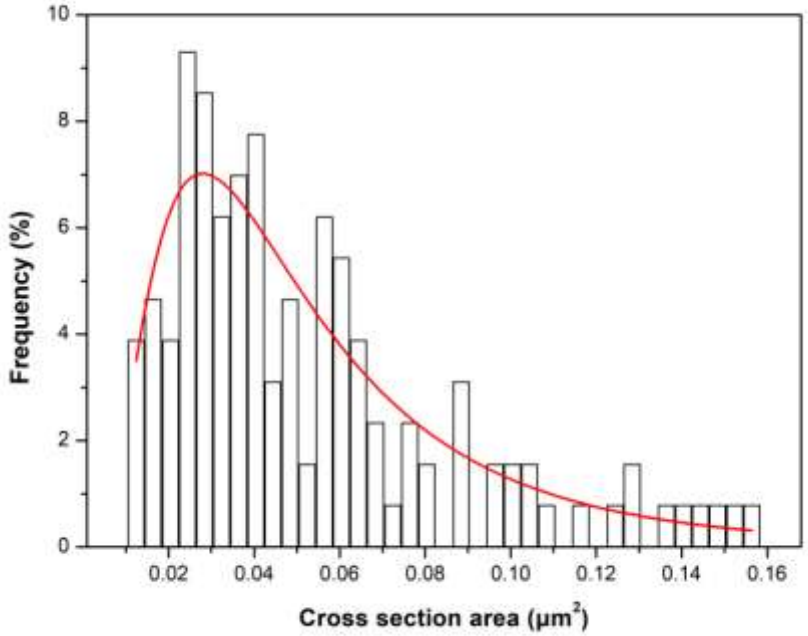
3.1 Materials and lab-scale cooling experiments

The initial materials were DC cast AA6082 and AA6061 billets. The chemistry of the alloys is listed in Table 3.

The lab-scale homogenization heat treatment of the two Al alloys were conducted in a programmable furnace. The heat treatment of AA6061 consists of heating from room temperature to 580 xC at a rate of 200xC/hour, soaking for up to up to 20 hours and cooling down at the rate of 125xC/hour*. The heat treatment of the AA6082 alloy consists of a heating of 200 Ņ/hour from room temperature and soaking at 560 Ņ for 2 hours and cooling down at

* The reported cooling rate is the average between the soaking temperature and 250 °C.


Frequency (%)
Length (μm)
0
2
4
6
8
10
12
14


Frequency (%)
Cross section area (μm²)
0
2
4
6
8
10
0.02
0.04
0.06
0.08
0.10
0.12
0.14
0.16

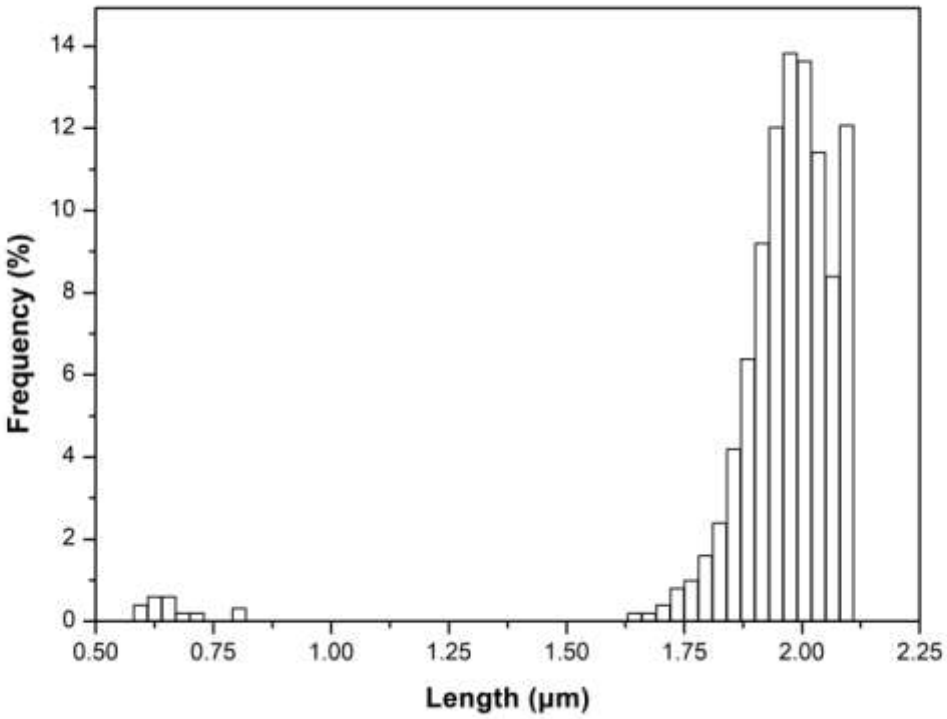


Fig. 6 The measured (a) cross area and (b) length and predicted (c) length distributions of MgSi particles obtained at the end of cooling of the AA6061 alloy

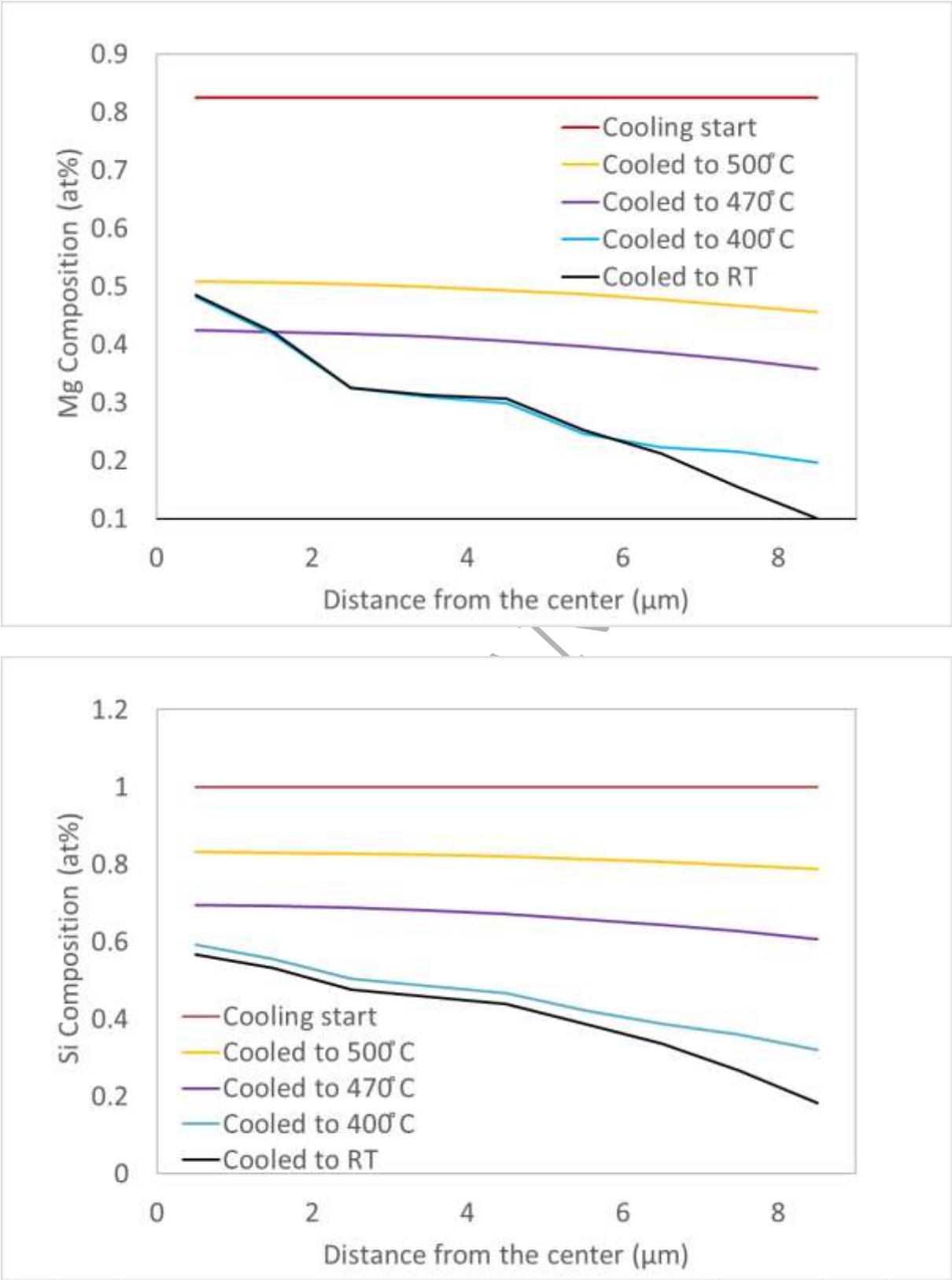


Fig. 7 the average composition profiles of (a) Mg and (b) Si at the start, middle and end of the cooling.

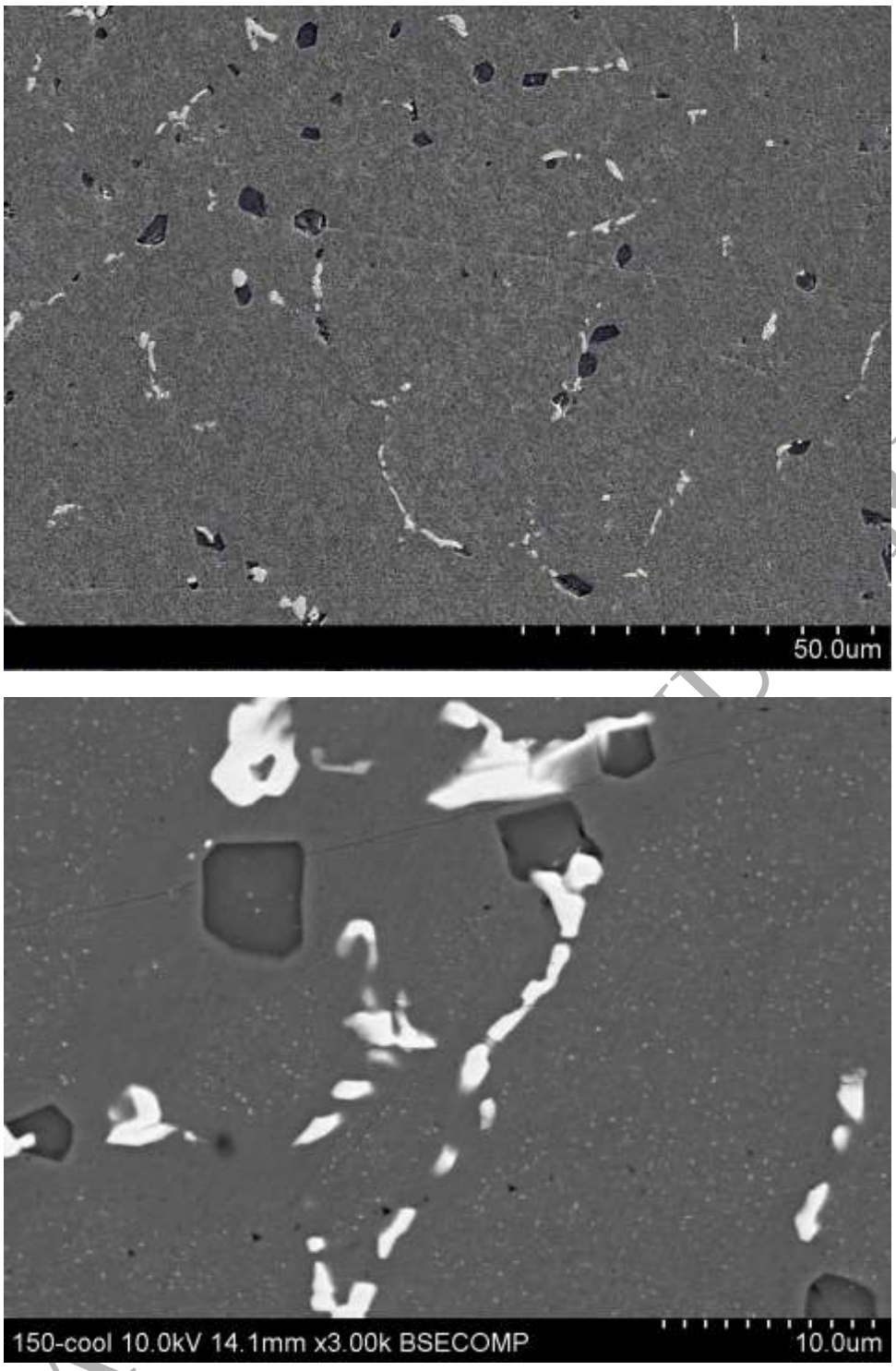


Fig. 8 The SEM images taken at the end of the cooling of the AA6082 alloys.

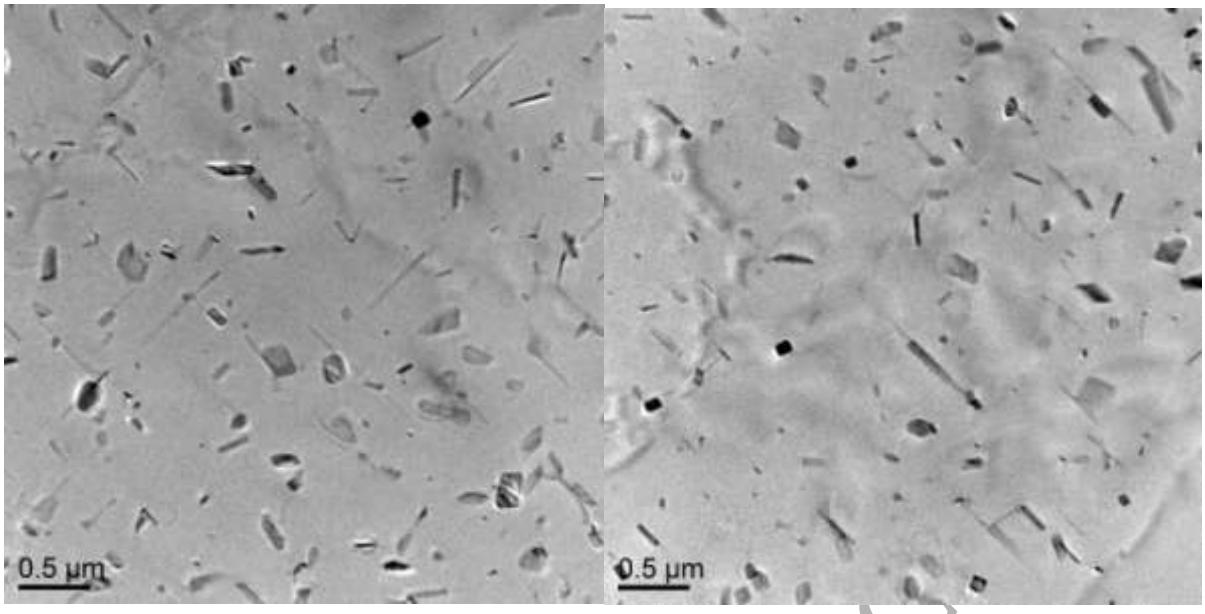


Fig. 9 The TEM images taken at the end of the cooling of the AA6082 alloys.

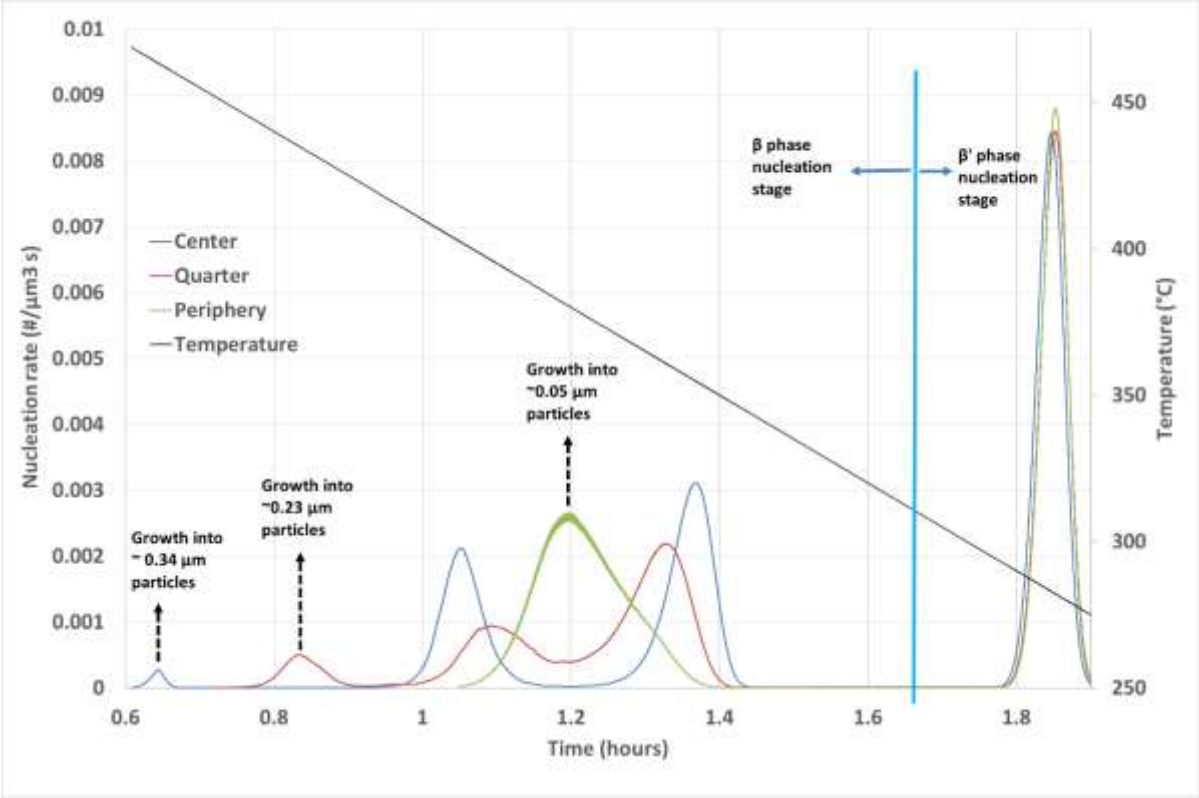


Fig. 10 The predicted nucleation rates at the different locations of a dendrite arm (periphery, 1 quarter from the periphery and centre) during the cooling of the AA6082 alloy.

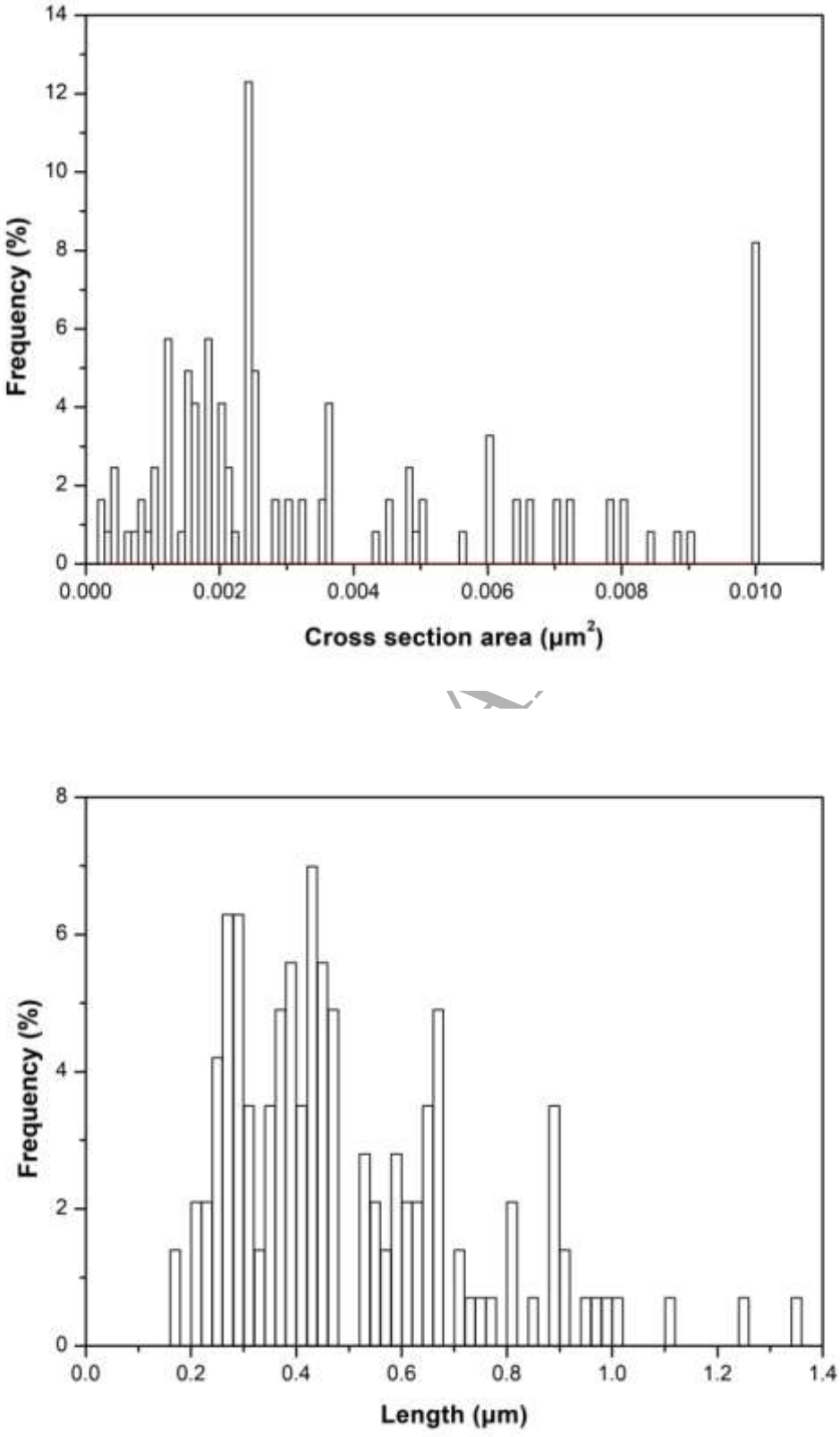


Fig. 11 The measured Mg-Si particles cross section area (a) and length (b) distributions obtained at the end of cooling of the AA6082 alloy.

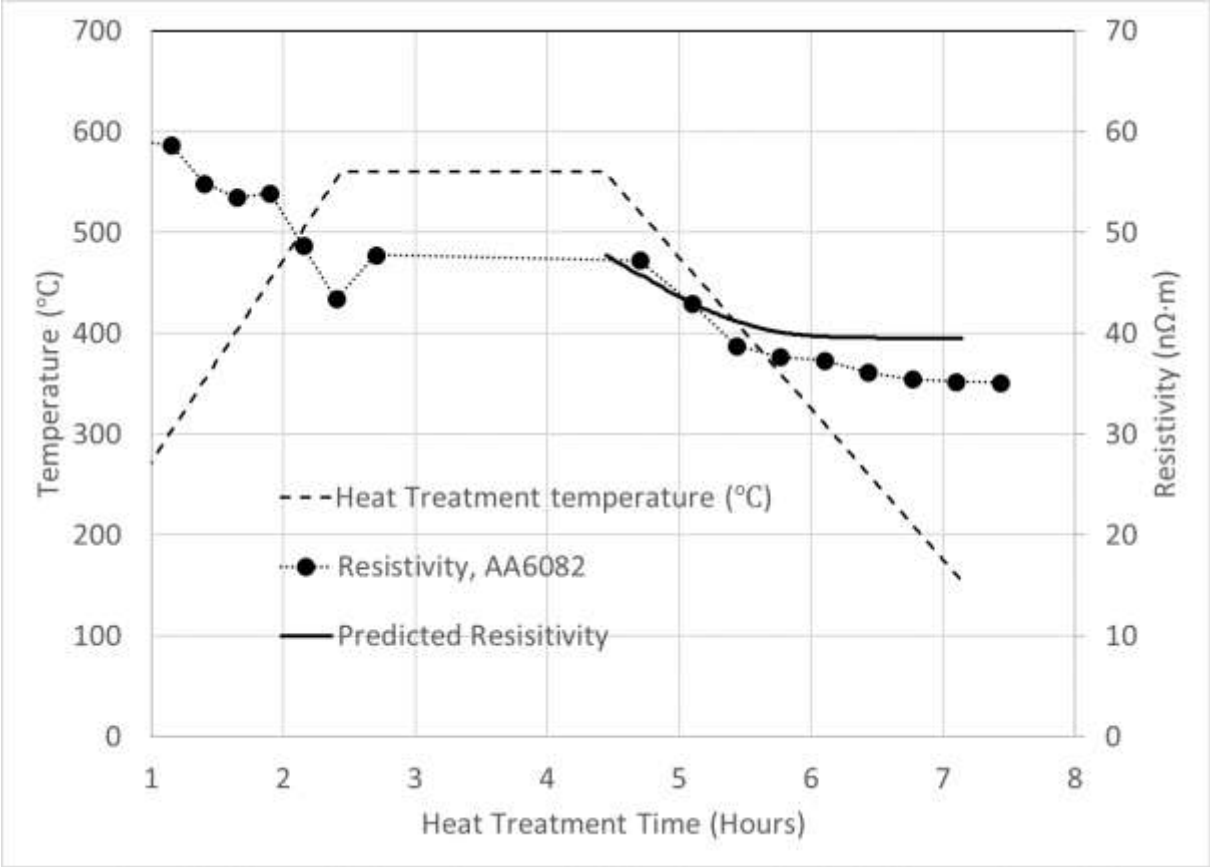


Fig. 12 The measured and predicted resistivity evolution during the cooling of the AA6082 alloy

Table 1 Interfacial energies used in the simulations and calculated using Kozeschnik's solution enthalpy method

Interface with FCC phase	Used in the simulations	Calculated using Kozeschnik's solution enthalpy, averaging in {100}, {111} and {110} planes
	0.09	0.198
ú	0.12	0.227

Table 2 Other material property and nucleation input parameters used in the simulations

0 R O D U Y R O X P H R I phases		1.0×10^{-5} m^3/mol	
Lattice parameters in FCC phase		0.404 nm	
Diffusivities in the FCC phase [40]		Diffusion constant (m^2/s)	activation energy (kJ/mole)
	Mg	1.49×10^{-5}	120.5
	Si	1.38×10^{-5}	117.6
Gibbs-Thomson phase diagram		Metastable phase diagram generated by first principle calculations in [39].	

Table 3 Chemistry of the two alloys investigated in the current work

Alloy	Mg (wt%)	Si (wt%)	Fe (wt%)	Mn (wt%)	Cu (wt%)
AA6082	1.08	1.23	0.27	0.62	0.08
AA6061	0.8085	0.4935	0.0089	0.1365	0.7057

Table 4. The predicted solid solution levels and constituent particles fractions at the end of soaking treatment

	Solid solution solute level (wt%)				Constituent particles fraction (%)	
	Mg	Si	Fe	Mn	Fe/Mn particles	Mg2Si
AA6082	0.85	0.88	0.0018	0.13	1.07	0.43
AA6061	0.81	0.49	0.0045	0.13	0.03	0

Table 5. The predicted and measured intragranular MgSi particles and Mg_2Si constituent particles fractions at the end of cooling of the AA6082 alloy

	Intragranular MgSi particles fraction (%)	Mg_2Si Constituent particles fraction (%)
Prediction	0.29	1.7
Measurement	0.47±0.13	1.78±0.5

Graphical abstract

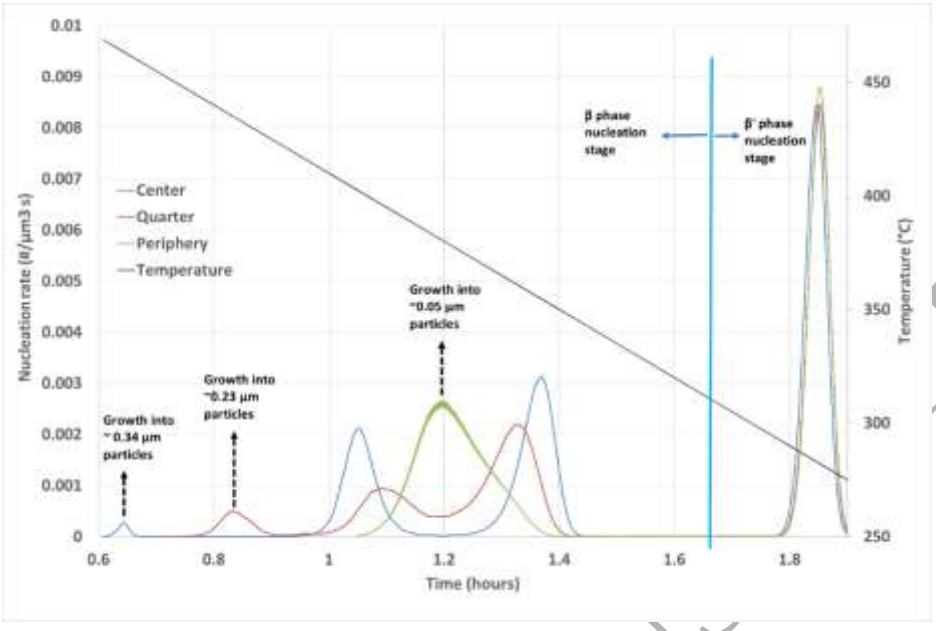
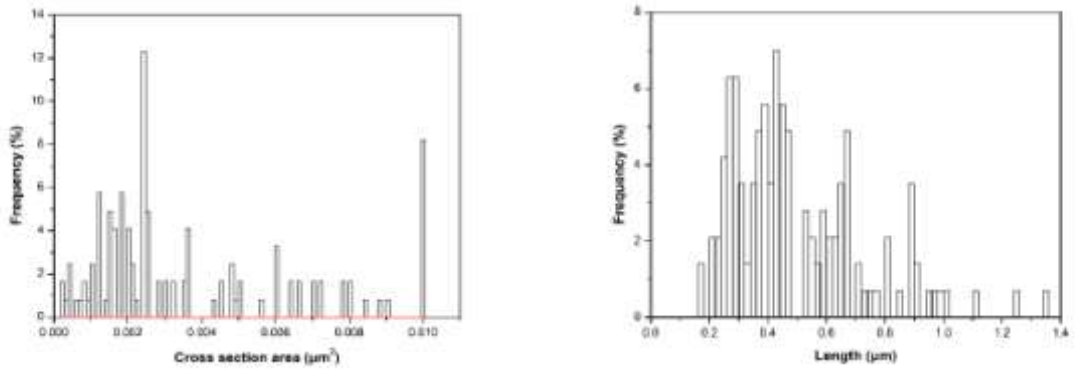


Fig. 1 The predicted nucleation rates at the different locations of a dendrite arm (periphery, 1 quarter from the periphery and centre) during the cooling of the AA6082 alloy.

The proposed model reveals that multi nucleation peaks arise because of:

1. the existences of local microchemistry variations along a dendrite arm
2. the continuous cooling enables the build-up of sufficient thermodynamic nucleation driving forces.

The complex nucleation behaviour shown in this figure is useful in interpreting the multi-modal particles distribution curves from the TEM measurements shown in the following figures.



(a)

(b)

Fig. 2 The measured Mg-Si particles cross section area (a) and length (b) distributions obtained at the end of cooling of the AA6082 alloy.