ASSESSMENT AND MODELLING OF PARTICLE CLUSTERING IN CAST ALUMINUM MATRIX COMPOSITES

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ASSESSMENT AND MODELLING OF PARTICLE CLUSTERING IN CAST ALUMINUM MATRIX COMPOSITES

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ABSTRACT

ASSESSMENT AND MODELLING OF PARTICLE CLUSTERING IN
CAST ALUMINUM MATRIX COMPOSITES

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The damage and deformation behaviour of particle reinforced aluminum matrix composites can be highly sensitive to local variations in spatial distribution of reinforcement particles, which markedly depend on melt processing and solidification stages during production. The present study is aimed at understanding the mechanisms responsible for clustering of SiC particles in an Al-Si-Mg (A356) alloy composite during solidification process and establishing a model to predict the risk of cluster formation as a function of local solidification rate in a cast component. Special emphasis has been given to spatial characterization methods in terms of their suitability to characterize composite microstructures. Result indicate that methods that present a summary statistics on the global level of heterogeneity have limited application in quantitative analysis of discontinuously reinforced composites since the mechanical response of such materials are highly sensitive to dimensions, locations and spatial connectivities of clusters. The local density statistics, on the other hand, was observed to provide a satisfactory description of the microstructure, in terms of localization and quantification of clusters. A macrotransport - solidification kinetics model has been employed to simulate solidification microstructures for estimation of cluster formation tendency. Results show
that the distribution of SiC particles is determined by the scale of secondary dendrite arms (SDAS). In order to attain the lowest amount of particle clustering, the arm spacings should be kept within the limit of $2d_{SiC} > SDAS > d_{SiC}$, where $d_{SiC}$ is the average particle diameter.

*Keywords*: Discontinuously reinforced composites, clustering, thermal analysis, solidification modelling, quantitative metallography.
ÖZ

DÖKÜM YOLUYLA ÜRETİLMİŞ ALÜMİNYUM TABANLI
KOMPOZİTLERDE PARÇACIK TOPAKLANMASININ
DEĞERLENDİRİLMESİ VE MODELLEMESİ

Çetin, Arda
Doktora, Metalurji ve Malzeme Mühendisliği Bölümü
Tez Yöneticisi: Prof. Dr. Ali Kalkanlı

Nisan 2008, 114 sayfa

Parçacık takviyeli alüminyum tabanlı kompozitlerin hasar ve deformasyon tepkileri üretimin sivi faz ve katlama aşamalarında şekillenen parçacık dağılımasına yüksek duyarlılık göstermektedir. Bu çalışma SiC parçacıklarıyla desteklenmiş Al-Si-Mg (A356) kompozitlerin katlama sürecinde parçacık topaklanmasına yol açan mekanizmaları anlamayı ve döküm parçalarda yerel katlama hızına bağlı topak oluşma riskini değerlendirebilecek bir model oluşturmayı amaçlamaktadır. Çalışma kapsamında birçok uzamsal analiz yöntemi yer verilmiş ve kompozit mikroyapısı analizine uygunlukları değerlendirilmiştir. Sonuçlar, kompozitlerin mekanik tepkilerinin topakların boyutları, pozisyonları ve uzamsal bağlantılarıyla yakından ilişkili olması nedeniyle genel heterojenlik seviyesini özetleyen istatistiklerin bu malzemelerin karakterizasyonunda sınırlı kullanımı olduğunu göstermektedir. Yerel yoğunluk istatistiğinin sonuçları ise topakların niceliksel değerlendirmesi ve pozisyonlarının tespiti açılarında tatmin edici bulunmuştur. Topak oluşma eğilimini tahmin edebilmek amacıyla katlama mikroyapılarının benzetimi için makro transfer - katlama kinetiği yaklaşımı kullanılmıştır. Elde edilen benzetimsel mikroyapı düzeltmelerinin deneySEL sonuçlarla oldukça uyumlu oldukları görülmüştür. Sonuçlar, SiC parçacık dağılımının ikincil dendrit
kol aralıkları (İDKA) tarafından belirlendiğini göstermektedir. En düşük topaklanma seviyesine ulaşılmak için ikincil dendrit kol aralıklarının $2d_{sc} > \text{İDKA} > d_{sc}$, ($d_{sc}$ ortalama parçacık boyutunu göstermektedir) aralığında tutulması gerekmektedir.

Anahtar kelimeler: Parçacık takviyeli kompozitler, topaklanma, termal analiz, katılma modellemesi, niceliksel metalografi.
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Looking back from the end of this long journey, it is interesting to see how many wonderful people have contributed to this work in a huge variety of ways. This thesis is the account of approximately five years of devoted work, which would not have been possible without the help of many.

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# TABLE OF CONTENTS

ABSTRACT ....................................................................................... iv  
ÖZ ................................................................................................. vi  
ACKNOWLEDGEMENTS ........................................................... viii  
TABLE OF CONTENTS ............................................................... x  
NOMENCLATURE ..................................................................... xiii

CHAPTERS

1 INTRODUCTION ............................................................................. 1

2 QUANTITATIVE ANALYSIS OF PARTICLE DISTRIBUTION .......... 4  
   2.1 Brief Review of Literature ....................................................... 4  
   2.2 Spatial Analysis Methods ......................................................... 6  
      2.2.1 Refined nearest neighbour analysis .................................. 6  
      2.2.2 K-Function ....................................................................... 8  
      2.2.3 Inference of local clustering ........................................... 9  
      2.2.4 Visualization of clusters ............................................... 11  
      2.2.5 Voronoi tessellation ....................................................... 12

3 THERMAL ANALYSIS OF MMC SOLIDIFICATION...................... 13  
   3.1 Newtonian and Fourier Thermal Analysis Methods .................. 13  
      3.1.1 Calculation of latent heat of solidification ....................... 18  
      3.1.2 Calculation of instantaneous solid fraction ....................... 19  
   3.2 Thermal Analysis of Composite Solidification ....................... 19
3.3 Dendrite Coherency Point ......................................................... 21

4 MODELLING OF MMC SOLIDIFICATION .................................... 23
4.1 Theoretical Formulation of Macroscopic Heat Transfer ............... 24
4.2 Microscopic Modelling ............................................................. 26
  4.2.1 Pseudo-binary alloy assumption: Calculation of equivalent solute ... 26
  4.2.2 Nucleation of $\alpha$-Al dendrites ........................................... 27
  4.2.3 Dendritic growth ............................................................... 28
  4.2.4 Eutectic nucleation ........................................................... 30
  4.2.5 Eutectic grain growth and impingement ............................... 31
  4.2.6 Coarsening of secondary dendrite arms ............................... 32

5 EXPERIMENTAL & COMPUTATIONAL DETAILS ......................... 38
5.1 Materials ................................................................................. 38
  5.1.1 Matrix alloy ........................................................................ 38
  5.1.2 The reinforcement phase ...................................................... 39
5.2 Stir Casting of Aluminum Matrix Composites ......................... 41
5.3 Thermal Analysis of MMC Solidification .................................. 44
5.4 Image Analysis ........................................................................ 45
5.5 Computational Details .............................................................. 47
  5.5.1 Quantitative analysis of particle distribution ......................... 47
    5.5.1.1 Computer generated point data ...................................... 47
    5.5.1.2 Spatial analysis programs ............................................. 48
  5.5.2 Macrotransport - solidification kinetics modelling ............... 48

6 RESULTS & DISCUSSION ................................................................. 51
6.1 Thermal Analysis of Composite Solidification ......................... 52
  6.1.1 Evolution of solid fraction .................................................. 54
  6.1.2 Comparison of dendrite coherency point estimations .............. 56
6.2 Quantitative Analysis of Particle Distribution ......................... 58
  6.2.1 Simulated point data .......................................................... 58
  6.2.2 Quantitative analysis of composite microstructures ............. 66
    6.2.2.1 Refined nearest neighbour analysis ............................. 66
    6.2.2.2 Voronoi tessellation .................................................... 69
    6.2.2.3 $K$-function .............................................................. 71
6.2.2.4 Local density statistics .................................................. 73
6.2.2.5 Effect of metallographic field size on cluster dimensions .......... 79
6.2.2.6 Comparison of methods .................................................. 82
6.3 Effect of Solidification Rate on Clustering of SiC Particles .......... 83
   6.3.1 Effect of solidification rate on dendritic structure ................ 83
   6.3.2 Effect of solidification rate on particle distribution ............... 84
6.4 Modelling of MMC Solidification ............................................. 90
   6.4.1 Prediction of local solidification rate ................................ 90
   6.4.2 Prediction of secondary dendrite arm spacings ................... 96
7  CONCLUSIONS ......................................................................... 99
REFERENCES ............................................................................. 102
APPENDICES
   A. Phase diagram and solidification path of A356 alloy .................. 107
   B. Mold dimensions and thermocouple locations ........................... 111
VITA ......................................................................................... 113
## NOMENCLATURE

<table>
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<td>eutectic growth constant</td>
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<td>$\mu_N$</td>
<td>nucleation parameter</td>
<td>m$^2$ °C$^{-2}$</td>
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\[ \rho \] density \hspace{2cm} \text{kg m}^{-3} \\
\[ \sigma \] standard deviation \\
\[ \xi \] significance parameter \\
\[ \psi \] impingement correction factor \\
\[ \Gamma \] Gibbs-Thomson coefficient \hspace{2cm} \text{m °C} \\

**SUPERCRIPTS & SUBSCRIPTS USED**

\[ A \] area \\
\[ L \] liquid \\
\[ P \] particle \\
\[ S \] solid \\
\[ \text{comp} \] composite \\
\[ \text{eq} \] equivalent \\
\[ \text{eut} \] eutectic \\
\[ g \] growing \\
\[ m \] mold \\
\[ \text{matrix} \] \\
\[ \text{obs} \] observed \\
\[ pois \] Poisson \\
\[ s \] shrinking \\
\[ \infty \] surroundings
CHAPTER 1

INTRODUCTION

Metal matrix composites (MMC) have proven to offer distinctive advantages over a number of conventional materials being used in aerospace, ground transportation (auto and rail), defense, thermal management and infrastructure industries. These advantages include improved strength, stiffness, fatigue and wear resistances with good thermal properties, while maintaining low weight. Another advantage is that the extent of such improvements can be tailored by altering the type, size and morphology of the reinforcement phase. There are two basic groups of reinforcements, which are referred as continuous and discontinuous. Continuous reinforcements are typically fibers or monofilament wires, offering attractive improvements in longitudinal properties by sacrificing transverse properties. Isotropic improvements can be obtained by employing discontinuous reinforcements, such as whiskers, particles and short fibers. Among these alternatives, the most attractive options for commercial practice are silicon carbide and alumina in the form of “particles” with aluminum alloy matrices due to their distinct advantage in terms of affordability. Hence, in today’s industry, discontinuously reinforced aluminum composites (DRA) account for majority of MMC annual production.

Due to extensive research on production and characterization of such materials, today DRA are an established technology. However, there are still certain problems, one and most important, being the clustering or agglomeration of reinforcement particles. The extent of property degradation associated with particle clustering have numerous been underlined by many researchers throughout the period covering from early 80’s, where DRA technology was first emerged, onwards to very recent investigations. Although certain solutions were derived by altering the processing method (such as infiltrating the liquid metal to a packed preform of reinforcements) or controlling processing variables
(such as the particle size ratio in powder metallurgy routes), no neat solutions could be proposed for casting processes, where bulk of MMC production are carried out. There are only certain beliefs about solidification routes that finer dendritic arm spacings would produce uniform distribution of reinforcements (which is proved to be wrong in this study), however no quantitative data were reported in the literature.

In order to understand the origin of clustering problem and to be able to quantify the amount of reinforcement clustering, one should have a very clear idea of what a cluster is. The literature on this aspect of MMC technology sadly lacks a thorough understanding of spatial characterization methods. There are only a few studies that came into prominence in this aspect, which are briefly reviewed in the following pages. In general, there is an ambiguity concerning the type of information that an engineer needs regarding the microstructure of a MMC. The problem is that, the terms inhomogeneity and clustering are commonly used as synonyms and the microstructures of MMCs are considered in terms of global trends in the distribution. However, as far as the fracture mechanics of MMCs is concerned, one also needs to gather information on the amount, locations and spatial connectivities of reinforcement clusters, since failure mechanisms of such materials include crack nucleation and propagation with a high sensitivity to cluster locations. Therefore attempting to characterize MMC microstructures by simple scalar descriptions, although holds a practical value, lacks a great deal of information to successfully associate the mechanical response of the composite to its microstructure.

The present study is structured in three parts. The first part attempts to answer the questions; what is a cluster and how can it be detected? The effort to answer these questions actually forms the backbone of the study, since the clusters can neither experimentally be detected nor computationally predicted without a clear understanding of what we are looking for. The second part is an experimental study on relation of clustering with local microstructure, which will help to understand cluster formation tendencies as a function of location within castings with complex geometries. The third and final part is a computational study aimed at simulating solidification microstructures as a function of local solidification time and therefore to predict the tendency of reinforcement clustering. Putting these parts together results in a thorough picture in which certain locations in a casting, where there exists a tendency towards clustered particle arrangements can be predicted before production. Such an ability will obviously help engineers to come up
with solutions to produce these components with more uniform microstructures and hence, less prone to failure. These solutions may include interventions to production process such as refinement of microstructure or varying reinforcement size, or altering component designs to improve microstructural uniformity.

Due to combinatorial nature of the present work, the theoretical background is presented in three different chapters. Firstly, the spatial characterization methods are given in Chapter 2. Chapter 3 focuses on thermal analysis of metal matrix composites, which provides a clear description of MMC solidification. Chapter 4 introduces the numerical approach to MMC solidification. Chapter 5 presents the experimental and computational details, followed by the results and discussion in Chapter 6. Finally, concluding remarks are summarized in Chapter 7.
2.1 Brief Review of Literature

It is well known that introduction of hard ceramic particles into soft metallic matrices leads to profound improvements in mechanical properties such as increased strength, stiffness and fatigue resistance while maintaining low weight\(^1,2\). The origin of these property enhancements is primarily attributed to two mechanisms. In the first one, the strengthening is ascribed to direct load transfer from the matrix to the reinforcement phase\(^3,6\). The second mechanism, on the other hand, attributes strengthening of the material to increasing dislocation density of matrix due to developed residual plastic strain, which results from thermal mismatch between the ceramic particles and the matrix material\(^5,9\).

In structural composites, one of the most critical design criteria is fatigue resistance, which is particularly important in automotive and aerospace industries where resistance to high cycle fatigue resistance is strictly necessary\(^6\). The failure process of discontinuously reinforced composites is generally described over two stages; damage localization and damage globalization\(^10\). The first stage includes various mechanisms such as particle and matrix interface debonding, particle fracture, void formation in the ductile matrix, short crack initiation and crack coalescence. The second stage, on the other hand, consists of a long crack growth stage, which follows Paris’ Law in the intermediate stress intensity factor range\(^10\). There are various factors that influence localization and globalization of the damage. One such parameter is the size of reinforcement particles. In
general, as the particle size increases, it is more likely that it contains a surface crack\textsuperscript{11, 12}. This assumption was verified through various experimental studies that directly relate the particle cracking frequency with increasing particle size\textsuperscript{2, 13}. Orientation and morphology of the reinforcing particles were also reported to contribute to damage behaviour of the composites\textsuperscript{14, 15}. One other important factor that determines the mechanical response of the composite is the spatial distribution of second phase particles. Although the aforementioned factors other than spatial arrangement of particles can well be altered by varying the size or morphology of reinforcements, control of particle distribution requires a thorough understanding of the factors that determine their arrangement.

Composite microstructures often display clustered arrangements. Although such clusters were reported to have negligible effect on elastic properties,\textsuperscript{16, 17} they have profound influence on damage evolution and failure mechanisms. One such influence is the stress localization in these regions\textsuperscript{15}, which increases the probability of particle failure to a significant extent\textsuperscript{17, 18}. Presence of particle clusters was also reported to decrease the yield strength, strain hardening rate and failure strain of the composite over the monolithic material\textsuperscript{16}. The origin of this property degradation is attributed to preferential nucleation of cracks\textsuperscript{12, 14} in clustered regions and final fracture is produced by crack propagation through the matrix to other clusters\textsuperscript{11, 19, 20}. Finally, Ayyar and Chawla’s\textsuperscript{14, 21} work have shown through finite element simulations that the crack growth resistance of the material is also lower in composites with clustered distributions compared to random arrangements.

The very first attempts in quantitative metallography of metal matrix composites were carried out by Wray et al\textsuperscript{22} and Spitzig et al\textsuperscript{23} by characterizing the composite microstructures by Dirichlet tessellation and nearest neighbour methods. Among the various studies that came into prominence in this field; Everett et al\textsuperscript{24} have compared the Monte-Carlo based computer generated patterns with actual composite microstructures by utilizing Dirichlet tessellation, nearest neighbour statistics and radial distribution function. Various characterization functions of Voronoi cell based geometric parameters for characterization and response modeling of composites was introduced by Ghosh et al\textsuperscript{25}. Li et al\textsuperscript{26} computationally constructed 3D microstructure models by sequentially assembling digital section micrographs obtained by serial sectioning and presented a systematic approach to 2D and 3D microstructural characterization. A computational approach based
on Voronoi tessellation to determine the local reinforcement area fraction contour maps was adopted by Ganguly and Poole\textsuperscript{27}. Finally, Scalon \textit{et al}\textsuperscript{28} investigated the distribution of second phase particles by various pattern descriptor functions and modeled their spatial distribution by a Strauss point process model.

Those studies have either concentrated on application of various quantitative characterization methods or attempted to correlate the spatial configuration of second phase particles to the mechanical response of composite systems. They have mainly described the observed spatial patterns by considering the global trends in the distribution of particles and used suitable descriptors to discriminate between random and clustered arrangements. However, such approaches may fail to predict the actual failure susceptibility of these materials since damage and deformation behaviour of composites can be highly sensitive to local variations in particle content and spatial correlations between these local variations. For example, previous research\textsuperscript{18, 19} on fracture behaviour of discontinuously reinforced composites showed that in composites with clustered particle arrangements, damage preferentially initiates from clusters and final fracture is produced by crack propagation through the matrix to other clusters. Therefore, the spatial heterogeneity in these systems should be characterized by considering the locations, dimensions and spatial connectivity of the clusters rather than simple scalar descriptions of microstructures.

2.2 Spatial Analysis Methods

2.2.1 Refined Nearest Neighbour Analysis

The refined nearest-neighbour analysis\textsuperscript{29} is based on comparison of the complete distribution functions of either $w$, the nearest-neighbour distance between events ($G$-function), or $x$, the distance from a sampling point (not event) to the nearest event ($F$-function) with that of expected distribution of events for complete spatial randomness (CSR). The theoretical cumulative distribution function of nearest-neighbour distances under the null hypothesis of CSR is given by
\[ G(w) = 1 - \exp(-\gamma \pi w^2) \]  

(2.1)

where \( \gamma \) is the intensity. An appropriate edge corrected estimate of the observed distribution function would be

\[
\hat{G}(w) = \frac{\sum I(w_i \leq w, r_i > w)}{\sum_i I(r_i > w)}
\]  

(2.2)

where \( I \) is the indicator function that denotes the count of events, and \( r_i \) and \( w_i \) are the distances from the \( i \)th event to its nearest boundary and to its nearest neighbour, respectively. The deviation of an observed pattern from randomness can be detected from the difference of estimates of observed and theoretical distribution functions\(^{29}\). The deviation of an observed pattern from CSR can also be brought out by utilizing the ratio of means, given by

\[
Q = \frac{E_{\text{obs}}(\bar{w})}{E_{\text{pois}}(\bar{w})}
\]  

(2.3)

where \( E_{\text{obs}}(\bar{w}) \) is the observed mean of nearest neighbour distances and \( E_{\text{pois}}(\bar{w}) \) is the expected mean of nearest neighbour distances for a Poisson process. The expected mean of a Poisson distribution is given by the expression\(^{30}\)

\[
E_{\text{pois}}(\bar{w}) = 0.5 \left( \frac{N}{A} \right)^{-0.5}
\]  

(2.4)

where \( N/A \) gives the area density of the events within the study region. Different types of spatial distributions can therefore be classified according to:

- \( Q \approx 1 \) denotes random event sets,
- \( Q < 1 \) implies clustered distribution,
- \( Q > 1 \) implies uniform distribution.
2.2.2 $K$-Function

One drawback of the refined nearest-neighbour distance methods is that they only consider the distances to the closest events, ignoring the larger scales of pattern. The $K$-function, on the other hand, provides a summary of spatial dependence over a wide range of scales including all event to event distances, not only the nearest-neighbour distances. The most commonly used edge-corrected estimator of the $K$-function is given by Ripley\textsuperscript{31} as

\[
\hat{K}(t) = \frac{1}{\gamma} \sum_i \sum_{j \neq i} \frac{I_i(u_{ij})}{w_{ij}}
\]  

(2.5)

where $\gamma$ is the intensity, $u_{ij}$ is the distance between the $i$th and $j$th events and $I_i$ is the indicator function that denotes the count belonging to a value of $t$ for the distance comparisons $u_{ij} \leq t$. The weight function $w_{ij}$ provides the edge correction by considering the proportion of the circumference of the circle around event $i$. The $K$-function for a homogeneous Poisson distribution of events is given by $K(t) = \pi t^2$. $K$-function is commonly transformed to

\[
\hat{L}(t) = \sqrt{\frac{\hat{K}(t)}{\pi}} - t
\]  

(2.6)

as Eqn. (2.6) yields a theoretical value of zero under the null hypothesis of CSR. Positive values above the confidence interval suggest the presence of clusters whereas negative values below the confidence interval suggest a uniform distribution of events.
2.2.3 Inference of Local Clustering

Although the $K$-function successfully measures the local density around each event over many distance scales, the presence of clustering can be inferred only when the local density around an event exceeds a certain threshold value. In order to determine this threshold value and to estimate the local amount of clustering around an event, we have adopted a cumulative radial distribution function, $\rho(P_i, r)$, which in the present study is defined as the number of reinforcement particles per unit area within the radius $r$ from an original particle $P_i$, divided by the number of particles per unit area of the whole study region. Fig. 2.1 illustrates the evaluation of $\rho(P_i, r)$.

![Figure 2.1](image)

Figure 2.1 Evaluation procedure of $\rho(P_i, r)$ illustrated at five distance scales; $r_1$, $r_2$, $r_3$, $r_4$ and $r_5$. $\rho(P_i, r)$ is also illustrated for particles $P_{81}$ and $P_{114}$ at distance scales of 100 and 160 units, respectively.
A similar concept of density calculation was previously introduced by Prodanov et al\textsuperscript{32} by defining a parameterized threshold function $T_\xi(r)$, which expresses the probability that the local density around a particle, as depicted by $\rho(P_i, r)$, exceeds a certain threshold value in an associated Poisson (random) point process. In the present study the threshold function $T_\xi(r)$ for each study region was estimated from 200 simulations of homogeneous Poisson process with the same intensity as the studied pattern. The significance parameter $\xi$ was set to 0.95; meaning that only 5\% of the points in an associated Poisson point process were expected to exceed the threshold $T_{0.95}(r)$. Therefore, a particle in the study region can be inferred as a part of a cluster if the local density around that particular particle exceeds the amount present in an associated Poisson process; $\rho(P_i, r) > T_{0.95}(r)$, at a particular radius, $r$. In order to consider the edge effects, a weighted edge correction scheme, which is based on weighing the proportion of the circumference of the disc that remains inside the study region to entire circumference, was applied\textsuperscript{11}.

An important aspect of this approach is that the local density around a particle may or may not exceed the corresponding threshold value depending on the scale of observation, $r$. Following the method of Prodanov et al\textsuperscript{32} the above-threshold particles, which were accepted to be a part of a cluster were updated at each scale of observation and the information regarding the coordinates of these particles were collected in a set of above-threshold particles, $S(r)$. The percent ratio of above-threshold events averaged over all scales of observation is then calculated from

\[
\%\text{Threshold} = \frac{\frac{1}{T} \int_0^T N(t)dt}{N_{\text{events}}} \tag{2.7}
\]

where $N_{\text{events}}$ is the total number of events in the study region and $T$ denotes the overall scale of observation.
2.2.4 Visualization of Clusters

In order to reveal the spatial correlations between the detected clusters, one needs to visualize the locations, dimensions and relative intensities of the clusters. One common way to obtain a spatially smooth intensity estimate of points within a study region is the kernel estimation\textsuperscript{33}. The intensity, \( \gamma(x) \), at location \( x \) can be estimated by

\[
\hat{\gamma}_h(x) = \frac{1}{n} \sum_{i=1}^{n} \frac{1}{h^2} k\left(\frac{x-x_i}{h}\right)
\]

(2.8)

where \( h \) is the bandwidth parameter that regulates the degree of smoothness, \( x \) is the location for intensity estimation, \( x_i \) is the observed event location, \( n \) is the number of points and \( k\{\} \) represents the kernel weighting function. Scalon \textit{et al.}\textsuperscript{28} successfully utilized this method for intensity estimation of the second phase particles in an Al/SiC\textsubscript{p} composite. A more refined approach to visualize the clusters would be to associate the set of above-threshold particles to the sum of kernels by only considering the above-threshold particles that belong to \( S(r) \), at a particular scale of observation, \( r \). With this approach, one can locate the clusters and compare their relative intensities instead of complicated contour plots that represent the intensity variation of the whole data set. Since different suitable alternatives of the kernel weighting functions have relatively small effect on the resulting intensity estimate,\textsuperscript{34} we have implemented a simple quartic density function denoted by

\[
k(u) = \begin{cases} 
\frac{3}{\pi} (1-u^T u)^2 & \text{for } |u| < 1 \\
0 & \text{otherwise}
\end{cases}
\]

(2.9)

where \( |u| \) represents the Euclidean distance of the vector \( x \) (e.g. \( \sqrt{u^T u} = x \)). The relative intensities of the above-threshold particles can therefore be estimated by

\[
\hat{\gamma}_h(x) = \sum_{i \in S(r)} \frac{3}{\pi h^2} \left[ 1 - \frac{d_i^2}{h^2} \right]^{-2}
\]

(2.10)
where, \( d_i \) is the distance between the location of intensity estimation and the observed particle location, and the summation is only over values of \( d_i \) which do not exceed \( h \).

### 2.2.5 Voronoi Tessellation

The Voronoi diagram\textsuperscript{35} built on a set of events represents partition of the area into a set of adjacent regions that have the shape of convex polygons such that each polygon contains exactly one event. The relevant properties of a Voronoi polygon, such as its area, may be used to distinguish between different point processes. The dispersion of the probability density function, known as the coefficient of variation, can be defined for polygon area as

\[
CV_A = \frac{\sigma_A}{\mu_A}
\]  

(2.11)

where \( \sigma \) is the standard deviation and \( \mu \) is the mean of the probability density function, and the subscript \( A \) refer to polygon area. A lower \( CV_A \) value indicates uniform distribution whereas higher values imply clustering of events.
3.1 Newtonian and Fourier Thermal Analysis Methods

Newtonian\textsuperscript{36} and Fourier\textsuperscript{37} thermal analysis methods are simplified but convenient techniques to monitor the average temperature variations and phase changes during the course of solidification. They are actually applicable at the limiting cases when solidification is fully controlled by the thermal resistance at the metal / mold interface or by low thermal diffusivity of the metal.

Both methods are based on interpretation of cooling curves obtained from one or more locations in the casting. The difference of these method is the way their baseline, or zero curve, is generated. The zero curve represents the hypothetical variation of the metal’s cooling rate, if there would be no phase transformation within the covered temperature range. Newtonian analysis neglects the presence of thermal gradients and generates the zero curve from the thermal data obtained from a single thermocouple. In Fourier analysis, on the other hand, the zero curve is a function of temperature Laplacian and therefore requires minimum of two (in cylindrical geometry) or three (in Cartesian geometry) thermocouples.

As mentioned above, in Newtonian analysis only one thermocouple, placed at the geometrical center of the casting is used and the presence of thermal gradients is neglected. This assumption is valid only when the Biot number for a particular geometry and metal-mold system is less than 0.1. Biot number\textsuperscript{38} is a dimensionless number that relates the resistance to heat transfer inside and at the surface of a sample (i.e. the metal-mold interface in the present case). Values smaller that 0.1 imply that the heat conduction
inside the solidifying sample is much faster than at the metal-mold interface, so that the
temperature gradients within the sample may be neglected. In all samples analyzed in the
present study, the Biot numbers were verified to be smaller than 0.1 for validity of the
Newtonian thermal analysis procedure.

In Newtonian analysis, the zero curve is derived from a simple energy balance for
a hypothetical case where the metal does not go under any phase transformation. The rate
of heat loss to the surroundings is given by the expression

\[
\frac{dQ}{dt} = -h_i A(T - T_\infty)
\]  (3.1)

where \(dQ/dT\) is the rate of heat loss to the surroundings, \(t\) is time, \(h_i\) is the coefficient of
heat loss to the surroundings, \(A\) is the effective surface area over which heat transfer
occurs, \(T\) is instantaneous temperature and \(T_\infty\) is the temperature of the surroundings. The
rate of heat evolved to the surroundings can be described by the equation

\[
\frac{dQ}{dt} = V \rho c_p \frac{dT}{dt}
\]  (3.2)

where \(V\) is the volume of solidifying metal, \(\rho\) is density and \(c_p\) is the specific heat
capacity. If the metal does not go under any phase transformation within the covered
temperature range, these two equations can be equated to yield

\[
\frac{dT}{dt} = -\left(\frac{h_i A}{V \rho c_p}\right)(T - T_\infty)
\]  (3.3)

Eqn. (3.3) represents the derivative of the cooling rate of a sample, which does not
go under any phase transformation; i.e. the zero curve. However, there are certain
difficulties associated with this equation. First one is the calculation of the heat transfer
parameter, since it is affected from many parameters such as; variations in pouring
temperature, sample geometry and reactions between the sample and the sand mold. The
value of specific heat is the second potential difficulty, since it is a strong function of
composition and temperature, and therefore depends on the relative amounts of phases present at any temperature. These difficulties can be overcome by using the thermal data obtained from cooling curve to generate the zero curve. Rearranging and integrating Eqn. (3.3) yields

\[
\int_{T_i}^{T} \frac{dT}{T-T_\infty} = -\int_{0}^{t} \frac{h_i A}{\rho \alpha c_p} dt
\]

(3.4)

\[\ln \left( \frac{T-T_\infty}{T_i-T_\infty} \right) = -\left( \frac{h_i A}{\rho \alpha c_p} \right) t\]

(3.5)

Rearranging to give the instantaneous temperature;

\[T = (T_i - T_\infty) \exp \left( -\frac{h_i A}{\rho \alpha c_p} \right) t + T_\infty\]

(3.6)

where \(T_i\) is the initial temperature at \(t = 0\). Derivative of Eqn. (3.6) finally yields the expression for zero curve

\[\left( \frac{dT}{dt} \right)_{zc} = -(T_i - T_\infty) \frac{h_i A}{\rho \alpha c_p} \exp \left( -\frac{h_i A}{\rho \alpha c_p} t \right)\]

(3.7)

Eqn. (3.7) can be rewritten as

\[\left( \frac{dT}{dt} \right)_{zc} = -C_1 C_2 \exp(-C_2 t)\]

(3.8)

where the subscript \(zc\) denotes the zero curve and \(C_1 = T_i - T_\infty\) and \(C_2 = \frac{h_i A}{\rho \alpha c_p}\) are constants to be determined experimentally.
Therefore, one can obtain the Newtonian zero curve experimentally by curve fitting of an exponential function from one no-phase transformation region (i.e. above liquidus) to the other (i.e. after solidification is completed). The obvious source of error in this approach is the chosen time steps for curve fitting, which can create significant differences in the resulting zero curve. There are various suggestions for curve fitting of the arbitrary function in the literature\textsuperscript{39, 40} however; no common agreement or a procedure could be established to obtain this curve.

The derivation of the Fourier zero curve is much more straightforward. It starts with the Fourier equation, including a heat source;

\[
\frac{\partial T}{\partial t} = \alpha \nabla^2 T + \frac{1}{c_v} \frac{\partial Q}{\partial t}
\]  

(3.9)

where \( \alpha \) is the thermal diffusivity, \( \nabla T \) is the temperature Laplacian and \( c_v \) is the volumetric specific heat and \( Q \) is the heat evolved during solidification. Rearranging

\[
\frac{\partial Q}{\partial t} = c_v \left( \frac{\partial T}{\partial t} - Z_F \right)
\]  

(3.10)

where \( Z_F = \alpha \nabla^2 T \) is the Fourier zero curve. In order to obtain the Fourier zero curve, therefore, one should be able to describe the temperature Laplacian within the solidifying casting. In a Cartesian geometry, a minimum of three thermocouples are necessary to describe the temperature field. However, the required number of thermocouples can be reduced to two in a symmetric temperature field\textsuperscript{37}. The temperature Laplacian in a cylindrical casting, which is symmetrical with respect to the vertical axis, is given by

\[
\nabla^2 T = \frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r}
\]  

(3.11)
When two thermocouples are placed in a cylindrical casting at two different locations, \( P_1 \) and \( P_2 \), from the center, the differentials in Eqn. (3.11) can be expressed as

\[
\nabla^2 T = \frac{T_2 - T_1 - 0}{P_2 - P_1} \left( \frac{P_2 - P_1}{P_1 + \frac{P_2 - P_1}{2}} \right) + \frac{1}{P_1 + \frac{P_2 - P_1}{2}} \left( \frac{T_2 - T_1}{P_2 - P_1} \right)
\]

(3.12)

where \( T_1 \) and \( T_2 \) are temperature values read from thermocouple locations \( P_1 \) and \( P_2 \), respectively. Upon rearranging, Eqn. (3.12) reduces to

\[
\nabla^2 T = \frac{4(T_2 - T_1)}{P_2 - P_1}
\]

(3.13)

When there is no phase transformation \((Q_s = 0)\) the thermal diffusivity of the system can be obtained from Eqn. (3.10) as

\[
\alpha = \frac{\partial T}{\partial t} \frac{1}{\nabla^2 T}
\]

(3.14)

Therefore, when the cooling rates and Laplacians are known before and after solidification, thermal diffusivities of the solid, \( \alpha_s \), and liquid, \( \alpha_l \), can be calculated. The instantaneous values of the thermophysical properties during solidification can be calculated by an iterative method. The iteration starts with a linear assumption of the variation in solid fraction

\[
f_s = \frac{t - t_{end}}{t_{end} - t_{start}}
\]

(3.15)

where, \( t \) is the instantaneous time and the subscripts \( start \) and \( end \) denote the start and end of solidification, respectively. At each time step, the instantaneous values of \( \alpha \) and \( c_s \) of the solid liquid mixture are calculated as
\[ \alpha = \alpha_L \left[1 - f_S \right] + \alpha_S f_S \quad (3.16) \]
\[ c_v = c_{v,L} \left[1 - f_S \right] + c_{v,S} f_S \quad (3.17) \]

where, \( c_{v,L} \) and \( c_{v,S} \) are the volumetric heat capacities of the liquid and solid, respectively.

### 3.1.1 Calculation of Latent Heat of Solidification

As mentioned above, the zero curve represents the variation in the cooling rate of a hypothetical sample if it would go under no phase transformation. Consequently, it should be clear that, the difference between the cooling rate of a sample (i.e. the derivative of cooling curve) and its zero curve results from the released latent heat associated with the solidification of the sample. Therefore, one can calculate the amount of released latent heat by quantifying this difference.

Latent heat calculation in Newtonian analysis starts with including the heat generation term. When phase transformation occurs, Eqn. (3.3) can be expressed as

\[ \frac{dQ}{dt} = V \rho c_p \frac{dT}{dt} + \frac{dQ_L}{dt} \quad (3.18) \]

where \( Q_L \) is the heat resulting from latent heat release during phase transformation. Combining Eqns. (3.1) and (3.18) and rearranging

\[ \left( \frac{dT}{dt} \right)_{cc} = \frac{1}{V \rho c_p} \left( h_i A (T - T_e) - \frac{dQ_L}{dt} \right) \quad (3.19) \]

The magnitude of latent heat released during solidification can be calculated by subtracting Eqn. (3.7) from Eqn. (3.19) and integrating within the interval of solidification

\[ L = -c_p \left[ \int_{t_e}^{t_s} \left( \frac{dT}{dt} \right)_{cc} \ dt - \int_{t_e}^{t_s} \left( \frac{dT}{dt} \right)_{zc} \ dt \right] \quad (3.20) \]
In Fourier analysis, latent heat is calculated by integrating Eqn. (3.10). This integrated form is actually very similar to Eqn. (3.20) since the zero curve term is already included in Eqn. (3.10). The governing equation is

$$L = \int_{t_s}^{t_e} \left( \frac{\partial Q}{\partial t} \right) dt \quad (3.21)$$

where $t_s$ and $t_e$ denote the start and end of solidification, respectively.

### 3.1.2 Calculation of Instantaneous Solid Fraction

The solid fraction evolution is calculated with the assumption that the latent heats associated with primary and eutectic solidifications are equal. The instantaneous fraction of the solid phase at time $t$ can therefore be calculated as

$$f_s(t) = \frac{1}{L} \int_{t_s}^{t} \left( \frac{\partial Q}{\partial t} \right) dt \quad (3.22)$$

### 3.2 Thermal Analysis of Composite Solidification

Newtonian and Fourier methods need some corrections to be applied to composite solidification. Firstly, the reinforcement particles do not go under any phase transformation during solidification of the matrix alloy. Therefore the mass content of particles should be considered since the latent heat values are calculated per unit mass. The latent heat associated with the solidification of the composite therefore can be expressed as

$$L_{\text{comp}} = L \left( 1 - M_p \right) \quad (3.23)$$
where $M_p$ is the mass content of particles and the subscript $comp$ denotes composite. The second point to be corrected is the effect of particles on the thermophysical properties of the composite materials. The governing equations are presented in Table 3.1.

**Table 3.1. Equations for thermophysical properties**

<table>
<thead>
<tr>
<th>Temp Interval</th>
<th>Derived Equations</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T &gt; T_L$</td>
<td>$c_p^{comp} = \frac{V_{SiC} \cdot \rho_{SiC} \cdot c_p^{SiC} + (1-V_{SiC}) \cdot c_p^{L} \cdot \rho_{L}}{\rho_{comp}}$ (3.24)</td>
</tr>
<tr>
<td></td>
<td>$\rho_{comp} = V_{SiC} \cdot \rho_{SiC} + (1-V_{SiC}) \cdot \rho_{L}$ (3.25)</td>
</tr>
<tr>
<td></td>
<td>$c_p^{comp} = \frac{V_{SiC} \cdot \rho_{SiC} \cdot c_p^{SiC} + (1-V_{SiC}) \cdot c_p^{m} \cdot \rho_{m}}{\rho_{comp}}$ (3.26)</td>
</tr>
<tr>
<td>$T_L &gt; T &gt; T_{end}$</td>
<td>$\rho_{comp} = V_{SiC} \cdot \rho_{SiC} + (1-V_{SiC}) \cdot \rho_{m}$ (3.27)</td>
</tr>
<tr>
<td></td>
<td>$c_m^{u} = c_p^{L} \cdot (1-f_s) + c_p^{S} \cdot f_s$ (3.28)</td>
</tr>
<tr>
<td></td>
<td>$\rho_m = \rho_{L} \cdot (1-f_s) + \rho_{S} \cdot f_s$ (3.29)</td>
</tr>
<tr>
<td>$T_{end} &gt; T$</td>
<td>$c_p^{comp} = \frac{V_{SiC} \cdot \rho_{SiC} \cdot c_p^{SiC} + (1-V_{SiC}) \cdot c_p^{S} \cdot \rho_{S}}{\rho_{comp}}$ (3.30)</td>
</tr>
<tr>
<td></td>
<td>$\rho_{comp} = V_{SiC} \cdot \rho_{SiC} + (1-V_{SiC}) \cdot \rho_{S}$ (3.31)</td>
</tr>
</tbody>
</table>

where $V_{SiC}$ is the volume content of SiC particles, and the sub- and superscripts $l, s, m$ and SiC denote liquid, solid, matrix and SiC particles, respectively.
3.3 Dendrite Coherency Point

Dendrite coherency point (DCP) is defined as the solid fraction at which the freely growing equiaxed dendrites impinge upon each other and a rigid, solid network of dendrites is established throughout the casting. It also defines a transition from mass feeding to interdendritic feeding\textsuperscript{41}. Casting defects such as interdendritic porosity, macrosegregation and hot tears start to develop after DCP\textsuperscript{42}.

SiC particles are not suitable substrates for nucleation of $\alpha$-Al. The dendrites therefore nucleate on available substrates other than SiC particles and push the reinforcement particles into the interdendritic regions, resulting in a segregated pattern of particles. Within the framework of the present study, we have also analyzed whether DCP can be regarded as an index to the distribution of second phase particles, since the distance scales that the particles are being pushed by the growing dendrites are related to the point where impingement occurs.

DCP can be determined by either mechanical methods\textsuperscript{43, 44} or thermal analysis\textsuperscript{45, 46}. The mechanical methods are based on rheological measurements during solidification. The abrupt increase in viscosity of the melt at some stage during solidification is attributed to impingement of primary dendrites. Thermal analysis method, on the other hand, monitors the variation in thermal gradient during the course of solidification. One thermocouple is placed at the geometrical center of the casting and another next to the mold wall and the temperature difference, $\Delta T$, read from these thermocouples are recorded. When the freely growing dendrites touch each other and a solid network is established all throughout the casting, the rate of heat transfer increases due to higher thermal conductivity of the solid phase. This results in a decrease in the magnitude of the thermal gradient in the casting, which can be monitored from the $\Delta T$ curve in Fig. 3.1. The first minimum in this curve is the dendrite coherency since the magnitude of thermal gradient starts to decrease after this point.
Figure 3.1 Cooling curves obtained from the center ($T_{\text{center}}$) and wall ($T_{\text{wall}}$) thermocouples and the $\Delta T$ curve for an A356 alloy reinforced with 20\% SiC$_p$. The first minimum in the $\Delta T$ curve is taken as the dendrite coherency point (DCP).
CHAPTER 4

MODELLING OF MMC SOLIDIFICATION

The present study is aimed at building a comprehensive approach to MMC solidification to assess the as-cast microstructure and reinforcement distribution. Predicting the cluster formation tendency in a part before casting would provide a distinct advantage due the significant savings in prototyping and production costs. Such an ability would also help engineers to come up with new designs to produce these parts with a more uniform microstructure and therefore less prone to failure.

The distribution of second phase particles during solidification is determined by the primary dendrites. Subsequent reactions after dendritic solidification, such as the eutectic reaction or solidification of various intermetallic phases do not yield any influence on the final arrangement of particles, since the particles are already stabilized within the solid network of dendrite arms during the period of these transformations. Therefore, an attempt to assess the as-cast particle distribution through numerical simulations should concentrate on predicting the local solidification rate, since the dendritic spacings (both primary and secondary) are functions of the local cooling rate\(^{47}\).

The main focus of the present work regarding the solidification process is, therefore, correct prediction of cooling curves at various locations within the casting as a function of casting geometry. The coupling of macrotransport equations with solidification kinetics was achieved through the latent heat method. The ripening of secondary dendrite arms was described by a dynamic coarsening model.
4.1 Theoretical Formulation of Macroscopic Heat Transfer

The main issue in continuum modelling of solidification process is to simultaneously solve the mass, energy and momentum transport equations. The standard transport equation for advection-diffusion is

\[
\frac{\partial}{\partial t} (\rho \phi) + \nabla (\rho \mathbf{V} \phi) = \nabla (\rho \gamma \nabla \phi) + S
\]

(4.1)

where \( t \) is time, \( \rho \) is density, \( \phi \) is the phase quantity, \( \mathbf{V} \) is the velocity vector, \( \gamma \) is the general diffusion coefficient and \( S \) is source term. However, as far as the casting solidification is concerned, the entire process is primarily controlled by diffusion of heat and, to a small extent, convection in the liquid. Therefore, neglecting the diffusion of species (i.e. constant density) and momentum transfer (\( \mathbf{V} = 0 \)) the transport equation can be written in terms of temperature rather than enthalpy as

\[
\frac{\partial T}{\partial t} = \alpha \nabla^2 T + \frac{\dot{Q}}{\rho c_p}
\]

(4.2)

where \( T \) is temperature, \( \alpha \) is the thermal diffusivity, \( c_p \) is the specific heat and \( \dot{Q} \) is the heat source term. This source term represents the latent heat released during solidification and is given by

\[
\dot{Q} = \Delta H_f \frac{\partial f_s}{\partial t}
\]

(4.3)

where \( \Delta H_f \) is the latent heat of fusion and \( f_s \) is the solid fraction. During the experimental studies, the composites were cast into cylindrical molds with insulation in the axial surfaces to ensure the heat transfer to occur in radial coordinates. In order to simulate the experimental conditions, composites were assumed to solidify in sand molds with cylindrical cavities and with zero heat flux in \( \theta \) and \( z \) directions. The definition of cylindrical coordinates is presented in Fig 4.1. The equation for transient heat conduction is given by
\[
\rho c_p \frac{\partial T(r, t)}{\partial t} = k \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T(r, t)}{\partial r} \right) + \Delta H_f \frac{\partial f_s(r, t)}{\partial t}
\]  

where, \( r \) is the radial coordinate and \( k \) is the thermal conductivity. In order to solve this equation, the evolution of solid, \( f_s(r, t) \) must be described as a function of time and location inside the casting. This can be achieved through various schemes for coupling of the macroscopic heat flow with the microscopic solidification kinetics\(^{50, 51}\). In the present study, the latent heat method (LHM)\(^{52}\) was used to incorporate the latent heat release during phase transformation to the macroscopic heat transfer. Heat transfer was assumed to be controlled by the resistance at the metal/mold interface and a prescribed flux boundary condition was used to account for the transfer at the interface\(^{48}\).

**Figure 4.1** Definition of \( \hat{r} \), \( \hat{\theta} \) and \( \hat{z} \) directions in cylindrical coordinates.
4.2 Microscopic Modelling

4.2.1 Pseudo-Binary Alloy Assumption: Calculation of Equivalent Solute

Formation of intermetallic phases at the later stages of eutectic solidification does not have any influence on the as-solidified particle distribution. Therefore, for the ease of calculations, it was assumed that solidification of the A356 matrix alloy results in two distinct microstructural regimes; the equiaxed $\alpha$-Al dendrites and the eutectic phase. The silicon equivalency ($Si_{eq}$) method\(^{53}\) was used to treat the A356 alloy as a pseudo-binary Al-$Si_{eq}$ alloy. The $Si_{eq}$ value expresses the amount of alloying elements other than Si (only 0.35% Mg in this case) in terms of an equivalent amount of silicon by the following expression

$$Si_{eq} = Si + \sum X_{eq}^{Si}$$  \hspace{1cm} (4.5)

where, $X$ denotes the alloying elements other than Si. The $\sum X_{eq}^{Si}$ values for some major and minor alloying elements and the resulting depression in the liquidus temperature are presented in Table 4.1.

**Table 4.1** $Si_{eq}$ (wt%) values and the resulting liquidus depression ($\Delta T_L$) of some alloying elements for 3XX aluminum alloys\(^{53}\).

<table>
<thead>
<tr>
<th>Alloying elements (1wt%)</th>
<th>Cu</th>
<th>Fe</th>
<th>Mg</th>
<th>Mn</th>
<th>Zn</th>
<th>Ni</th>
<th>Pb</th>
<th>Sn</th>
<th>Bi</th>
<th>Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Si_{eq}$</td>
<td>0.323</td>
<td>0.650</td>
<td>0.017</td>
<td>0.787</td>
<td>0.123</td>
<td>0.536</td>
<td>0.889</td>
<td>0.752</td>
<td>0.898</td>
<td>0.770</td>
</tr>
<tr>
<td>$\Delta T_L$ (°C)</td>
<td>1.98</td>
<td>4.00</td>
<td>0.10</td>
<td>4.84</td>
<td>0.75</td>
<td>5.47</td>
<td>4.63</td>
<td>5.53</td>
<td>4.74</td>
<td></td>
</tr>
</tbody>
</table>
4.2.2 Nucleation of α-Al Dendrites

It was assumed that solidification of the A356 matrix alloy results in two distinct microstructural regimes; the equiaxed α-Al dendrites and the eutectic phase. It was previously reported\(^2\) that the SiC particles are not suitable substrates for nucleation of α-Al dendrites. Therefore, we have assumed that the presence of SiC particles have no influence on nucleation kinetics of the α-Al dendrites.

The nucleation models used in the present study are mainly intended for equiaxed solidification. Bulk nucleation heterogeneously takes place within the melt on foreign particles already existing in the liquid. A simple empirical instantaneous nucleation model was used to account for the nucleation of equiaxed α-Al dendrites in order to avoid computational complications related to definition of the micro-volume element\(^{48}\) as will be explained in the following section. The model assumes that all nuclei are generated at the nucleation temperature (Fig 4.2a) and the nucleation site density is determined by the cooling rate at the onset of solidification. The governing equation is given by

\[
N_s = a + b \left( \frac{\partial T}{\partial t} \right)^2
\]

(4.6)

where, \(N_s\) is the volumetric nucleation site density and \(a\) and \(b\) are experimentally determined constants\(^{54}\).
4.2.3 Dendritic Growth

There is still no complete theoretical solution that can describe the complexity of dendrite growth\textsuperscript{48}. Although the diffuse interface approaches\textsuperscript{55} (also known as the phase field method) can handle the morphological evolution of a dendrite quite successfully, they have limited applicability in terms of engineering usefulness. Furthermore, focusing on morphological evolution of each dendrite in a three dimensional space would require excessive computational capabilities, which does not seem possible at the current state of the art. Therefore, for the problem under consideration in this study, we have employed a simplified volume averaged dendrite model\textsuperscript{48}.

The schematic representation of the volume average model for an equiaxed condensed dendrite is given in Fig. 4.3. The dendrite envelope defines a pseudo-interface that separates the intradendritic and extradendritic liquid phases. It embraces the solid phase and the intradendritic liquid. The equivalent dendrite envelope ($r_E$), on the other hand, defines a spherical volume identical to the dendrite envelope. This volume also includes the solid and the intradendritic liquid. For further ease of calculations, this volume can be assumed as approximately equal to the volume of solid phase. Therefore, the growth rate of the condensed dendrite can be described over an averaged volume of sphere. The growth proceeds until all micro-volume element is filled, which has a radius of $r_f$. 

![Figure 4.2 Schematic comparison of (a) instantaneous and (b) continuous nucleation models\textsuperscript{48}.](image)
Figure 4.3 Schematic representation of assumed morphology and the associated concentration profile of globular equiaxed dendrites as given by Nastac and Stefanescu\(^{56}\).

Figure 4.4 The concentration and temperature profiles ahead of the dendrite tip. \(\Delta T_L\) and \(\Delta T_t\) shows the solutal and thermal undercoolings, respectively. \(\langle C_L \rangle^*\) is the intrinsic volume average concentration\(^{56}\).
The growth rate of the equiaxed dendrites was calculated by the model developed by Nastac and Stefanescu\textsuperscript{56} which relies on the melt undercooling at the dendrite tip (Fig. 4.4), which is given by

$$\Delta T = \Delta T_c + \Delta T_t$$

(4.7)

where $\Delta T_c$ and $\Delta T_t$ are the solutal and thermal undercoolings, respectively. The governing equation to describe the mean growth velocity of the dendrite tip is given by Eq. (4.8). The original derivation is given by Nastac and Stefanescu\textsuperscript{56}.

$$V_s = \left[ 2\pi^2 \Gamma \left( \frac{m(\kappa-1)C_L^*}{D_L} + \frac{\Delta H_f}{c_p \alpha_L} \right) \right]^{-1} \cdot \Delta T^2$$

(4.8)

where, $m$ is the liquidus slope, $\kappa$ is the partition coefficient, $C_L^*$ is the liquid interface concentration, $D_L$ is the liquid diffusion coefficient, $\Gamma$ is the Gibbs-Thomson coefficient, $\alpha_L$ is the liquid thermal diffusivity and $\Delta T$ is the undercooling. The melt undercooling is calculated by a linear liquidus assumption;

$$\Delta T = T_m + m \langle C_L \rangle - T_{\text{bulk}}$$

(4.9)

where, $T_m$ is the melting point of pure aluminum, $\langle C_L \rangle$ is the volume average extradendritic liquid concentration and $T_{\text{bulk}}$ is the average temperature in the volume element (see Fig. 4.4).

### 4.2.4 Eutectic Nucleation

The eutectic microconstituent may form by heterogeneous nucleation on the primary phase or as independently nucleated equiaxed grains on nucleant particles in the interdendritic liquid\textsuperscript{57}. However, the literature reports contradictory results on whether the SiC particles could act as nucleants for heterogeneous nucleation of the eutectic phase.
Although SiC particles were previously reported\textsuperscript{58, 59} as suitable substrates for heterogeneous nucleation of the eutectic phase, Nagarajan \textit{et al}\textsuperscript{59} did not detect any significant alteration in the eutectic undercooling due to presence of SiC particles. For the ease of calculations, following the method of Gonzalez-Rivera \textit{et al}\textsuperscript{54}, we have assumed that the eutectic phase forms only as independently nucleated equiaxed grains. This assumption leads to prediction of higher eutectic growth rates than the actual case, which was compensated by a suitable impingement treatment, as will be explained in the following section.

A continuous nucleation model (Fig. 4.2b) was adopted to account for the nucleation of eutectic grains in order to obtain a more realistic impression of eutectic undercooling and recalescence periods on the simulated cooling curves. Oldfield’s empirical model\textsuperscript{60} was used to describe the nucleation site distribution. The governing equation is given by

\begin{equation}
\frac{\partial N_s}{\partial t} = -n\mu_N (\Delta T)^{n-1} \frac{\partial T}{\partial t} (1 - f_S)
\end{equation}

where, $\mu_N$ is a nucleation parameter, $n = 2$ and the term $(1 - f_S)$ was included to account for the residual volume fraction of liquid.

\subsection*{4.2.5 Eutectic Grain Growth and Impingement}

The growth of the equiaxed eutectic grains was calculated by using the Johnson-Mehl model\textsuperscript{61}

\begin{equation}
V_s = \frac{\partial R}{\partial t} = \mu_{\text{eut}} \Delta T_{\text{eut}}^2
\end{equation}

where, $R$ is the grain radius, $\mu_{\text{eut}}$ is the eutectic growth constant and $\Delta T_{\text{eut}}$ is the eutectic undercooling. Since the equiaxed grains have spherical morphology, the evolution of solid fraction can be described by\textsuperscript{48}
\[
\frac{\partial f_S}{\partial t} = 4\pi \left( \frac{\partial N_r}{\partial t} \frac{R^3}{3} + N_r r^2 \frac{\partial R}{\partial t} \right) (1 - f_S)^{\Psi/6}
\] (4.12)

The effect of grain impingement was considered by weighting the effective area between the solid grains and liquid by the term \((1 - f_S)^{\Psi/6}\) in Eq. (4.12) with \(\Psi = 3\). This correction is analogous to previous approximations by Johnson-Mehl\textsuperscript{61} and Avrami\textsuperscript{62}. \(\Psi\) is a constant introduced to account for the delay in the eutectic growth rate\textsuperscript{54} due to impingement of growing eutectic grains with each other, with the preexisting primary dendrites and also with the reinforcing SiC particles.

### 4.2.6 Coarsening of Secondary Dendrite Arms

The main aim of the numerical studies in this work is to estimate the secondary dendrite arm spacing distribution in a casting as a function of local cooling rate. Therefore, special emphasis will be given to this section and the applied model will be described in more detail.

The secondary arms are morphological instabilities or branches that grow perpendicular to the primary trunk. Although in early theories of dendritic solidification it was assumed that the secondary arms form in beginning of dendritic solidification and grow cooperatively with the primary trunk, it has later been recognized that the secondary arms coarsen with a mechanism analogous to Oswald ripening process in precipitate growth\textsuperscript{63}. During coarsening of secondary dendrite arms, smaller branches shrink and melt and remaining larger branches continue their growth. The main reason behind this phenomenon is the effect of curvature on the liquidus temperature and the concentration profiles along the surfaces of these instabilities.

As the dendrite arm gets smaller, the contribution of the surface energy to the free energy of the solid phase increases due to increasing surface energy to volume ratio. This increment in free energy results in a depression in the equilibrium melting temperature of the solid phase (Fig. 4.5). Therefore, the smaller dendrite arms with higher curvature and therefore lower melting point dissolve to the melt and eventually disappear, whereas; the
larger arms continue their growth. This phenomenon is known as the dynamic coarsening of secondary dendrite arms\cite{48}.

\textbf{Figure 4.5} Variation of free energy of the solid and liquid phases as a function of temperature. Additional surface due to curved interface results in a depression in the equilibrium melting temperature of the solid phase by $\Delta T$\cite{66}.

The coarsening of secondary arms is a diffusion controlled process. It is well established that the secondary dendrite arm spacing can be related to the solidification time by the empirical relationship

$$\lambda_2^3 = \mu_0 \delta_f$$

(4.13)

where, $\lambda_2$ is the secondary dendrite arm spacing, $t_f$ is the solidification time and $\mu_0$ is the coarsening constant. The correct prediction of as-solidified arm spacings depends on the derived expressions of $\mu_0$. Various derivations of $\mu_0$ were previously proposed in the literature\cite{63, 64, 65}. In the present study, the dynamic coarsening model developed by Mortensen\cite{65} was used. The governing derivation in the following pages is for array of coarsening cylinders, which represents the secondary dendrite arms, held at a constant holding temperature, $T_{\text{hold}}$, for a certain time, $t$. 

33
As the dendrites grow, the secondary arms also get thicker. The average diameter of the dendrite arms, $\Phi$, therefore can be represented as a function of fraction solid formed. The governing relationship is

$$\Phi = \lambda_2 \sqrt{f_s}$$  \hspace{1cm} (4.14)

where $f_s$ is the solid fraction. Since under isothermal conditions the solid fraction remains constant, the rate of thickening of the arms can be expressed by

$$\frac{d\Phi}{dt} = \frac{d\lambda_2}{dt} \sqrt{f_s}$$  \hspace{1cm} (4.15)

While other approximations cited above generally focus on dissolution time of small dendrite arms into the melt, the Mortensen’s model focuses on the fraction of dendrites that continue their growth. Some fraction of dendrites, $F$, is assumed to continue their growth, while others shrink. If the diameters of the growing and shrinking arms are denoted as $\Phi_g$ and $\Phi_s$, respectively; then the average dendrite arm diameter can be given by the following relationship

$$\Phi = F\Phi_g + (1 - F)\Phi_s$$  \hspace{1cm} (4.16)

Assuming $F \approx 0.5$ and $\Phi_g \approx 2\Phi_s$ and combining Eqns. (4.14) and (4.16); we have

$$0.75\lambda_2 \sqrt{f_s}$$  \hspace{1cm} (4.17)
Figure 4.6 Schematic illustration of the situation between two solid cylinders of different radii, placed in a locally isothermal melt.
Fig. 4.6 illustrates the situation between two solid cylinders of different radii placed in a locally isothermal melt. Due to effect of curvature, (see Fig. 4.5) the two cylinders will have different liquidus temperatures and hence, the concentration along the surfaces of these cylinders will also be different. The surface of the cylinder with smaller radius will be at a lower solute concentration. Therefore, the solute will diffuse along the concentration gradient from the larger to smaller cylinder, while the solvent atoms will diffuse from the smaller to larger cylinder. As a consequence, the smaller cylinder will dissolve to the melt, while the larger cylinder will continue its growth. If it is assumed that the spacing between these cylinders are sufficiently small and the coarsening rate is sufficiently low so that local equilibrium is established between them, then the solute flux from the larger to smaller cylinder and solvent flux from the smaller to larger cylinder are given by

\[ J_{\text{solute}} = -D_L \frac{\partial C}{\partial x} \]  
\[ J_{\text{solvent}} = -C_L' (1 - \kappa) \frac{1}{2} \frac{d\Phi_s}{dt} \]

where \( \kappa \) is the partition coefficient. Now, if we assume that the difference between surface concentrations is very small so that \( C_L' = C_L^R = \langle C_L \rangle \), where \( \langle C_L \rangle \) is the average liquid concentration, by combining Eqns. (4.18) and (4.19) and writing the flux of the solvent for the growing arms, we obtain the expression for the thickening rate of the larger arms;

\[ \frac{1}{2} \frac{d\Phi_s}{dt} = \frac{D_L}{(1 - \kappa) \langle C_L \rangle} \frac{\partial C}{\partial x} \]

The rate at which the larger sphere coarsens is therefore a function of the concentration gradient; that is the term \( \partial C/\partial x \) in Eqn (4.20), which in turn depends on the amount of depression in liquidus temperatures (see Fig. 4.5). The liquidus temperatures at the surfaces of the cylinders are given by Kurz and Fisher\(^63\) as

\[ T_L' = T_m + mC_L' - \frac{2\Gamma}{\Phi_s} \]
\[ T_L^R = T_m + mC_L^R \frac{2\Gamma}{\Phi_g} \]  \hspace{1cm} (4.22)

where \( T_m \) is the melting point of the solvent, \( m \) is the liquidus slope and \( \Gamma \) is the Gibbs-Thomson coefficient. Under isothermal conditions, combining Eqns. (4.21) and (4.22)

\[ m(C_L^R - C_L^D) = \Gamma \left( \frac{2}{\Phi_s} - \frac{2}{\Phi_g} \right) \]  \hspace{1cm} (4.23)

Therefore the concentration difference is

\[ \Delta C = -\frac{2\Gamma}{m\Phi_g} \]  \hspace{1cm} (4.24)

Combining Eqns. (4.17) and (4.24), the difference in concentration can be written as

\[ \Delta C = \frac{-3\Gamma}{2m\sqrt{f_s}\lambda_2} \]  \hspace{1cm} (4.25)

The average diffusion distance, \( \Delta l \), in Fig. 4.6 is given by Mortensen as

\[ \Delta l = (1 - \sqrt{f_s})\lambda_2 \]  \hspace{1cm} (4.26)

Then, it can be estimated that

\[ \frac{-\Delta C}{\Delta l} \approx -\frac{\partial C}{\partial x} = \frac{3\Gamma}{2m\lambda_2^2 \left(1 - \sqrt{f_s}\right)\sqrt{f_s}} \]  \hspace{1cm} (4.27)

Combining Eqns. (4.17), (4.20) and (4.27) we obtain

\[ \frac{\lambda_2^2}{dt} \frac{d\lambda}{dt} = \frac{9\Gamma D_L}{4m(1 - \kappa)C_L \left(1 - \sqrt{f_s}\right)\sqrt{f_s}} \]  \hspace{1cm} (4.28)
Integrating Eqn. (4.28) with in the interval of isothermal holding time, }{t_h},

\[
\lambda_{2,f}^3 - \lambda_{2,0}^3 = \frac{27 \Gamma D f t_b}{4m(1-\kappa)\left(C_L\right)\left(1-\sqrt{f_s}\right)f_s} = \mu_0 t_b
\]  

(4.29)

where \( \lambda_{2,0}^3 \) and \( \lambda_{2,f}^3 \) are the spacings between secondary arms at the beginning and end of the isothermal holding. Assuming that \( \lambda_{2,0} < \lambda_{2,f} = \lambda_2 \), the final secondary arm spacing is given by

\[
\lambda_2^3 = \mu_0 f_{sf} \quad \text{with} \quad \mu_0 = \frac{27 \Gamma D f_l}{4m(1-\kappa)\left(C_L\right)\left(1-\sqrt{f_s}\right)f_s}
\]  

(4.30)

Eqn. (4.30) describes the coarsening of secondary arms under isothermal conditions. In order to integrate this equation to the actual process of solidification, one needs to know the functional dependence of the average liquid concentration and solid fraction with time\(^6\). The dependence of these variables to time was directly calculated from their corresponding alterations within the micro-volume element, which was presented in section 4.2.3, and implemented to Eqn. (4.30) to achieve the as solidified secondary dendrite arm spacing distribution within the casting.
CHAPTER 5

EXPERIMENTAL & COMPUTATIONAL DETAILS

5.1 Materials

5.1.1 Matrix Alloy

The matrix alloy chosen for the present study was A356 aluminum alloy due to its excellent castability and rather wide solidification range (see Appendix A). The composition limits of the A356 alloy are presented in Table 5.1. Typical applications are aircraft structures, machine parts, truck chassis parts and other structural applications requiring high strength.

Table 5.1 Composition limits of A356 aluminum alloy

<table>
<thead>
<tr>
<th></th>
<th>Si</th>
<th>Mg</th>
<th>Cu</th>
<th>Mn</th>
<th>Fe</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>6.5% - 7.5%</td>
<td>0.25% - 0.45%</td>
<td>0.20% (max)</td>
<td>0.10% (max)</td>
<td>0.20% (max)</td>
</tr>
<tr>
<td>Zn</td>
<td>0.10% (max)</td>
<td>0.20% (max)</td>
<td>0.05% (max)</td>
<td>0.15% (max)</td>
<td>Balance</td>
</tr>
<tr>
<td>Ti</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Other (each)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Others (total)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

5.1.2 The Reinforcement Phase

F320 type, green silicon carbide (SiC) particles with an average particle size of 29.2 ± 1.5 μm were used as the reinforcement phase. Table 5.2 presents the chemical and physical properties of the SiC particles. The surface chemical values of the particles are given in Table 5.3.
Table 5.2 Chemical and physical properties of the SiC particles

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystal form:</td>
<td>$\alpha$-SiC hexagonal</td>
</tr>
<tr>
<td>True density (kg m$^{-3}$):</td>
<td>3.20</td>
</tr>
<tr>
<td>Color:</td>
<td>Green</td>
</tr>
<tr>
<td>Decomposition point (°C):</td>
<td>2300</td>
</tr>
<tr>
<td>Hardness - Knoop (kg cm$^{-2}$):</td>
<td>2500 - 2900</td>
</tr>
<tr>
<td>Reaction with acids:</td>
<td>Very slight surface action with hydrofluoric acid</td>
</tr>
<tr>
<td>Oxidation-reduction:</td>
<td>Oxidation slowly starts at 800 °C. No reduction.</td>
</tr>
</tbody>
</table>

Table 5.3 Surface chemical values of F320 silicon carbide

<table>
<thead>
<tr>
<th>Product</th>
<th>%SiC</th>
<th>%Free C</th>
<th>%Si</th>
<th>%SiO$_2$</th>
<th>%Fe$_2$O$_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>F240 - F800</td>
<td>99.50</td>
<td>0.10</td>
<td>0.10</td>
<td>0.10</td>
<td>0.05</td>
</tr>
</tbody>
</table>

Figure 5.1 Morphology of SiC particles used in this work.
Fig. 5.1 shows the morphology of the SiC particles used in this study. The X-ray diffraction pattern (Cu Kα) of reinforcement particles is shown in Fig. 5.2 with JCPDS (Joint Committee on Powder Diffraction Standards) data 29-1131 (6H-SiC).

**Figure 5.2** (a) X-ray diffraction pattern of the SiC particles and (b) standard powder pattern of 6H-SiC (JCPDS 29-1131).
5.2 Stir Casting of Aluminum Matrix Composites

The aluminum alloy matrix composites were synthesized by the double stir-casting method. A weighed quantity of high purity aluminum electrical wires were melted in a clay bonded graphite crucible and alloyed with required amount of silicon. After complete dissolution of silicon, the melt was degassed with high purity argon (99.998%) for 20 min with a flow rate of 5 liter per min. In order to improve the wettability of SiC particles, the melt was alloyed with 1% Mg and temperature was dropped below liquidus, to the semi-solid state. SiC particles, preheated at 300 °C for 2 hours were added to the slurry and manually stirred until the particles were completely wetted. The composite slurry was then reheated to $T_L \pm 5 ^\circ C$, where $T_L$ is the liquidus temperature, and stirred with a stainless steel four-blade impeller at 300 rpm for 15 minutes. The blades of the impeller were coated with a zirconia based suspension to avoid iron contamination of the melt. In order to compensate the oxidized magnesium during the stirring period, the melt was brought to fully liquid state and alloyed with required amount of magnesium to reach the nominal composition. Extreme care was taken for temperature control of the melt (720 ± 10 °C) during processing to avoid Al$_4$C$_3$ formation.

![Schematic diagram of the experimental setup for synthesis of aluminum matrix composites](image1)

![Schematic of a typical cylindrical thermal analysis cup](image2)

Figure 5.3 (a) Schematic diagram of the experimental setup for synthesis of aluminum matrix composites; (b) schematic of a typical cylindrical thermal analysis cup.
During production of samples, the amount of charge materials, stirring duration and position of stirrer in the crucible were almost kept constant to minimize the contribution of variables related to stirring on distribution of reinforcement particles. Fig 5.4 illustrates the position of the stirrer in the ladle and the schematic of the four-blade stirrer. In all experiments, the total amount of charge materials ranged between 1800 to 2400 gr, which resulted in a liquid metal height of 6.5 to 8 cm. Nagata\textsuperscript{68} stated that, in order to avoid accumulation of particles at the bottom of the ladle, the position of the stirrer should not exceed 30\% of the height of the liquid metal from the base. In all experiments, care was taken to position the stirrer 1.5 to 2 cm from the base to ensure uniform dispersion of particles within the liquid.

**Figure 5.4** (a) Position of stirrer in the ladle; (b) schematic of the four-blade stirrer.
5.3 Thermal Analysis of MMC Solidification

The composites were poured into cylindrical resin-coated shell sand molds (see Fig. 5.3b) with varying dimensions (see Appendix B) to obtain different solidification rates and allowed to cool down to room temperature. The investigated range of solidification rates were between 0.164 and 2.417 °C sec\(^{-1}\).

The temperature of the solidifying composites was monitored from three alumina sheathed K-type thermocouples placed inside each cylindrical mold. In each mold, one thermocouple was positioned at the geometrical center of the mold and the others at two different radial distances from the center and at the same vertical height as the center thermocouple (Fig. 5.3b). Apart from its suitability for both Fourier and Newtonian thermal analysis procedures, this set-up is also typical for estimation of dendrite coherency point (DCP) with two thermocouples, which is based on the temperature difference (\(\Delta T\)) between the wall and center thermocouples\(^{41}\).

The thermocouple tips were in direct contact with the liquid. The top and bottom surfaces of the cylindrical molds were isolated with layers of zirconia and kaolin wool placed on preheated ceramic plates to ensure the cooling of the samples only by heat transfer from the radial surfaces for estimation of dendrite coherency points and also to establish similar heat transfer conditions with the 2D computational algorithm presented in Chapter 4. The thermal data were obtained by recording the time and temperature values by using an Elimko multi-channel data acquisition system at a rate of 1 Hz. The recorded data were processed with a program developed in Mathcad environment (Mathsoft Eng. & Ed. Inc. v11), which allowed application of Newtonian (NTA) and Fourier thermal analysis (FTA) procedures, calculation of solid fraction (\(f_S\)) curves and estimation of dendrite coherency points (DCP). Solidification rate of the samples were calculated from the slopes of the cooling curves between liquidus and eutectic reactions, as received from the center thermocouples. The solidification rates were defined to be positive so that the calculated values indicate their magnitude.
The DTA measurements were carried out by the Setaram SETSYS DTA device already available in the Department of Metallurgical and Materials Engineering in METU. The investigated temperature range was from 660 to 480 °C, at a cooling rate of 10 °C min\(^{-1}\) under argon atmosphere.

5.4 Image Analysis

The solidified samples were sectioned and prepared for metallographic analysis. The Cartesian coordinates of the centroids of the SiC particles and the secondary dendrite arm spacing (SDAS) values were obtained by Clemex Image Analysis system. At least 20 random fields of each sample were analyzed to achieve the spatial and metallographic data and care was taken to avoid overlapping of the analyzed fields. The metallographic fields were 1376.3 × 1017.2 μm\(^2\) in all cases (Fig. 5.5).

![Figure 5.5](image)

**Figure 5.5** (a) An optical micrograph of sample no A2010; (b) selected SiC particles for quantitative analysis and; (c) centroids of SiC particles, presented with the Voronoi diagram.
In order to capture the long range spatial patterns and to analyze the effect of metallographic study area on the resulting statistics, the calculations were also performed on digital montages of contiguous microstructural fields. Fig. 5.6 shows such a montaged microstructure of an A356-20% SiC composite and a close-up image showing the high resolution of the image. The photographs of contiguous metallographic fields were collected and montaged with Adobe Photoshop CS2 image editor software. This procedure is necessary to capture relatively large microstructural fields that cannot be captured by an optical microscope with high resolution. The high resolution of the image is particularly important for the image analysis program to capture the microstructural details.

Figure 5.6 (a) A high resolution montage of contiguous fields from an A356-20% SiC composite microstructure; (b) a close-up image from the metallographic field, together with the binary image.
5.5 Computational Details

5.5.1 Quantitative Analysis of Particle Distribution

5.5.1.1 Computer generated point data

Computer simulated artificial distributions of events, where each event represents a reinforcement particle, were generated in order to test the response of applied statistical methods to various point patterns and levels of clustering. Three types of point processes; random (Poisson), regular and clustered were considered. Random event sets were generated by simulation of homogeneous Poisson process, in which the \(x\) and \(y\) coordinates were sampled from two independent uniform distributions within the boundaries of the study region, \(R\). Inhibition model was used for regular event sets, which commences with generation of a Poisson distribution, followed by elimination of events that are found to be less than some specified distance, \(d\), from their nearest-neighbour. The clustered event sets were generated by a three step Neyman-Scott cluster process. First, parent events were distributed according to a Poisson process with a pre-defined intensity. Then a number of events, obtained from a random distribution with mean \(\mu\) were assigned to each parent. Finally, the assigned events were distributed around the parents according to a Gaussian probability distribution function centered on the parents and with standard deviation \(\sigma\). In order to monitor the response of the applied functions to local variations in particle content, we have generated regular, random and clustered data sets with three different intensities. Each representative distribution was limited to a \(1000 \times 1000\) square area and consisted of 150, 300 and 450 events. The clustered data sets were generated with varying \(\mu\) and \(\sigma\) to test the robustness of the applied functions to alterations in clustering levels. Nine different clustered distributions were generated with the following properties:

- For 150 events: \(\mu = 10; \sigma = 50, 100, 150\)
- For 300 events: \(\mu = 20; \sigma = 50, 100, 150\)
- For 450 events: \(\mu = 30; \sigma = 50, 100, 150\)
5.5.1.2 Spatial analysis programs

Four different analysis methods were used in the present study; Voronoi tessellation, refined nearest neighbour analysis, K-function and the local density statistics. The Voronoi polygons for the set of coplanar points, which represent the centroids of SiC particles, were first determined with MATLAB (The Mathworks, Inc. v7). Area distribution of the resulting cells was evaluated by Clemex image analysis software. The polygon areas could also be determined by the inbuilt function ‘polyarea’ in MATLAB. However, since the ‘polyarea’ function also takes the marginal polygons into consideration and makes no edge correction; the analysis ends up with incorrect results. In order to overcome this problem, the marginal polygons were eliminated and the area distributions were evaluated via the image analysis software. A set of programs developed in-house by the author were used for the refined nearest neighbour analysis, K-function and the local density statistics. All programs were developed in Mathcad environment (Mathsoft Eng. & Ed. Inc. v11).

5.5.2 Macrotransport – Solidification Kinetics Modelling

A program developed by the author was used for numerical assessment of composite solidification. The program calculates the evolution of fraction of solid during the course of solidification and couples the calculated evolution with the governing macrotransport equations. The coupling was achieved through the latent heat method (LHM). The flowchart of the coupling procedure is given in Fig. 5.7. The numerical parameters used in simulations and thermal analysis are presented in Table 5.3.
Figure 5.7 Basic flowchart of macrotransport – solidification kinetics coupling algorithm by latent heat method (LHM)\textsuperscript{52}.
Table 5.3 Selected data used in simulations and in thermal analysis of composites.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Mold (shell sand)</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Density</td>
<td>$\rho_m = 1600$</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>$k_m = 0.52$</td>
<td>W m$^{-1}$ °C$^{-1}$</td>
</tr>
<tr>
<td>Specific heat</td>
<td>$c_{p,m} = 1170$</td>
<td>J kg$^{-1}$ °C$^{-1}$</td>
</tr>
<tr>
<td><strong>Matrix(A356) - Reinforcement (SiC)</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Solid density</td>
<td>$\rho_S = 2700$</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>Liquid density</td>
<td>$\rho_L = 2400$</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>Particle density</td>
<td>$\rho_P = 3200$</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>Solid specific heat</td>
<td>$c_{p,S} = 1084$</td>
<td>J kg$^{-1}$ °C$^{-1}$</td>
</tr>
<tr>
<td>Liquid specific heat</td>
<td>$c_{p,L} = 963$</td>
<td>J kg$^{-1}$ °C$^{-1}$</td>
</tr>
<tr>
<td>Particle specific heat</td>
<td>$c_{p,P} = 1300$</td>
<td>J kg$^{-1}$ °C$^{-1}$</td>
</tr>
<tr>
<td>Solid thermal conductivity</td>
<td>$k_S = 159$</td>
<td>W m$^{-1}$ K$^{-1}$</td>
</tr>
<tr>
<td>Liquid thermal conductivity</td>
<td>$k_L = 121$</td>
<td>W m$^{-1}$ K$^{-1}$</td>
</tr>
<tr>
<td>Particle thermal conductivity</td>
<td>$k_P = 250$</td>
<td>W m$^{-1}$ K$^{-1}$</td>
</tr>
<tr>
<td>Latent heat (Pure Al)</td>
<td>$\Delta H_f^p = 3.91 \times 10^5$</td>
<td>J kg$^{-1}$</td>
</tr>
<tr>
<td>Latent heat (Eutectic)</td>
<td>$\Delta H_f^{eut} = 5.63 \times 10^5$</td>
<td>J kg$^{-1}$</td>
</tr>
<tr>
<td>Partition coefficient</td>
<td>$\kappa = 0.132$</td>
<td></td>
</tr>
<tr>
<td>Liquidus slope</td>
<td>$m = -6.656$</td>
<td>°C wt%$^{-1}$</td>
</tr>
<tr>
<td>Liquid diffusion coefficient</td>
<td>$D_L = 3 \times 10^{-9}$</td>
<td>m$^2$ s$^{-1}$</td>
</tr>
<tr>
<td>Gibbs-Thomson coefficient</td>
<td>$\Gamma = 0.9 \times 10^{-7}$</td>
<td>m °C$^{-1}$</td>
</tr>
<tr>
<td><strong>Nucleation and Growth</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Primary nucleation parameter</td>
<td>$a = 3.57 \times 10^9$</td>
<td>m$^3$</td>
</tr>
<tr>
<td>Primary nucleation parameter</td>
<td>$b = 2.3 \times 10^7$</td>
<td>s$^2$ m$^{-3}$ °C$^{-2}$</td>
</tr>
<tr>
<td>Eutectic nucleation parameter</td>
<td>$\mu_N = 1 \times 10^6$</td>
<td>m$^2$ °C$^{-2}$</td>
</tr>
<tr>
<td>Eutectic growth constant</td>
<td>$\mu_{eut} = 5 \times 10^{-6}$</td>
<td>m s$^{-1}$ °C$^{-2}$</td>
</tr>
</tbody>
</table>
CHAPTER 6

RESULTS & DISCUSSION

The present work aims to study the clustering of reinforcement particles in metal matrix composites through experimental studies and numerical simulations. The scope of the study consists of (i) understanding the effect of solidification rate on clustering of reinforcement particles; (ii) studying suitable statistical descriptions of composite microstructures that can successively quantify the amount of particle clustering and (iii) establishing a model that predicts the cluster formation tendency in complex geometry cast parts.

Prediction of clustering is an important issue for two reasons. Firstly, being able to predict the cluster formation tendency in complex geometry cast parts before production would help the engineers to come up with new designs to produce these parts with a more uniform microstructure and therefore less prone to failure. The second benefit would be for the modelling studies aimed at predicting the mechanical response of complex composite parts with varying section thicknesses produced by solidification processing. Until now, such studies could only introduce the effect of particle clustering by some scalar descriptions. However, being able to predict and describe the particle distribution as a function of solidification rate would offer the possibility to consider the effect of local microstructural features as well.

The first section to be presented in this chapter concerns the thermal analysis results. The aim of the thermal analysis studies is to describe the evolution of solid fraction during the course of solidification and estimate the dendrite coherency points. The former also holds significance in verifying the numerical model, which describes the solidification of composites. The results regarding the quantitative analysis of particle
distribution will be presented in the following section, which will be followed by the
effect of solidification rate on as-solidified particle distribution and the results of the
numerical analysis study. As described previously, the aim of the numerical analysis study
is to establish a model that can predict the solidification microstructures, which
determines the as-cast particle distribution.

6.1 Thermal Analysis of Composite Solidification

Distribution of SiC particles was evaluated in terms of two parameters related to primary
solidification of composites; secondary dendrite arm spacing (SDAS) and volume fraction
of solid phase at the dendrite coherency ($f_{S}^{DCP}$). Secondary dendrite arm spacing is a
function of local cooling rate and is known to decrease as the cooling rate increases\(^47\). The
volume fraction of the solid phase at the dendrite coherency, on the other hand, depends
on the evolution of microstructure during the course of solidification. Refinement of the
dendritic structure postpones the coherency whereas; increased growth rate results in an
earlier formation of the dendritic skeleton\(^44, 69-72\). From a macro-scale point of view, one
could expect earlier occurrence of coherency with an increase in the magnitude of cooling
rate as a result of increased volumetric transformation rate. However, an increase in the
magnitude of cooling rate also results in refinement of the dendritic structure due to
increased undercooling. Therefore, the solid fraction at which coherency occurs is actually
determined by the competition between these two processes with counteracting effects\(^44, 72\).

In order to obtain the variation of solid fraction during the course of solidification,
the thermal data obtained from various locations within the cylindrical molds were
processed according to Newtonian (NTA) and Fourier (FTA) thermal analysis methods.
Fig. 6.1 presents sample cooling curves, their derivatives, the zero curves and the solid
fraction curves on the same time scale. The range of solidification rates investigated in the
present study is tabulated in Table 6.1.
Figure 6.1 Sample cooling curves, derivative curves, zero curves and solid fraction curves obtained by (a) Newtonian and (b) Fourier thermal analysis methods for A356 alloy composites reinforced with 20% SiC particles.
### Table 6.1 Range of investigated solidification rates.

<table>
<thead>
<tr>
<th>SiC Content</th>
<th>Sample</th>
<th>SR (°C sec(^{-1}))</th>
<th>SiC Content</th>
<th>Sample</th>
<th>SR (°C sec(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A1001</td>
<td>0.164</td>
<td></td>
<td>A2001</td>
<td>0.213</td>
</tr>
<tr>
<td></td>
<td>A1002</td>
<td>0.242</td>
<td></td>
<td>A2002</td>
<td>0.309</td>
</tr>
<tr>
<td></td>
<td>A1003</td>
<td>0.280</td>
<td></td>
<td>A2003</td>
<td>0.353</td>
</tr>
<tr>
<td></td>
<td>A1004</td>
<td>0.345</td>
<td></td>
<td>A2004</td>
<td>0.429</td>
</tr>
<tr>
<td></td>
<td>A1005</td>
<td>0.411</td>
<td></td>
<td>A2005</td>
<td>0.566</td>
</tr>
<tr>
<td></td>
<td>A1006</td>
<td>0.429</td>
<td></td>
<td>A2006</td>
<td>0.652</td>
</tr>
<tr>
<td></td>
<td>A1007</td>
<td>0.596</td>
<td></td>
<td>A2007</td>
<td>0.698</td>
</tr>
<tr>
<td>10%</td>
<td>A1008</td>
<td>0.682</td>
<td></td>
<td>A2008</td>
<td>0.775</td>
</tr>
<tr>
<td></td>
<td>A1009</td>
<td>0.725</td>
<td></td>
<td>A2009</td>
<td>0.806</td>
</tr>
<tr>
<td></td>
<td>A1010</td>
<td>0.795</td>
<td></td>
<td>A2010</td>
<td>1.107</td>
</tr>
<tr>
<td></td>
<td>A1011</td>
<td>0.981</td>
<td></td>
<td>A2011</td>
<td>1.211</td>
</tr>
<tr>
<td></td>
<td>A1012</td>
<td>1.121</td>
<td></td>
<td>A2012</td>
<td>1.421</td>
</tr>
<tr>
<td></td>
<td>A1013</td>
<td>1.329</td>
<td></td>
<td>A2013</td>
<td>1.861</td>
</tr>
<tr>
<td></td>
<td>A1014</td>
<td>1.928</td>
<td></td>
<td>A2014</td>
<td>2.417</td>
</tr>
<tr>
<td></td>
<td>A1015</td>
<td>2.319</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### 6.1.1 Evolution of Solid Fraction

The difference between the FTA and NTA lies in the way they generate the baselines, also known as the zero curves. Instead of NTA, which arbitrarily calculates the zero curve by some extrapolation or curve fitting techniques, in FTA the zero curve is obtained from the temperature Laplacian, \( \nabla^2 T \). The extent of this difference can be seen on the generated zero curves in Fig. 6.1. The zero curve generated by the NTA exhibits a smooth trend during the course of solidification, since it is obtained by fitting of an exponential function. The zero curve generated by FTA, on the other hand, is a function of the cooling rate and temperature Laplacian and therefore exhibits similar trends with the derivative curve.
Figure 6.2 Comparison of solidification rates obtained from NTA and FTA methods.

The differences between the solid fraction ($f_S$) estimations of NTA and FTA can better be understood by close examination of solidification rates in Fig. 6.2. NTA calculates a higher solidification rate at the onset of solidification compared to FTA. Due to this difference within the interval of primary solidification of $\alpha$-Al dendrites, the solid fraction values at the dendrite coherency point obtained from NTA and FTA also differs. The second peaks in Fig. 6.2 account for the eutectic solidification. FTA demonstrates another third peak corresponding to the solidification of intermetallic phases such as Mg$_2$Si and $\beta$-Al$_5$FeSi. The solidification of these phases cannot be monitored from the curve obtained from NTA. Formation of these phases was verified by DTA (Fig. 6.3) and also from the literature$^{73,74}$. 

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55
6.1.2 Comparison of Dendrite Coherency Point Estimations

As far as the as-cast particle distribution is concerned, formation of these intermetallics holds no importance. The main difference that needs to be underlined is the calculated solid fraction evolutions during dendritic solidification, since the distribution of reinforcement particles is determined during this interval. Therefore, it should again be noted that, NTA calculates a higher solidification rate at the onset of solidification compared to FTA. Consequently, the solid fraction values at the dendrite coherency point obtained from NTA generally, though not necessarily, corresponds to a higher solid fraction compared to FTA.
Figure 6.4 A typical output data: cooling curves obtained from center and wall thermocouples, the $\Delta T$ curve and the solid fraction curve for an A356 – 20% SiC$_p$ composite, presented on the same time scale.

Figure 6.5 Calculated solid fractions at the dendrite coherency point by NTA and FTA.
Fig. 6.4 illustrates a typical $f_s^{DCP}$ estimation procedure with two thermocouples. The solid fraction that corresponds to the first negative peak on the $\Delta T$ curve is accepted as the dendrite coherency point. Fig. 6.5 shows the comparison of $f_s^{DCP}$ values by NTA and FTA. Although no clear relationships could be observed, the solid fractions calculated by NTA were found to correspond to higher values compared to FTA. This difference was also reported by Barlow and Stefanescu\(^{40}\). The shaded region in Fig. 6.5 is added to emphasize this tendency.

6.2 Quantitative Analysis of Particle Distribution

The spatial data obtained from composite microstructures were analyzed with various methods in spatial statistics to quantify the amount of particle clustering. The discussed methods are the refined nearest neighbour statistics, Voronoi tessellation, $K$-function and the local density statistics method. Series of point processes with known properties were simulated in order to explore the reaction of the aforementioned methods to various levels of clustering. The applicability of these methods to quantify composite microstructures was discussed.

6.2.1 Simulated Point Data

Computer simulated artificial distributions of events, where each event represents a reinforcement particle, were generated in order to test the response of applied statistical methods to various point patterns and levels of clustering. Three types of point processes; random (Poisson), inhibition (uniform) and clustered were considered. Summary statistics of the simulated events are presented in Table 6.2. The tabulated results are calculated from 200 simulations of each point process with 150, 300 and 450 events. Fig. 6.6 demonstrates some examples of the simulated point processes.
Figure 6.6 Examples of simulated point processes with 300 events: (a) Neyman-Scott (NS) cluster process with $\sigma = 50$; (b) NS with $\sigma = 100$; (c) NS with $\sigma = 150$; (d) Poisson (random); (e) Inhibition (uniform) $d = 25$. All point processes were simulated on a 1000 × 1000 square.
Table 6.2 Summary statistics of the simulated point processes

<table>
<thead>
<tr>
<th>Events</th>
<th>Process</th>
<th>Q-ratio</th>
<th>% Threshold</th>
<th>CV_A</th>
</tr>
</thead>
<tbody>
<tr>
<td>150</td>
<td>NS (μ = 10; σ = 50)</td>
<td>0.614 ± 0.025</td>
<td>34.911 ± 9.032</td>
<td>1.457 ± 0.288</td>
</tr>
<tr>
<td></td>
<td>NS (μ = 10; σ = 100)</td>
<td>0.770 ± 0.060</td>
<td>29.001 ± 6.672</td>
<td>0.966 ± 0.194</td>
</tr>
<tr>
<td></td>
<td>NS (μ = 10; σ = 150)</td>
<td>0.818 ± 0.047</td>
<td>21.200 ± 4.709</td>
<td>0.758 ± 0.082</td>
</tr>
<tr>
<td></td>
<td>Poisson</td>
<td>0.988 ± 0.044</td>
<td>5.371 ± 1.194</td>
<td>0.548 ± 0.020</td>
</tr>
<tr>
<td></td>
<td>Inhibition (d = 25)</td>
<td>1.127 ± 0.045</td>
<td>0.396 ± 0.137</td>
<td>0.533 ± 0.017</td>
</tr>
</tbody>
</table>

| 300    | NS (μ = 20; σ = 50) | 0.657 ± 0.026 | 38.010 ± 10.706 | 1.517 ± 0.218 |
|        | NS (μ = 20; σ = 100) | 0.797 ± 0.022 | 29.199 ± 11.403 | 1.017 ± 0.208 |
|        | NS (μ = 20; σ = 150) | 0.840 ± 0.020 | 22.479 ± 5.766  | 0.932 ± 0.081 |
|        | Poisson          | 1.025 ± 0.024 | 4.802 ± 1.129   | 0.611 ± 0.043 |
|        | Inhibition (d = 25) | 1.285 ± 0.014 | 0.275 ± 0.088   | 0.410 ± 0.031 |

| 450    | NS (μ = 30; σ = 50) | 0.669 ± 0.020 | 40.567 ± 11.592 | 1.856 ± 0.248 |
|        | NS (μ = 30; σ = 100) | 0.793 ± 0.034 | 28.926 ± 6.669  | 1.275 ± 0.245 |
|        | NS (μ = 30; σ = 150) | 0.859 ± 0.018 | 18.378 ± 3.933  | 0.923 ± 0.124 |
|        | Poisson          | 0.992 ± 0.016 | 4.962 ± 1.103   | 0.597 ± 0.032 |
|        | Inhibition (d = 25) | 1.334 ± 0.014 | 0.134 ± 0.098   | 0.338 ± 0.022 |

Values are presented as mean ± standard deviation. NS refers to Neyman – Scott clustering process.

The Q-ratios derived from the nearest neighbour statistics were observed to be robust discriminators of varying clustering levels and different point processes with various intensities. However, it should be noted that, the clustered patterns were generated by Poisson (Neyman - Scott) clustering process and different clustering levels correspond to alterations in the standard deviation of the Gaussian probability function that specifies the distance of clustered events from the parent event. Therefore, the mechanism of clustering in the synthetic data is closely related to the nearest neighbour distances, which may be reason for the success of nearest neighbour analysis in the present case. The graphical representations of the refined nearest neighbour analysis are demonstrated in Fig. 6.7.
Figure 6.7 (a) The cumulative density function and (b) $\hat{G}(w) - G(w)$ plots of random (CSR), clustered (Neyman-Scott) and uniform (inhibition) point processes. Confidence intervals plotted at $\alpha = 0.01$ level, based on 200 replicates.
The generated point patterns were also analyzed by the local density statistics and the Voronoi tessellation method. Threshold values in Table 6.2 were calculated from the ratio of average number of clustered events to the total number of events in each particular study region (Eqn. 2.7). The dispersions of the probability density functions for area ($CV_a$) distributions were also found to be sensitive to the different point patterns and to alternating cluster levels, which may again be due to same reason as discussed for the nearest neighbour method.

Figure 6.8 Sample Voronoi plots derived from generated point data: (a) Neyman Scott cluster process with $\sigma = 50$, (b) Neyman Scott cluster process with $\sigma = 100$, (c) Poisson (random) and (d) uniform distribution. Marginal polygons are shown in dark color and excluded from analysis since they are not statistically significant.
One drawback of the Voronoi tessellation method is the lack of information regarding the theoretical distribution functions for either Poisson or other known spatial processes. Consequently, the distinction between clustered and uniform distributions cannot be brought out by comparing the observed values with some theoretical statistics. Therefore, the results can only be evaluated in terms of the alterations in $CV_a$ values. A lower $CV_a$ value indicates uniform distribution, whereas higher values imply a tendency of cells to aggregate into clusters. Sample Voronoi plots derived from point data are presented in Fig. 6.8.

Fig. 6.9 presents the $K$-function plots of the generated data for clustered, random and uniform distributions. The advantage of $K$-function over the other methods discussed so far is that, it can characterize the pattern over many length scales. For example, the nearest neighbour statistics cannot identify the distinction between different patterns beyond a length scale of approximately 60 units (see Fig. 6.7). $K$-function, on the other hand, was observed to identify any deviation from randomness all throughout the analyzed length scales.

![Figure 6.9](image)

**Figure 6.9** $K$-function plots presented with the $\hat{L}(r)$ correction for clustered, random and uniform distributions. Confidence intervals at $\alpha = 0.01$ level, based on 200 replicates.
The % Threshold values presented in Table 6.2 were obtained from the ratio of above-threshold events to all events within considered study regions averaged across all scales of observation. The decision whether an event belongs to a cluster or not depends on the number of events surrounding that particular event at some specific radius $r$. If the number of events surrounding an event exceeds the threshold $T\text{h}_{0.95}(r)$ at some distance $r$, that event was inferred to be a part of a cluster for that scale of observation. Therefore, the global clustering set $S(r)$ was updated at each distance scale, meaning that the number of above-threshold events varies at each scale of observation. Fig. 6.10a presents this variation in percentages as a function of distance $r$ for a generated clustered data set composed of 300 events with $\mu = 20$ and $\sigma = 100$ (see Fig 6.6b).

Three different distance scales were considered (80, 200 and 360) to monitor the effect of scale parameter $r$ on localization of detected clusters. The kernels associated to the global cluster sets $S(r)$ for each indicated distance scale are presented in Fig. 6.10b. Increasing the scale parameter from $r = 80$ to $r = 200$ expands the clusters, meaning that extra events were considered to be a part of a cluster. Further increasing the scale parameter from $r = 200$ to $r = 360$ results in rejection of some events from the global clustering set (the decrease in Fig. 6.10a). However, the kernels emerging from individual clusters merge into a single large cluster due to their interaction as a result of relatively large scale of observation as compared to the distance between the cluster centers. Note that while varying the scale parameter, the cluster centers remain approximately at the same positions.
According to Fig. 6.10a, clustering is most pronounced at a distance scale of $r = 200$ units. At this scale, the percentage of events that exceed the threshold reaches to a maximum. Therefore, this scale of observation can accepted to be the most informative scale regarding cluster geometries. Scales lower than $r = 200$ result in small individual clusters distributed as spikes over the matrix and higher scales result in formation of big merged clusters.
6.2.2 Quantitative Analysis of Composite Microstructures

6.2.2.1 Refined nearest neighbour analysis

The results of the nearest neighbour statistics indicate that, although the method is sensitive to various levels of clustering, it is not the best choice for quantitative analysis of particle distribution in metal matrix composites. The first reason is that, the method only considers the interaction of nearest particles and do not take the second, third or any other nearest particle into account. However, clusters in composites occur by involvement of many reinforcement particles and over many distance scales. The lack of information regarding the distribution of particles beyond the nearest neighbours makes this method a weak candidate in quantitative analysis of composite microstructures.

The refined nearest neighbour statistics can be presented by a variety of plots, which were demonstrated through Figs. 6.11 and 6.12. Fig. 6.11 shows the cumulative density function plots and the corresponding $\hat{G}(w) - G(w)$ variations. $\hat{G}(w) - G(w)$ plots simply express the difference between observed and theoretical (Poisson) distributions. Values below the confidence interval suggest uniform distribution, whereas values above the confidence interval indicate clustering of particles. Another way to present the nearest neighbour statistics is demonstrated through Figs. 6.12a, 6.12c and 6.12e. These plots present the nearest neighbour statistics of the observed pattern with reference to the Poisson distribution. Any deviation from linearity also indicates deviation from randomness. Similarly, values below the confidence interval suggest uniform distribution, whereas values above the confidence interval indicate clustering of particles.
Figure 6.11 Sample cumulative density function plots (on the left hand side) and $\hat{G}(w) - G(w)$ plots (on the right hand side) for A356 alloy composites reinforced with 10% SiC$_p$ for samples A1002 and A1009.

The refined nearest neighbour analysis was found to be effective in characterizing the global trends in distribution; however, cannot provide information about the local variations in the microstructure. The main issue in quantitative characterization of particle distribution is to relate the microstructure to the mechanical response of the composites. The nearest neighbour statistics fail to overcome this problem since the mechanical response is mainly determined by local variations in particle content and therefore cannot be described over a statistics that presents a summary that considers the interaction between closest particles.
Figure 6.12 Sample refined nearest neighbour analysis results for A356 alloy composites reinforced with 20% SiC$_p$ for sample A2004, A2009 and A2014.

It is interesting to note that all plots indicate a solid tendency towards regularity at length scales up to approximately 30 $\mu$m. This value corresponds to the average SiC size used in the present study (29.2 ± 1.5 $\mu$m). Since the particles cannot approach each other closer than their diameter, the statistics suggest regularity within this length scale.
6.2.2.2 Voronoi tessellation

The Voronoi tessellation method considers the area distribution of the polygonal cells to assess the level of inhomogeneity. The results indicate that, although the method yields a good pictorial impression of the spatial pattern of the reinforcement particles, it cannot provide a comprehensive description of the microstructure. Sample tessellated composite microstructures are presented in Fig. 6.13.

![Sample Voronoi plots derived from composite microstructures together with the corresponding CVA values for each particular micrograph. A higher CVA value indicates tendency of cells to aggregate into clusters.](image)

**Figure 6.13** Sample Voronoi plots derived from composite microstructures together with the corresponding $CV_x$ values for each particular micrograph. A higher $CV_x$ value indicates tendency of cells to aggregate into clusters.
The probability density functions of polygon area, \( f(A) \), for some composite microstructures are plotted in Fig. 6.14. One problem associated with this approach is the sensitivity of the method to the volume fraction particles. At higher volume fraction (20% SiC), the distinction between levels of clustering is not as clear as in lower volume fraction of particles (10% SiC). Yet, this comparison is only valid for composites with similar volume fraction of particles. Normally, the intensity of the peaks ceases as the level of inhomogeneity increases. However, the reduction in the intensity of the peaks with increasing volume fraction occurs due to the decrease in area gradients as a result of high particle density and should not be interpreted as an increase in inhomogeneity.

The terms “inhomogeneity” and “clustering” are frequently used as synonyms with regard to distribution of second phase particles in metal matrix composites. However, the clusters in such materials cannot be directly detected by measuring the level of inhomogeneity by some statistical method. The Voronoi diagrams and their corresponding probability density function plots indicate that, the method is actually informative when one needs to assess the level of inhomogeneity of a given spatial pattern. However, in the
case of metal matrix composites, one also needs to gather information on the locations, relative intensities and spatial connectivities of the clusters. Description of whole microstructures in terms of some scalar numbers obviously cannot provide these information. Therefore, Voronoi tessellation was also not considered as a suitable method in quantitative characterization of metal matrix composites.

6.2.2.3 $K$-function

$K$-function (also known as Ripley’s $K$-function) is a second order intensity function and is known as the most effective method for assessing departure from spatial randomness\textsuperscript{75, 76}. Similar to radial distribution function, it is very sensitive to local variations and it has the ability to characterize a given morphological pattern over many length scales. The $K$-functions for some composite microstructures are plotted in Fig. 6.15.

Figure 6.15 Sample $K$-function plots for a number of composites reinforced with (a) 10% SiC and (b) 20% SiC particles.
Figure 6.16 Sample $\hat{L}(r)$ function plots for composites reinforced with 10% and 20% SiC. The confidence intervals are not shown on the plots for simplicity, since each sample has different intensity resulting in different confidence limits.

Although $K$-function is quite sensitive to the trends in the pattern, it is commonly presented with a linearization transformation in order to bring out the differences between various patterns. Corresponding $\hat{L}(r)$ functions are plotted in Fig. 6.16. Instead of nearest neighbour statistics and the Voronoi tessellation, $K$-function is observed to detect any deviation from regularity over many length scales. The reinforcement particles aggregate into clusters at all distance scales greater than approximately 30 μm, which corresponds to the average SiC size used in the present study (29.2 ± 1.5 μm). The $K$-function results reveal that, clustering of reinforcement particles occur with arrangement of many particles over various distance scales. Therefore, the nature of these clusters can hardly be inferred by statistics that only considers the interaction with the nearest neighbours. This is another limitation that indicates the deficiency of nearest neighbour statistics and Voronoi tessellation to be employed in quantitative characterization of composite microstructures.
6.2.2.4 Local density statistics

As mentioned previously, although the $K$-function successfully measures the local density around each particle, presence of clustering can be inferred only when the local density around a particle exceeds a certain threshold value. These threshold values were obtained from a threshold function, $T_{0.95}(r)$, which expresses the probability that the local density around a particle, as depicted by $\rho(P_i, r)$, exceeds a certain value in an associated Poisson point process. Figs. 6.17 through 6.24 show sample microstructural patterns, together with the variation of above-threshold particles as a function of observation scale.

Figure 6.17 Evaluation of a micrograph from sample A1001: (a) Effect of observation scale on the amount of above-threshold particles and (b) clusters demonstrated by kernel surfaces at the observation scale where clustering is most pronounced ($r = 360 \mu m$).
Figure 6.18 Evaluation of a micrograph from sample A1002: (a) Effect of observation scale on the amount of above-threshold particles and (b) clusters demonstrated by kernel surfaces at the observation scale where clustering is most pronounced ($r = 400 \mu m$).

Figure 6.19 Evaluation of a micrograph from sample A1008: (a) Effect of observation scale on the amount of above-threshold particles and (b) clusters demonstrated by kernel surfaces at the observation scale where clustering is most pronounced ($r = 120 \mu m$).
Figure 6.20 Evaluation of a micrograph from sample A1009: (a) Effect of observation scale on the amount of above-threshold particles and (b) clusters demonstrated by kernel surfaces at the observation scale where clustering is most pronounced ($r = 200 \, \mu m$).

Figure 6.21 Evaluation of a micrograph from sample A2001: (a) Effect of observation scale on the amount of above-threshold particles and (b) clusters demonstrated by kernel surfaces at the observation scale where clustering is most pronounced ($r = 240 \, \mu m$).
Figure 6.22 Evaluation of a micrograph from sample A2001: (a) Effect of observation scale on the amount of above-threshold particles and (b) clusters demonstrated by kernel surfaces at the observation scale where clustering is most pronounced ($r = 260 \, \mu m$).

Figure 6.23 Evaluation of a micrograph from sample A2009: (a) Effect of observation scale on the amount of above-threshold particles and (b) clusters demonstrated by kernel surfaces at the observation scale where clustering is most pronounced ($r = 140 \, \mu m$).
As described previously, the local density around a particle may or may not exceed the corresponding clustering threshold depending on the scale of observation, \( r \). The subfigures (a) in Figs. 6.17 through 6.24 show the influence of scale on the amount of above-threshold particles for actual composite microstructures. From these plots, the amount of deviation from randomness can be quantified over many length scales in terms of the amount of particles that have higher local density as compared to an associated Poisson process with the same intensity. The intensity of the peaks in these plots indicates the amount of particles that exceeds the clustering threshold.

The detected clusters are demonstrated by kernel surfaces in subfigures (b) in Figs. 6.17 through 6.24 at observation scales where clustering is most pronounced. It is interesting to note that, samples solidified with relatively slow and fast cooling rates display large clusters with arrangement of many above-threshold particles. The samples solidified with relatively moderate cooling rates, on the other hand, display small clusters, randomly distributed throughout the matrix. This result was verified to hold for all samples produced in this study and will be discussed in detail in section 6.3.2. No information regarding the cluster dimensions could be inferred from previously described statistical functions. Demonstration of the intensity variation of all particles also cannot help to obtain a clear picture about the cluster dimensions. One such example is illustrated.
in Fig. 6.25, which compares the visual effectiveness of the local density statistics with the intensity variation of all particles, as demonstrated by kernel surfaces. The kernel surfaces, when associated with all particles, display similar trends with the intensity variation of the above-threshold particles in the clustered regions. However, these plots cannot provide information on the scales of clustered regions. The kernel surfaces associated with above-threshold particles, on the other hand, provide a much better description of the clustered regions by avoiding the particles, which do not agglomerate into clusters.

![Figure 6.25 Kernel surfaces demonstrating the intensity variation of (a) all particles and (b) above-threshold particles.](image)

The main aim of quantitative characterization of particle distribution is to relate the microstructure to the mechanical response of the composite materials. The results reveal that methods such as the nearest neighbour statistics or Voronoi tessellation have limited use in these systems since the mechanical response of the composites is determined by local variations in particle content and therefore cannot be described over a statistics that presents a summary of the interaction between nearest particles. The presented methodology, on the other hand, describes the pattern from the perspective of individual particles and even in a microstructure that globally conforms to random distribution, local clusters, which may actually control the mechanical response can be detected.
One problem, however, associated with this method is that not every particle that belongs to a cluster is included in the set of above-threshold particles, $S(r)$. As an example, let us consider an individual cluster composed of many SiC particles and surrounded by matrix only. Although the local density around the particles in the cluster core will exceed the clustering threshold, some particles close to cluster surface may not be accepted to $S(r)$ since the local density around these particles may be lower than the threshold. This limitation will especially be more pronounced as the particles form compact and well-separated clusters. In such cases, although the cluster locations can still be determined precisely, the amount of above-threshold particles may not reflect the actual amount of clustered particles. However, since the clusters in discontinuously reinforced composites are in the form of local variations in particle content rather than compact, well-separated agglomerates, the extent of this problem will not be realized significantly.

6.2.2.5 Effect of metallographic field size on cluster dimensions

The results of the local density statistics show that clusters in metal matrix composites can successfully be detected and visualized by the described approach. However, in all attempts the metallographic field size was constant ($1376 \times 1017 \mu m$). We have also investigated the effect of metallographic field size on the size of the detected clusters. In order to capture the long range spatial patterns with high resolution we have performed the calculations on digital montages of contiguous microstructural fields (see Fig. 5.6).

Fig.6.26 shows the corresponding results of the local density statistics. A comparison from Fig. 6.26a between the samples reinforced with 10% and 20% SiC particles indicates that the sample reinforced with 10% SiC has higher level of clustering at all studied length scales, including the scale at which clustering is most pronounced.
Figure 6.26 (a) Variation in the amount of above-threshold particles with scale of observation; (b-c) kernel surfaces associated to the set of above-threshold particles at scales where clustering is most pronounced and; (d-e) at an observation scale of 140 µm for composites reinforced with 10% and 20% SiC$_p$. 
Figs. 6.26b-e shows the kernel surfaces associated to the set of above-threshold particles at scales where clustering is most pronounced to analyze the influence of scale on cluster locations. While varying the scale of observation from $r = 260 \mu m$ to $r = 140 \mu m$, the cluster locations do not vary significantly in the sample with 10% SiC since the differences in length scales and amount of above-threshold particles at these scales are relatively small. In the sample with 20% SiC, on the other hand, although the locations where the clusters are detected remain almost the same, there is significant difference in the dimensions of the detected clusters. This is due to the fact that when the calculations are performed at a length scale of $r = 140 \mu m$, the nature of the pattern beyond 140 $\mu m$ cannot be seen and therefore remains unevaluated. Consequently, the analysis results in small individual clusters distributed throughout the matrix, which do not seem to represent the actual cluster dimensions since no information is available beyond 140 $\mu m$. Upon increasing the length scale to $r = 440 \mu m$, some particles are rejected from $S(r)$ and some small clusters disappear since the local density around these particles does not exceed the clustering threshold anymore. However, a larger number of them are found to exceed the clustering threshold (the increasing trend in Fig. 6.26a). At this larger scale, the kernels merge to form bigger clusters, which can be accepted to represent a more accurate picture, since the number of particles that exceed the threshold reach to a maximum at this scale. Further increasing the length scales result in a decrease in the amount of above-threshold particles and the difference of the patterns from random distribution start to diminish.

It is interesting to note that, although there are considerable differences in cluster dimensions, the scales where clustering is most pronounced do not vary with metallographic field size. In all attempts, regardless of the metallographic field size, clustering was most pronounced at scales ranging between 150 – 500 $\mu m$. Therefore, in order to quantify the scale of clustering, one can make calculations on relatively small metallographic fields and end up with correct results. However, in order to reveal the actual dimensions of clusters, it is always better to work on micrographs that represent larger metallographic fields.
6.2.2.6 Comparison of methods

The refined nearest neighbour analysis and Voronoi tessellation were found to be effective in characterizing the global trends in distribution; however, cannot provide information about the local variations in the microstructure. The main issue in quantitative characterization of metal matrix composites is to relate the microstructure to the mechanical response of the composites. The nearest neighbour statistics and Voronoi tessellation fail to overcome this problem since the mechanical response is mainly determined by local variations in particle content and therefore cannot be described over a statistics that presents a summary that considers the interaction between closest particles. K-function was observed to be very sensitive to local variations in particle content and can effectively characterize the pattern over many distance scales. However, this method also fails to provide visual information on locations and dimensions of the clusters.

The local density statistics was observed to provide a much better description of the spatial pattern over many distance scales. Plots that show the variation of above-threshold particles with observation scale reveal that, the actual nature of clusters can hardly be inferred from the distances between nearest particles since clustering is evidenced over much larger scales of observation compared to nearest neighbour distances. The presented method considers the distribution from the perspective of individual particles, so that even in a pattern that globally conforms to random distribution, local clusters, which may actually control the mechanical response, can be detected. Results show that the presented methodology is an effective discriminator of clustered particles and can successfully be used for quantitative characterization of particle distribution.
6.3 Effect of Solidification Rate on Clustering of SiC Particles

6.3.1 Effect of Solidification Rate on Dendritic Structure

Distribution of SiC particles was evaluated in terms of two parameters related to primary solidification of composites; secondary dendrite arm spacing (SDAS) and volume fraction of solid phase at dendrite coherency point ($f_{SDCP}$). We have examined the influence of solidification rate on these parameters for A356 alloy composites reinforced with 10% and 20% SiC_p. The influence of different SiC_p contents on SDAS was negligible, in agreement with the previous results reported in literature. Power law fits to the experimental data for composites with 10% and 20% SiC_p contents yield;

\[
\begin{align*}
A356-10\%\text{ SiC}_p: \quad \lambda_2 &= 37.2 \cdot T^{-0.52} \quad (R^2 = 0.954) \\
A356-20\%\text{ SiC}_p: \quad \lambda_2 &= 38.1 \cdot T^{-0.56} \quad (R^2 = 0.949)
\end{align*}
\]

where, $\lambda_2$ is SDAS in $\mu$m and $T$ is cooling rate in $^\circ$C sec$^{-1}$. There exists a discrepancy regarding the cooling rate dependence of coherency point in the literature. Arnberg et al. reported a decrease in $f_{SDCP}$ with increasing cooling rate. Veldman et al. on the other hand, concluded from their research that an alteration to cooling rate alone has a negligible effect on $f_{SDCP}$. In the present study, no relationships could be established between the solidification rate and the calculated coherencies. Although the coherency predictions obtained by NTA were observed to be in a slowly decreasing trend, FTA results were randomly scattered within the range of investigated solidification rates. As far as the effect of particle content is concerned, FTA results seem to yield a more accurate picture since, contrary to NTA, the $f_{SDCP}$ values obtained by FTA decrease as the particle content is increased from 10% to 20%. This is actually expected since the thermal conductivity of SiC is much higher than that of A356 alloy at the temperature range of coherency. Therefore, the SiC particles between growing dendrite arms contribute to the established fast channel of heat transfer and manipulate the coherency values, resulting in early predictions in general.
6.3.2 Effect of Solidification Rate on Particle Distribution

It was observed that the relationship of particle distribution to solidification rate can be characterized by three intervals for both reinforcement contents, which were denoted as intervals 1, 2 and 3 in Tables 6.3 and 6.4 and Figs. 6.27 and 6.28. First interval was characterized by a continuous decrease in SDAS with increasing solidification rate. Both statistical functions suggested a decrease in amount of clustering with increasing solidification rate in this interval for both reinforcement contents. The $f_S^{DCP}$ values were observed to be almost independent of solidification rate. In the second interval, further increasing the cooling rate did not create significant alterations in SDAS and the spacings remained almost constant around 40 $\mu$m. Interestingly, the corresponding statistics also did not suggest any significant tendency in this range. The $f_S^{DCP}$ values were still fluctuating, however at relatively lower values as compared to the first interval. In the third interval SDAS values were smaller than the average particle size ($29.2 \pm 1.5 \mu$m). In contrary to first two intervals, the statistical functions suggest tendency towards clustering. The $f_S^{DCP}$ values exhibited sharp alterations, maintaining its scattered nature.

The tabulated results of statistical functions indicate that distribution of SiC particles is mainly determined by secondary dendrite arm spacings. This dependence is characterized by two steps in the first two intervals: (i) the growing secondary dendrite arms push the SiC particles into interdendritic regions, where the particles start to agglomerate; (ii) during growth of dendrite arms, there also occurs a fluid flow through these arms to feed the solidification shrinkage. This flow carries some particles from interdendritic spaces towards the roots of secondary arms, therefore preventing formation of larger clusters.
<table>
<thead>
<tr>
<th>Interval</th>
<th>Sample</th>
<th>SR (°C sec⁻¹)</th>
<th>SDAS (μm)</th>
<th>(f_s^{DCP}) (NTA)</th>
<th>(f_s^{DCP}) (FTA)</th>
<th>% Threshold</th>
<th>(Q)-ratio</th>
<th>(CV_d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 – 10% SiC&lt;sub&gt;p&lt;/sub&gt;</td>
<td>A1001</td>
<td>0.164</td>
<td>86.99 ± 12.51</td>
<td>0.271</td>
<td>0.203</td>
<td>17.781 ± 6.183</td>
<td>0.809 ± 0.067</td>
<td>0.827 ± 0.111</td>
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<td>A1002</td>
<td>0.242</td>
<td>72.70 ± 11.32</td>
<td>0.278</td>
<td>0.207</td>
<td>18.320 ± 7.949</td>
<td>0.815 ± 0.035</td>
<td>0.806 ± 0.103</td>
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<td>A1003</td>
<td>0.280</td>
<td>70.21 ± 11.45</td>
<td>0.273</td>
<td>0.174</td>
<td>18.539 ± 7.628</td>
<td>0.817 ± 0.063</td>
<td>0.839 ± 0.161</td>
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<td></td>
<td>A1004</td>
<td>0.345</td>
<td>72.23 ± 9.30</td>
<td>0.227</td>
<td>0.138</td>
<td>15.161 ± 6.427</td>
<td>0.853 ± 0.022</td>
<td>0.731 ± 0.242</td>
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<td>A1005</td>
<td>0.411</td>
<td>62.14 ± 8.69</td>
<td>0.270</td>
<td>0.212</td>
<td>16.021 ± 6.877</td>
<td>0.862 ± 0.028</td>
<td>0.749 ± 0.073</td>
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<td>A1006</td>
<td>0.429</td>
<td>60.58 ± 9.83</td>
<td>0.233</td>
<td>0.218</td>
<td>15.778 ± 7.496</td>
<td>0.858 ± 0.053</td>
<td>0.747 ± 0.097</td>
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<tr>
<td>2 – 10% SiC&lt;sub&gt;p&lt;/sub&gt;</td>
<td>A1007</td>
<td>0.596</td>
<td>42.44 ± 4.67</td>
<td>0.244</td>
<td>0.146</td>
<td>11.184 ± 6.764</td>
<td>0.892 ± 0.063</td>
<td>0.688 ± 0.078</td>
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<td>A1008</td>
<td>0.682</td>
<td>43.10 ± 7.42</td>
<td>0.238</td>
<td>0.141</td>
<td>11.925 ± 5.631</td>
<td>0.876 ± 0.067</td>
<td>0.808 ± 0.182</td>
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<td>A1009</td>
<td>0.725</td>
<td>48.20 ± 5.35</td>
<td>0.204</td>
<td>0.136</td>
<td>12.234 ± 3.447</td>
<td>0.881 ± 0.044</td>
<td>0.656 ± 0.101</td>
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<td>A1010</td>
<td>0.795</td>
<td>41.12 ± 6.12</td>
<td>0.209</td>
<td>0.195</td>
<td>12.285 ± 4.339</td>
<td>0.886 ± 0.035</td>
<td>0.682 ± 0.049</td>
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<td>A1011</td>
<td>0.981</td>
<td>37.40 ± 3.17</td>
<td>0.229</td>
<td>0.231</td>
<td>11.121 ± 3.842</td>
<td>0.878 ± 0.029</td>
<td>0.661 ± 0.071</td>
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<td>A1012</td>
<td>1.121</td>
<td>40.10 ± 2.95</td>
<td>0.196</td>
<td>0.147</td>
<td>12.282 ± 4.399</td>
<td>0.912 ± 0.016</td>
<td>0.692 ± 0.117</td>
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<td></td>
<td>A1013</td>
<td>1.329</td>
<td>35.12 ± 2.21</td>
<td>0.166</td>
<td>0.197</td>
<td>11.287 ± 3.371</td>
<td>0.891 ± 0.021</td>
<td>0.667 ± 0.058</td>
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<td>3 – 10% SiC&lt;sub&gt;p&lt;/sub&gt;</td>
<td>A1014</td>
<td>1.928</td>
<td>24.89 ± 2.42</td>
<td>0.171</td>
<td>0.231</td>
<td>14.882 ± 4.011</td>
<td>0.852 ± 0.049</td>
<td>0.726 ± 0.088</td>
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<td>A1015</td>
<td>2.319</td>
<td>21.36 ± 2.78</td>
<td>0.129</td>
<td>0.204</td>
<td>15.687 ± 4.971</td>
<td>0.861 ± 0.031</td>
<td>0.731 ± 0.078</td>
</tr>
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SDAS and statistical results are presented as mean ± standard deviation.
Table 6.4 Effect of solidification rate (SR) on SDAS, $f_S^{DCP}$ and corresponding summary statistics for A356 composites reinforced with 20% SiCₚ

<table>
<thead>
<tr>
<th>Interval</th>
<th>Sample</th>
<th>SR (°C sec⁻¹)</th>
<th>SDAS (μm)</th>
<th>$f_S^{DCP}$ (NTA)</th>
<th>$f_S^{DCP}$ (FTA)</th>
<th>% Threshold</th>
<th>Q-ratio</th>
<th>CV₄</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 – 20% SiCₚ</td>
<td>A2001</td>
<td>0.213</td>
<td>83.03 ± 6.32</td>
<td>0.268</td>
<td>0.117</td>
<td>13.952 ± 3.667</td>
<td>0.931 ± 0.019</td>
<td>0.684 ± 0.071</td>
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<td>A2002</td>
<td>0.309</td>
<td>76.68 ± 5.85</td>
<td>0.288</td>
<td>0.130</td>
<td>12.647 ± 7.241</td>
<td>0.952 ± 0.048</td>
<td>0.657 ± 0.048</td>
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<tr>
<td>A2003</td>
<td>0.353</td>
<td>69.57 ± 5.92</td>
<td>0.291</td>
<td>0.107</td>
<td>10.130 ± 2.526</td>
<td>0.969 ± 0.051</td>
<td>0.688 ± 0.063</td>
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<td>A2004</td>
<td>0.429</td>
<td>65.23 ± 7.21</td>
<td>0.252</td>
<td>0.168</td>
<td>11.329 ± 5.365</td>
<td>0.963 ± 0.043</td>
<td>0.624 ± 0.064</td>
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<td>A2005</td>
<td>0.566</td>
<td>54.40 ± 7.03</td>
<td>0.252</td>
<td>0.154</td>
<td>10.801 ± 4.221</td>
<td>0.957 ± 0.073</td>
<td>0.672 ± 0.051</td>
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<td>2 – 20% SiCₚ</td>
<td>A2006</td>
<td>0.652</td>
<td>42.83 ± 7.12</td>
<td>0.256</td>
<td>0.122</td>
<td>8.932 ± 3.541</td>
<td>0.994 ± 0.011</td>
<td>0.617 ± 0.054</td>
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<td>40.60 ± 5.19</td>
<td>0.231</td>
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<td>8.719 ± 4.050</td>
<td>0.986 ± 0.023</td>
<td>0.617 ± 0.054</td>
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<td>7.868 ± 3.999</td>
<td>1.036 ± 0.029</td>
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<td>1.024 ± 0.025</td>
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<td>0.973 ± 0.021</td>
<td>0.641 ± 0.052</td>
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SDAS and statistical results are presented as mean ± standard deviation.
Figure 6.27 Effect of SDAS on clustering of reinforcement particles in A356 alloy composites reinforced with 10% SiC<sub>p</sub>.

Figure 6.28 Effect of SDAS on clustering of reinforcement particles in A356 alloy composites reinforced with 20% SiC<sub>p</sub>.
In the third interval, where the spacings were smaller than the average particle size, however, there was a net increase in amount of clustering. Karnezis et al.\textsuperscript{80} also observed a similar behavior in squeeze cast samples in which the arm spacings were lower than the average particle size. This reversed trend occurs when the fluid flow due to shrinkage cannot carry the particles through the finer secondary arms. The particles were therefore pushed by the finer mesh of secondary arms, resulting in formation of larger clusters\textsuperscript{80}.

The microstructural trends in these intervals are illustrated in Fig. 6.29 through sample micrographs. In the first interval (Fig. 6.29a), the particles accumulate at relatively large interdendritic regions. In the second interval (Fig. 6.29b), where the arm spacings are slightly larger than the average particle size, the number of particles that could be contained in these interdendritic spaces decreases significantly, leading to a considerable decrease in the amount of clustering. In the third interval (Fig. 6.29c), where the arm spacings are lower than the average SiC size, the particles cannot penetrate through the finer mesh of secondary arms and the amount of clustering again increases.

There is also a significant difference in amount of clustering between composites reinforced with 10% and 20% SiC\textsubscript{p}. All statistical functions indicate a tendency towards regularity with an increase in SiC\textsubscript{p} content from 10% to 20%. This tendency can be explained by two factors: (i) the statistical functions presented in this study are quite sensitive to local variations in distribution at low volume fractions, however, as the volume fraction of the particles increase the functions start to lose their effectiveness and the distinction between different clustering levels diminishes\textsuperscript{25}; (ii) the presence of hard, insoluble particles increases the viscosity of the melt to a great extent\textsuperscript{81}. Increased viscosity at higher volume fraction of particles suppresses the convection during solidification of the composite and hinders the settling of reinforcements\textsuperscript{82, 83}. Therefore, the particles almost preserve their positions in the liquid state prior to solidification and show less tendency to agglomeration since faster settling rates at low fractions and convection effects contribute to clustering of reinforcement particles\textsuperscript{7}.
Figure 6.29 Micrographs from samples (a) A1002, (b) A2009 and (c) A1015, representing the microstructural trends in the first, second and third intervals, respectively.
6.4 Modelling of MMC Solidification

The results of the previous chapters indicate that, distribution of second phase particles is mainly determined by the secondary dendrite arm spacings. In general, finer arm spacings result in lower clustering levels. However, when the arm spacings are lower than the average particle size, this tendency is reversed, leading to an increase in the amount of clustering.

Therefore, in order to predict the cluster formation tendency in any cast part before production, one should be able to determine the dendritic arm spacing distribution throughout the casting. Such information would bring the advantage of assessing the critical regions in cast parts, where the particles are prone to agglomerate and help the engineers to come up with solutions by altering the solidification conditions or the design of the component. The final part of this study, therefore, focuses on numerical approaches to composite solidification to predict the local solidification rates and the resulting secondary dendrite arm spacings.

6.4.1 Prediction of Local Solidification Rate

A series of cooling curve simulations were performed to predict the local solidification rate distributions within the cylindrical castings. In each simulation, the experimental conditions were duplicated by entering the mold dimensions to the computer program. The simulated cooling curves were also plotted from the same radial locations with the experimental curves. The schematic illustrating the mold dimensions and the thermocouple locations is shown in Fig. 6.30.
Figure 6.30 Schematic illustrating the mold dimensions and thermocouple locations.

Figs. 6.31 through 6.33 show the experimentally determined and simulated cooling curves for A356 alloy composites reinforced with SiC particles. Figs. 6.31a, 6.32a and 6.33a present the cooling curves obtained from the thermocouples located at the geometrical center ($r = 0$ mm) of the castings and Figs. 6.31b, 6.32b and 6.33b show the cooling curves obtained from various radial distances from the center, together with the simulated curves. The results reveal good agreement between the experimental and simulated cooling curves, especially in terms of local solidification time predictions.
Figure 6.31 The simulated and experimental cooling curves of A356 alloy composite reinforced with 10% SiC<sub>p</sub> (sample A1002). The mold dimensions are $D = 50$ mm, $H = 60$ mm, $T = 12.5$ mm, and the outer thermocouples are located at $r_1 = 9$ mm and $r_2 = 16$ mm.
Figure 6.32 The simulated and experimental cooling curves of A356 alloy composite reinforced with 20% SiC_p (sample A2002). The mold dimensions are \( D = 55 \) mm, \( H = 60 \) mm, \( T = 12.5 \) mm, and the outer thermocouples are located at \( r_1 = 8 \) mm and \( r_2 = 14 \) mm.
Figure 6.33 The simulated and experimental cooling curves of A356 alloy composite reinforced with 20% SiC$_p$ (sample A2008). The mold dimensions are $D = 30$ mm, $H = 50$ mm, $T = 15$ mm, and the outer thermocouples are located at $r_1 = 5$ mm and $r_2 = 9$ mm.
There still exist some differences between the experimental and simulated curves, which may better be understood by close examination of the solidification kinetics in terms of evolution of the solid. Fig. 6.34 presents the variation of solidification rates during the course of solidification obtained from experimental cooling curves and simulation for sample A2002. The experimental solid fraction results were obtained by Fourier and Newtonian thermal analysis methods. The solidification rate of the simulated sample exhibits a higher initial peak (which corresponds to the nucleation and growth of primary $\alpha$-Al dendrites) compared to the initial peaks of the experimental curves obtained by NTA and FTA. The simulated curve also ceases more rapidly as compared to the solidification rate curves of the actual sample. This is most probably due to the instantaneous nucleation model used in the calculations, which assumes that all nuclei are generated at the nucleation temperature. Therefore, the model predicts a high value of latent heat release at the initial stage of primary solidification. However, because the actual transformation proceeds with continuous nucleation during the interval of primary solidification, the initial maxima of the experimental curves are lower and the subsequent decrease in the solidification rates are slower.

![Figure 6.34 Variation of solidification rate as a function of solid fraction for sample A2002 obtained by NTA and FTA, together with simulation result.](image-url)
The magnitudes of the second maxima are observed to be reasonably in agreement due to the continuous nucleation model used in eutectic solidification. However, the start of eutectic solidification in the simulated curve corresponds to a higher solid fraction as compared to the experimental observations as derived by FTA and NTA. The delay in eutectic nucleation cannot be detected when the transformation start times are considered on the cooling curves. This quantitative discrepancy indicates that further improvements are required to the model to simulate the nucleation kinetics of the eutectic grains.

There exists a third peak in the solidification rate curve of the FTA processed experimental cooling curve, which corresponds to formation of intermetallic phases such as Mg$_2$Si and β-Al$_5$FeSi, which was also illustrated in Fig. 6.2. This third peak is not visible on the simulated solidification rate curve. This is because of the pseudo-binary alloy assumption, which considers all alloying elements other than Si as solutes and calculates and equivalent solute for the eutectic reaction. Therefore, no extra transformation is visible after the eutectic reaction in the simulated curves.

6.4.2 Prediction of Secondary Dendrite Arm Spacings

As mentioned previously, we have assumed that the presence of SiC particles do not have any influence on the secondary dendrite arm spacings, which is in accordance with the previously reported results in the literature$^{1, 59}$ and with our experimental findings. Therefore, it was assumed that the dynamic coarsening model developed by Mortensen$^{65}$ can be applied both reinforcement contents (10% and 20%) without any modifications. Fig. 6.35 presents variation of SDAS as function of local solidification time for the composites, together with the predicted results from Eq. (4.30).
Figure 6.35 Predicted and experimental variations of secondary dendrite arm spacings as a function of local solidification time.

The experimental and predicted SDAS values are found to be in reasonably good agreement. Although the predicted and experimental variations have very similar trends, the predicted values are observed to be slightly higher than the actual spacings at local solidification times lower than approximately 50 seconds. This limitation may be a problem in correct prediction of SDAS values in rapidly cooled samples that belong to the third interval and therefore estimating the cluster formation tendencies. The origin of the quantitative discrepancies may be attributed to presence of factors not considered by the coarsening model.

Fig. 6.35 reveals that, the arm spacing distribution throughout any cast part can be predicted if the local solidification rate values at those locations are correctly calculated. Therefore, one can assess the local arm spacing at particular regions in a casting and evaluate the risk of cluster formation by looking at the particle size to SDAS ratio. One such example is shown in Fig. 6.36, which demonstrates the variation of SDAS values as a function of radial location within the casting for an A356 alloy composite reinforced with 20% SiC particles (sample A2009). It was observed that there exists a good agreement
between the experimental and predicted trends. However, the predicted values are slightly lower than the mean SDAS values all throughout the radial locations, which again may be attributed to the limitations discussed above and also to wrong estimation of local solidification times, which arises from incorrect description of the heat transfer system as a result of limitations in the thermophysical parameters.

![Graph showing predicted and experimental variations of secondary dendrite arm spacings as a function of radial location within the cylindrical casting for sample A2009.]

**Figure 6.36** Predicted and experimental variations of secondary dendrite arm spacings as a function of radial location within the cylindrical casting for sample A2009.
CHAPTER 7

CONCLUSIONS

1. The distribution of SiC particles in aluminum alloy composites was found to be highly dependent on secondary dendrite arm spacing (SDAS), which is a strong function of local solidification rate. In general, with decreasing arm spacings, particles exhibit less tendency towards clustering. At higher solidification rates, however, this tendency is reversed, resulting in formation of larger clusters. This reversed trend occurs when the average secondary dendrite arm spacing decreases below the average particle size. The particles cannot penetrate through the finer mesh of secondary arms and agglomerate in larger interdendritic channels. The results indicate that, in order to attain the lowest possible amount of particle clustering, the secondary dendrite arm spacings must lie within the range of $d_{SiC} < SDAS < 2d_{SiC}$, where $d_{SiC}$ is the average particle diameter.

2. No relationships could be established between the as-cast particle distribution and the fraction of solid phase at the dendrite coherency ($f_s^{DCP}$). $f_s^{DCP}$ indicates the fraction of solid phase when a solid network is established throughout the casting. This solid fraction value, however, does not give any information on the length scales that the particles were pushed by the growing dendrites during the course of solidification. Therefore, the fraction of solid phase at the dendrite coherency is not considered as a determining factor in clustering of reinforcement particles.
3. The applicability of various spatial characterization methods was investigated. Results indicate that, methods that describe the microstructural pattern over a statistics that presents a summary of the interaction between nearest particles have limited use in discontinuously reinforced composites since clustering in such materials is evidenced over many length scales. Methods such as the nearest neighbour statistics and Voronoi tessellation can describe the microstructural heterogeneity to a certain extent, however, cannot provide information on the characteristics of the pattern at length scales beyond nearest neighbour distances.

$K$-function is observed to be an effective solution to overcome this problem, since it has the ability to analyze the microstructural pattern over many length scales. However, similar to the above aforementioned methods, $K$-function also cannot provide any sort of information regarding dimensions, locations and spatial connectivities of the detected clusters. However, as far as the fracture mechanics of the discontinuously reinforced composites is concerned, one also needs to gather such information in order to successfully relate the mechanical response of the composite to its microstructure.

The local density statistics, on the other hand, was observed to provide a much better description of the composite microstructures. The method considers the microstructural pattern from the perspective of individual particles, therefore even in a pattern that globally conforms to random distribution, local clusters, that may actually control the mechanical properties, can be detected.

The variation of above-threshold particles with observation scale show that clustering of SiC particles is most pronounced at length scales ranging between 150 - 500 μm. In general, slower solidification rates result in large clusters. The samples solidified with higher cooling rates, on the other hand, display small clusters, randomly distributed throughout the matrix. Further increasing the solidification rate, again produces large clusters due to agglomeration of particles within large interdendritic spaces.
The effect of metallographic field size on the corresponding statistics was also analyzed. It was observed that the length scales where clustering is most pronounced do not vary with metallographic field size. However, there are considerable differences in observed cluster dimensions. When the metallographic field area is increased, the kernel surfaces emerging from individual clusters merge together to reveal the actual cluster morphologies. Therefore, although clustering levels can successfully be quantified from relatively small micrographs, the actual dimensions of clusters can only be revealed from micrographs that represent much larger visual fields compared to reinforcement particle size.

4. Coupling of macroscopic heat transport with solidification kinetics through the latent heat method was observed to provide a neat description of MMC solidification process, especially in terms of local solidification time predictions. The experimental and predicted SDAS values are found to be in reasonably good agreement. Small quantitative discrepancies can be attributed to the presence of factors not considered in the model and incorrect description of the heat transfer system as a result of limitations in the thermophysical parameters.

It was concluded that locations in a casting where there exists a tendency towards clustered particle arrangements can be successfully predicted prior to production with the described numerical approach. This advantage will obviously help engineers to come up with solutions to produce cast composites with more uniform microstructures and hence less prone to failure.

5. The present study differs from previous investigations in three ways. Firstly, the results hereby reported marks the first attempt in the literature to bring a quantitative approach to effect of dendritic solidification on spatial distribution of reinforcement particles.

Another difference is that, this study approaches to the problem of clustering from a different perspective and quantifies the amount of clustering from the perspective of individual particles rather than global trends in the pattern. This is a much appropriate way to look at the problem under consideration since clusters in MMCs are in the form of local variations in particle content rather than compact,
well separated agglomerates and therefore are hard to detect by conventional methods of spatial statistics.

Finally, the described computational strategy marks the first attempt reported in the literature to predict the risk of cluster formation as a function of local solidification rate. With this approach, certain locations in a casting, where there exists a tendency towards clustered particle arrangements can be predicted before production. Such an ability will obviously help engineers to come up with solutions to produce these components with more uniform microstructures and hence, less prone to failure.
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APPENDIX A

PHASE DIAGRAM AND SOLIDIFICATION PATH
OF A356 ALLOY

A356 is basically an Al - 7% Si - 0.35% Mg alloy. In order to present an accurate picture of the solidification path of the alloy and also the phases present at room temperature, various isothermal sections obtained from the ternary Al-Si-Mg phase diagram and solidification sequence of various phases were calculated by Pandat Multi-Component Phase Diagram Calculation Software (CompuTherm LLC, v5).

Figure A1 Isothermal sections from the Al-Si-Mg ternary phase diagram.
Figure A1 Isothermal sections from the Al-Si-Mg ternary phase diagram (continued).
Table A1 Theoretical non-equilibrium (Scheil) solidification sequence of A356 alloy calculated by Pandat Multi-Component Phase Diagram Calculation Software.

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<th>T (°C)</th>
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<th>X (MG)</th>
<th>X (SI)</th>
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Table A2 Theoretical non-equilibrium (Scheil) solid phase evolution of A356 alloy calculated by Pandat Multi-Component Phase Diagram Calculation Software.

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# APPENDIX B

## MOLD DIMENSIONS AND THERMOCOUPLE LOCATIONS

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There is also a third thermocouple ($r_1$) placed at the center of each cylindrical mold ($r = 0$ mm).
Figure B1 A sample cylindrical thermal analysis cup with three thermocouples placed at three different radial locations.
VITA

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Voice : +90-532-408-0902
E-mail : ardacetin@gmail.com

EDUCATION

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WORK EXPERIENCE

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PUBLICATIONS


