In situ Synchrotron
Tomographic Quantification of
Semi-solid Properties of
Aluminum-Copper alloys

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Abstract

Semi-solid deformation mechanisms are important in a range of manufacturing and natural phenomena, which range from squeeze casting to magma flows.

In this thesis, using high speed synchrotron X-ray tomography and a bespoke precision thermo-mechanical rig, a four dimensional (3D plus time) quantitative investigation was performed to study the mechanical / rheological behavior of semi-solid Al-Cu alloys. Various deformation techniques, namely, isothermal semi-solid compression, extrusion and indentation were used. The time-resolved dynamic 3D images were analyzed with the help of novel image quantification techniques including digital volume correlation and image-based simulations of fluid flow. The quantified dynamics at a microstructural scale was then linked with macroscopic mechanical properties.

The qualitative and quantitative analyses revealed a range of important semi-solid micromechanical mechanisms including the occurrence and effects of dilatancy, associated liquid flow through the equiaxed microstructure, intra-dendritic deformation, and strain localization during semi-solid deformation, not only shedding new insights into the mechanisms of deformation-induced solidification defect formation (solute segregation, porosity and hot tearing) of semi-solid alloys at both a macroscopic and microscopic level, but also providing benchmark cases for semi-solid deformation models and theories. The experimental methodology, techniques and analysis procedures developed in this thesis are generic in nature and can be applied to a wide range of research fields.
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Publications

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10. **B.Cai***, P.D. Lee et al, **In situ** synchrotron tomographic quantification of strain localization during hot compression of Al-15wt.%Cu alloy, *Euromat* 2014, Sep. 2013


Declaration

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Nomenclature

Roman Symbols

\(v\)  
Fluid velocity

\(\mu\)  
Viscosity of the fluid/slurry

\(P\)  
Pressure

\(T\)  
Deviatoric component of the total stress tensor

\(f\)  
Body forces per unit volume acting on the fluid

\(k\)  
Equilibrium partition coefficient

\(u\)  
Velocity

\(V\)  
Displacement

Greek Symbols

\(\tau\)  
Shear stress

\(\kappa\)  
Permeability

\(\mu\)  
Viscosity of the fluid/slurry

\(\varepsilon\)  
Strain

\(\gamma\)  
Surface tension

Other Symbols

\(\Delta T\)  
Freezing range

\(P_s\)  
Pressure drop due to solidification shrinkage

\(P_g\)  
Equilibrium pressure of dissolved gas

\(P_{s-t}\)  
Pressure due to pore-liquid surface tension

\(T_m\)  
Melting point of solvent
\(T_L\)  Liquid temperature of the alloy  
\(K_c\)  Kozeny constant  
\(\rho_l\)  Liquid density  
\(\dot{\gamma}\)  Shear rate  
\(f_s\)  Volume solid fraction  
\(f_\ell\)  Volume liquid fraction  
\(S_v\)  Surface area of the solid per unit volume of sample  
\(\rho_f\)  Fluid density  
\(S_v\)  Surface area of the solid per unit volume of sample  
\(\mu_0\)  Attenuation coefficient of a homogeneous material  
\(\varepsilon_n\)  Octahedral normal strain  
\(\varepsilon_s\)  Octahedral shear strain  

**Acronyms**  
DVC  Digital volume correlation  
LC  Liquid channel  
DC  Direct chill
Chapter 1. Introduction

1.1 Thesis overview

Solidification, a phase transformation from liquid to solid, is a science that has been studied for centuries [1]. In a multicomponent alloy, as the temperature drops, crystal embryos will nucleate from the melt, typically growing in the form of dendrites under most of relevant casting circumstances. The art of controlling this solidification process forms the backbone of modern metals engineering, ranging from turbine blades for the airplane engines [2–4] to metal houses for laptops [5]. There is also a strong drive for better engineered and manufactured components with recent emphasis on reducing energy consumption and carbon emissions. One of the keys to achieving this is via controlling the microstructure and reducing defects during solidification.

Deformation is a vital aspect of solidification processes, although it is a double-edged sword. On the one hand, it can lead to various solidification defects if not properly controlled, including micro-porosity [6], segregation [7] and hot tearing [8], which are detrimental to the final components. On the other hand, there are many benefits of dedicatedly imposing deformation upon solidifying alloys, for instance, refining and modulating the grain structure [9], and diminishing defects [10,11]. Notably, this has led to a number of advanced manufacturing technologies (e.g. high pressure die casting [12], twin roll casting [7,13,14], semi-solid processing [9,11]). Most of these techniques aim at reducing the number of processing routes in converting a liquid alloy into an engineering component, thus reducing costs and energy consumption. However, again, defects (e.g. deformation induced porosity and segregation bands [6]) have become issues of major concern restricting their widespread
applications in industry. It is therefore of significant importance to provide a better understanding of deformation responses of semi-solid alloys, which could be helpful to prevent solidification defects and develop innovative manufacture techniques.

Semi-solid deformation has been commonly used to describe deformation responses of solidifying alloys. Two of major aspects need to be accounted for when discussing semi-solid deformation: (i) to measure global mechanical responses, also known as rheological properties of semi-solid alloys; (ii) to visualize the microstructural evolution during the processes of semi-solid deformation. In terms of mechanical property measurements, numerous methods have been developed: tensile tests [15,16], compression [17], rotation viscometer [18,19]. Based on these measurements, theories and models were made to predict the global mechanical behavior of semi-solids [20,21]. The microstructural responses of semi-solids upon deformation are extremely complex mainly owing to variables such as solid fraction, grain morphology, and solid-solid interaction coupled with solute-enriched liquid flow. ‘Post-mortem’ microstructural analyses were used which only assessed the final results, not the kinetics and mechanisms. A few in situ experiments were also conducted. Analogous model alloys [22,23], X-ray radiography [15,24], and high speed X-ray tomography [25] have been developed combining various thermal mechanical deformation cells. Among them, high speed X-ray tomography is a relatively new but powerful technique which could provide time-resolved information at a microstructural scale in three dimensions [26]. A semi-solid tensile tester was the first coupled with high speed X-ray tomography, which indeed shed new insights into how damage formed due to granular responses of the semi-solid system [25,27].

This thesis endeavors to facilitate the understanding of semi-solid deformation mechanisms with the aim to refine answers to the following questions: (i) How do the solid grains respond to deformation, do they move, bend or fracture? (ii) How does the solute-rich liquid flow
through a deforming equiaxed microstructure? (iii) How does the strain evolve, does it localize? This will be achieved by developing novel methods to visualize and quantify the microstructural evolution *in situ* via high speed X–ray tomography during various semi-solid deformation processes while measuring the macroscopic mechanical properties simultaneously. The quantified results can also provide experimental benchmarks to validate physically correct models.

### 1.2 Thesis outline

The thesis outline is described as follows:

Chapter 2 provides a theoretic background for this thesis related to solidification and semi-solid deformation. It provides a brief introduction on the theory of solidification and important concepts and models related to semi-solid deformation. It focused on the methods which were used to measure semi-solid mechanical properties, whereas the use of *in situ* experiments (transparent alloys, X-ray radiography and high speed X-ray tomography) observing the microstructural responses during semi-solid deformation were thoroughly discussed. The basics of X-ray tomography and the recent development of high speed X-ray tomography have also been briefly introduced.

In Chapter 3, *in situ* high speed synchrotron tomography was used to visualize uniaxial compression of a semi-solid Al-Cu alloy. This study not only confirmed that semi-solid alloys of equiaxed dendritic structure with a liquid fraction of 30% can be considered as granular materials as the occurrence of dilatancy was observed, but also showed that the semi-solid can be a very complex system.

In Chapter 4, high speed synchrotron X-ray tomography was used to image an indirect extrusion of a semi-solid Al-Cu alloy, focusing on how deformation drives liquid flowing
through and out of the equiaxed microstructure. Using advanced image quantification and image based flow modelling, not only was the amount of liquid exudate and the compaction of the billet measured and quantified, the decrease of the permeability of the porous network during the course of deformation was also determined.

In Chapter 5, high speed synchrotron tomography was used to image the indentation process of a semi-solid Al-Cu alloy. The velocity and strain field in the vicinity of the indenter were directly measured by the use of digital volume correlation, illustrating how different deformation zones (compression, dilation and shear) evolved and localized.

Chapter 6 summarizes the results of the above studies and outlines prospective areas for future investigation.
Chapter 2. Literature review

2.1 Solidification

Many manufactured components include solidification at some stages [1]. For instance, single crystal Ni-based superalloys used for turbine blades are cast by directional solidification techniques [28]. Generally, solidification is the nucleation and subsequent growth of solid crystals. A common example of this is when water freezes to ice. Solidification processes involve numerous physical phenomena such as heat and mass transfer, phase transformation, and fluid flow. The understanding and control of these events help us to invent and improve manufacturing processes.

2.1.1 Nucleation

The nucleation starts when a crystal-like arrangement (a nuclei) form out of randomly distributed atoms in the liquid [1]. The nucleation of crystals mainly depends on two competing forces: (a) the thermal fluctuation which leads to the creation of variously sized embryos, and (b) the creation of a new liquid/solid interface. A homogeneous-formed embryo can form under the condition that the thermal energy provided by cooling the melt overcomes the energy required to create a new liquid/solid interface. The nuclei can also form at preferential locations (e.g. mould wall, impurities) which can substantially reduce the energy barrier to nucleation. This is known as heterogeneous nucleation.

2.1.2 Dendritic growth

After the embryo has formed, a crystal will grow as the melt cools down and its growth will follow a few rules determining its morphology and direction. Firstly, the growth morphology
is determined by the temperature gradient, concentration gradient and growth velocity [29]. When pure metals solidify, one may find a planar or stable solid/liquid interface under constraint conditions (e.g. columnar solidification of pure metals [1]). However, in most cases during solidification when alloying elements are present, the diffusion of elements in the solid is very slow, compared to that in the liquid which lead to the enrichment of solute in the liquid. This can maintain an undercooled region ahead of the interface (constitutive undercooling), in which a planar interface is unstable. Hence most often, branched tree-like structure (dendrites) will form (Figure 2.1) [1].

![Dendritic structure](image)

Figure 2.1 A scanning electron micrograph showing dendritic structure in a Ni-based superalloy, after [30]

When a crystal nucleates, it will grow along its preferred orientation (e.g. <100> for cubic metals like aluminum) but opposite to the direction of heat flow [1]. Therefore, columnar dendrites can form on the condition that the dendrites grow preferably opposite to the heat flow direction along its preferred growth orientation [1,31]. When the dendrites grow unconstrained in an undercooled melt, where the latent heat is transferred radially, the dendrites formed have similar dimensions in all directions. These dendrites are known as ‘equiaxed’.
Here solidification theory is very briefly introduced as a start point for this literature review. Further comprehensive and detailed description of solidification theory can be found in the textbooks by Kurz and Fisher [1] and Dantzig and Rappaz [8].

2.1.3 The origins of deformation in the mushy zone

During solidification, three distinguish states are present: the liquid, semi-solid where liquid and solid coexist, and solid region. In the semi-solid region, the liquid is entrapped between the solid grains. It is also known as the ‘mushy zone’. In this thesis, the mushy zone and the semi-solid are inter-changeable. The mushy zone is a solid-liquid (two phase) region where all of the microstructure characteristics of a solidifying materials are determined [32]. The mushy zone shows strong interplays between the solid and liquid phase during solidification, and presents distinguished properties from the sole liquid or solid state. One of the key differences is its response to deformation. In later sections, the significant influence of deformation on the mushy zone will be demonstrated. While in this section, where the deformation comes from during solidification will be discussed.

The origins of deformation during casting can be categorized into two types: (a) internal deformation inherent to solidification processes, and (b) external deformation (e.g. gravity, dedicated applied deformation and pressure etc.). Internal deformation is mainly due to solidification shrinkage and thermal contraction, which are the most common origination of stress and strain during solidification. The density difference between the liquid and solid phases during solidification leads to the volume shrinkage of the casting components, which is known as solidification shrinkage [33]. The contraction of the solid due to the temperature dependence of the solid phase density can result in thermal contraction [33]. For example, during direct chill (DC) casting, the cooling processes impose a strong thermal gradient on
the ingot, leading to stress and strain accumulation within the materials in the semi-solid state [34].

Dedicated applied deformation, on the other hand, is used in casting processes to: (i) enhance productivity, (ii) reduce defects, and (iii) produce engineering components from the liquid alloy in the shortest route. Depending on the processes used, the casting components can experience different deformation conditions with combined actions of compression, shear and/or tensile forces. Casting techniques include high pressure die casting [12,35], twin-roll casting [7,13] and squeeze casting [36,37]. High pressure die casting serves as a good example. This process injects molten metal into the mould (which consists of thin wall and tortuous cavities) at high velocity and pressure, allowing it to solidify under pressure.

Another set of techniques making good use of external deformation is semi-solid forming or thixo-processing [38]. These combine both casting and forming techniques together by forming the component in the semi-solid state with fine globular grains [39]. Semi-solid forming is an efficient net-shape forming process, which decreases costs by reducing the force applied during forming, and gives superior mechanical properties by reducing the defects. A number of reviews on these techniques can be found in Refs [10,11,40,41].

Although thermal contraction and dedicated applied external deformation are the most common sources to deform the mushy zone, others such as fluid flow, or even gas bubbles can also impose deformation, leading to significant changes of solidification structures.

2.2 Important concepts in semi-solid deformation

The semi-solid state consists of solid phase and liquid phase. Semi-solid deformation requires extensive understanding in fluid flow and solid mechanics. Material properties that are used in solidification, fluid flow and solid mechanics have been applied in semi-solid mechanics.
It is therefore important to introduce these concepts, including solid fraction, permeability and viscosity.

2.2.1 Solid fraction

Solid fraction \( f_s \), packing fraction, packing density or volume density has been used to describe the fraction of solid in liquid-solid mixtures, whereas solid fraction is commonly used to describe the progress of solidification. The solid fraction of a material formed by freezing from liquid state, is a function of temperature and the grain morphology (e.g. size, shape and spacing of the dendrites) [42]. In order to derive this solid fraction evolution, we firstly need to know the phase diagram of the alloy, which usually assumes that the solid/liquid interface were locally in a state of equilibrium. Al-Cu alloy phase diagram in the Al rich corner is shown in Figure 2.2. An equilibrium partition coefficient \( k = C_s / C_l \) and the liquid slope \( (m) \) are defined. In most cases for the theoretical treatment, the solidus and liquidus lines will be assumed to be straight, which means \( k \) (<1, here) and \( m \) are constant. Further assumptions need to be made: (1) the mass transport in the liquid is infinitely quick (no concentration gradient in the liquid); and (2) the mass is balanced between solid and liquid (all the solute which cannot be diffused into the solid must stay in the liquid) [1]. The first case to determine the relationship of solid fraction and temperature is the Lever rule (Eq. 2.1), which assumes that no concentration gradient in the solid and liquid (Figure 2.3 (a)). In practice, equilibrium will only be maintained at the solid/liquid interface, where the compositions agree with the phase diagram. For the second case, provided that a zero concentration gradient in the liquid and limited diffusion in the solid (shown in Figure 2.3 (b)), we can have the equation 2.2.
Figure 2.2 Phase diagram of Al-Cu alloy, after [43].

Figure 2.3 Solidification under equilibrium condition
\[ \frac{C_l}{C_0} = \frac{1}{1 - (1 - k)f_s} \quad (2.1) \]
\[ \frac{C_l}{C_0} = [1 - f_s(1 - 2\alpha k)]^{\frac{k-1}{2\alpha k}} \quad (2.2) \]

Where \( \alpha \) is the dimensionless solid-state back-diffusion parameter, and \( \alpha = D_s t_f / L^2 \), \( L \) is the length of the solidifying system. When \( \alpha = 0 \), we will have Scheil’s equation [44]:

\[ \frac{C_l}{C_0} = (1 - f_s)^{(k-1)} \quad (2.3) \]

Due to the fact that those attempts to model solid fraction involving empirics and a number of assumptions, the prediction of solid fraction still has difficulties in various practical situations, the detail of which will not be studied in this work. Instead, focus will be placed on the usage of solid fraction to categorize the mushy zone in terms of its mechanical response.

Usually, the solidification process is divided into three stages based on the mechanical response and liquid feeding capacity of the mushy zone [45]. (1) at the very early stage of solidification, the nucleated crystals float freely and the macroscopic behavior is close to the liquid [46]; (2) as solid fraction increasing, crystals grow to interact with each other, and shear strength starts to form due to grain entanglement. In the meantime, strain caused by solidification shrinkage can be easily counteracted by the liquid flow and grain arrangement; (3) after that, towards the very end of solidification, the solid dendrites start to coalesce so that: (a) grains can hardly move, (b) tensile forces, resulting from the solidification shrinkage and external strain, can transfer through the grain bridges, and (c) only thin liquid film exits between grains. Liquid feeding then becomes difficult. The deformation cannot be fully compensated by liquid flow and grain movement [46]. The thin liquid film also accounts for the fragile behavior of the mushy zone. So the mush becomes vulnerable to deformation. It is believed that this state is the period during which porosity develops or in the worst case, hot tears form [8].
Dendrite coherency and coalescence point are commonly used to determine different stages of solidification. Dendrite coherency is a term marking the point of crystal impingement where crystals come into mechanical contact [47–50]. It is strongly influenced by grain morphologies so for highly dendritic structures it occurs at a solid fraction of ~ 0.15 [51], and globular grains of ~ 0.55 [52]. The start of coalescence corresponds to the transition between a continuous network of liquid films and a fully coalesced solid skeleton through the formation of inter-granular solid bridges [53,54]. Sometimes, a term called ‘maximum packing solid fraction’ was used by some researchers [55], which marks a transition point when significant shear strength and some tensile strength develop during solidification [14].

### 2.2.2 Permeability

The permeability associated with interdendritic liquid flow plays a significant role in the formation of defects in metal casting processes. The permeability measures the capability of a porous median to transmit fluids. It is a parameter used in Darcy’s law [56]:

\[
\nu = \frac{\kappa \Delta P}{\mu \Delta x}
\]

where \(\nu\) is the fluid flow velocity through the porous network, \(\kappa\) is the permeability, \(\mu\) is the dynamic viscosity of the fluid, and \(\Delta p/\Delta x\) is the pressure gradient in the direction of flow. Darcy’s law is a simple relation to estimate the permeability in a porous media.

The Navier-Strokes equation is also used in numerical modelling of permeability. The general form of the Navier-Strokes equation is [57]:

\[
\rho_l \left( \frac{\partial \nu}{\partial t} + \nu \nabla \nu \right) = - \nabla p + \nabla \cdot T + f
\]

where \(\nu\) is the fluid flow velocity, \(\rho_l\) is the fluid density, \(p\) is the pressure, \(T\) is the deviatoric component of the total stress tensor, \(f\) represents the body forces per unit volume acting on the fluid.
The Kozeny-Carman relationship [56] is used to estimate the permeability of porous medium, which can be expressed as:

\[ \kappa = \frac{(1 - f_s)^3}{K_c S_v^2} \]

where \( f_s \) is the solid fraction, \( S_v \) is the surface area of the solid per unit volume of sample, and \( K_c \) is the Kozeny constant that depends on the characteristics of the porous medium.

Several experimental efforts have been carried out to measure the permeability of the mushy zone at various solid fractions [58–63]. Although the permeability of a semi-solid sample had been measured at a static condition [61] or during isothermal holding at semi-solid temperatures [59,60], it is difficult to apply to determine the permeability evolution during solidification across a range of solid fraction, or when the sample is under external disturbances (e.g. mechanical deformation and magnetic fields).

Another method of permeability determination is using meshed grain structures to run numerical flow simulations. The grain structure can be obtained by the construction of 3D grain geometry with X-ray tomography [64–68] or serial-section techniques [69]. Then the 3D meshed geometry was used as an input to a dynamic fluid flow simulator, usually to solve the Navier-Stokes equation (Eq. 2.5). This method can be effectively used to measure the permeability evolution as a function of solid fraction and grain structures. For instance, Puncreobutr et al [66] combined in situ 3D tomographic datasets with a numerical flow model simulating the permeability of Al-Si-Cu alloys with and without Fe additions. The study showed how the growth of Fe-rich intermetallic can significantly modulate the flow behavior of the solidifying dendritic grains. This method can potentially be used to study the effect of deformation on the permeability of solidifying structure although studies have not been carried out yet.
2.2.3 Viscosity

Viscosity (µ) is a concept in fluid mechanics which measures the resistance of fluid or slurries to gradual deformation by shear or tensile stress [57]. Viscosity is an important properties of semi-solid metallic alloys, and determines the required force for deformation and flow of semi-solid materials [70].

The fluid is categorized as Newtonian fluid or non-Newtonian fluid based on its viscosity. A Newtonian fluid’s viscosity is independent of the shear stress/rate. In another word, the viscosity is a constant. Newton’s fluid law for simple shear [57] is defined as:

\[ \tau = \mu \left( \frac{\partial V}{\partial y} \right) = \mu \dot{\gamma} \]  \hspace{1cm} (2.7)

\( \mu \) is the dynamic viscosity of the fluid, \( \tau \) is the shear stress, and \( V \) is the momentum velocity, \( \dot{\gamma} \) is the shear rate.

For non-Newtonian fluids, including shear thickening and shear thinning, the viscosity is dependent on shear rate history. The viscosity of a shear-thickening liquid increases as the shear strain rate increases, while that of a shear-thinning liquid decreases with rising shear strain rate. The use of the terms (thixotropic and pseudoplastic) has been commonly seen in literatures involving semi-solid deformation [71]. It is important to clarify its definition here. A thixotropic material is one whose flow properties are time-dependent. That is, the viscosity or shear stress required for flow of a thixotropic material depends on its past history [72]. A pseudoplastic material is one whose apparent viscosity depends upon and decreases with shear rate, which is basically shear-thinning.

It is important to note here, that the fluid viscosity of the liquid in semi-solid system is significantly different from the viscosity of the semi-solid mixture. Molten metals behave like Newtonian fluids. The pure Aluminum liquid at its melting point has a viscosity of 1.0–1.4
mPa·s. Although the viscosity of semi-solid Al alloys varied with liquid fractions and shear rates, it is much larger than the pure liquid. For instance, the semi-solid A356 Al-Si with volume solid fraction of 35% has a viscosity of $10^7$ to $10^8$ Pa s [73].

### 2.3 Effect of deformation on solidification microstructure and defects

Casting microstructure and defects determine the performance of final solidified products. Grain morphology ranging from globular, cellular to complex tree-like structures have been found in castings, which determines mechanical properties of final components [1]. In addition, typical defects, such as porosity, hot tearing, and segregation, are known to occur during solidification and are yet to be fully addressed. Those defects are all affected by heat and mass transfer during solidification, but in some way, they are also controlled by the imposed deformation. In this section, we will focus on the effect of external deformation on the grain structure and defect formation.

#### 2.3.1 Grain structure

Conventional solidification usually produces various sizes of dendrites [8]. Applying deformation to the solidifying alloys is known to change the grain morphology [9]. For example, dendrite arms were observed to bend due to gravity [74]. Under strong shearing of partial solidifying alloys, the morphology of the primary phase transformed from dendrites to globular [18] (Figure 2.4). This process of producing globular grains via mechanical stirring is referred to as vigorous agitation, which has been attributed towards a transition from dendritic to ‘rosette’; then spheroidal as a result of ripening, shear and abrasion with other grains (illustrated in Figure 2.5) [9]. However, the fragmentation of the secondary dendrite arms during solidification has also been considered as a possible mechanism [75,76]. As
shown in Figure 2.6, the dendrites when extensively sheared could break up into small pieces. Those pieces of grains can be the nuclei of new grains evolving to globular-shape. Nevertheless, there is still a paucity of experimental evidence to support any of the two mechanisms.

Figure 2.4 A quenched structure after continuously shearing by rotation speed 500 tpm, after [9].

Figure 2.5 Development of globular grain during solidification with vigorous agitation: (a) initial dendritic fragment (nucleation), (b) dendritic growth, (c) rosette structure, (d) ripened rosette, and (e) spheroid, after [9].
Figure 2.6 Schematic illustration of the evolution of structure during solidification under mechanical stirring: (a) initial nucleation, (b) dendritic growth, (c) breakdown of dendritic network to form new nuclei due to vigorous agitation, (d) the emergence of globular structure (modified from [76]).

2.3.2 Segregation

As far as normal solidification conditions are concerned, local equilibrium is assumed to hold at the solid/liquid interface [1]. Then at the interface, compositional differences will always lead to compositional non-uniformity in the solidified alloy, which is known as segregation [1]. When the variation of solute composition is at the scale of a casting component, it is known as macrosegregation, for example, the centerline segregation of a continuously cast ingot [77]. On the other hand, the localization of segregation at the micro scales, typically of the secondary arm spacing (10 μm–100 μm), is termed as microsegregation [1,78]. Segregation has been well reviewed in several books [1,8,79] and a few papers [77,80].

One important driving force for segregation is the deformation either occurring naturally or imposed externally during solidification. The deformation could cause serious segregation or even solid/liquid separation. And in turn, this segregation can have marked effects on the local and global mechanical properties in the final components. For instance, the positive centerline macrosegregation commonly observed in continuous casting of steels is believed to be associated with deformation. In the process of continuous casting, the deformation (owing to thermal stress and bulging between support rolls) draws solute-enriched liquid into the slab center, which gives rise to centerline macrosegregation [81,82]. Another example is the exudation formed in direct chill castings. The exudate layer results from transportation of solute rich liquid through the coherent mushy zone into the air gap between the mould and the casting component [83] (Figure 2.7(a)). Deformation induced segregation can also be found in twin roll casted sheets [7]. In the twin roll casted sheets, common but detrimental
deformation-induced segregation includes surface bleed (a pool of solute-rich materials), and segregation band with mixed grain morphology dispensed with low melting materials (Figure 2.7 (b)), limits the commercial exploitation of twin roll casting techniques [7,84].

The complex combination of solidification and accumulation of internal and external deformation indeed could result in various forms of severe segregation patterns which are rarely encountered in normal casting practices. The underlying mechanisms, however, have not been clearly understood, mainly due to the inadequate assessment of the link between the change of complex two phase microstructures and the imposed deformation.

Figure 2.7 Micrograph of (a) surface exudates (the exudate thickness varies from ~0.2 to 1 mm), after [83], (b) a segregation band, after [7].

2.3.3 Porosity

Microporosity formed during solidification is of pronounced importance because it can reduce the ductility and fatigue life of metallic engineering components. Considerable efforts have been attempted both experimentally and numerically to study the origination of microporosity and its effect on mechanical properties of components [85]. Comprehensive reviews of porosity formation as a result of gas and shrinkage can be found in Refs [8,85,86]. The formation of microporosity during solidification is generally attributed to two factors: shrinkage and evolution of gas [85,86]. If the liquid feeding is blocked in the semi-solid region and not able to compensate the liquid/solid volume change, the pressure in this region
is reduced locally, forming numerous small pores [85,87]. These pores are often irregular-shaped and are known as shrinkage porosity [85]. Due to the fact that gases are far less soluble in solid than in liquid metals, as solidification progresses, the concentration of dissolved gases in the liquid phase increases until gas bubbles are formed, hence forming ‘gas porosity’ in the final casting components [87]. In Al alloys, dissolved Hydrogen is also shown to diffuse into entrapped oxide bi-films, leading to the formation of Hydrogen porosity in the solid cast [86,88].

Deformation can also induce porosity, although it is difficult to be distinguished it from shrinkage porosity in the solid casts as it is also irregular-shaped. The effect of external pressure on pore formation can be explained by Eq. 2.8 [12]. When the sum of the gas pressure and the solidification shrinkage is greater than the combined effect of applied external pressure and gas/liquid surface tension, a pore will form out of the melt [12].

\[ P_g + P_s \geq P_{ext} + P_{s-l} \] \hspace{2cm} (2.8)

where \( P_g \) is the equilibrium pressure of dissolved gas, \( P_s \) is the pressure drop due to solidification shrinkage, \( P_{ext} \) is the applied external pressure, and \( P_{s-l} \) is the pressure due to pore-liquid surface tension. Therefore, when the radius, \( r \), of a pore nucleus exceeds a critical radius, \( r_c \) [12]:

\[ r \geq r_c = \frac{2\gamma}{P_g + P_s - P_{ext}} \] \hspace{2cm} (2.9)

where \( \gamma \) is the surface tension and:

\[ P_{s-l} = \frac{2\gamma}{R} \] \hspace{2cm} (2.10)

Eq. 2.9 can be used to demonstrate the observation that the amount of porosity decreases with the increase in the applied external pressure during high pressure die casting [12]. However,
it has limited application to the formation of porosity under complex deformation conditions. A localized band of porosity had been found within high pressure die casted components [35], which could not be explained by Eq. 2.9. It is believed that the deformation imposed on the solidifying structure can initiate strain localization and dilatancy which leads to such a band of porosity (Figure 2.8) [6]. It is observed that the porosity band is normally accompanied with a band of segregation in the products of high pressure die cast [35], and twin roll casted components (Figure 2.7b) [7].

![Figure 2.8 Micrograph showing a band of porosity, after [6].](image)

### 2.3.4 Hot tearing

Hot tearing is a very catastrophic casting defect. If it occurs in a cast or a weld, the components are usually rendered unusable. It is also extremely challenging to predict and prevent. The desires to obtain a comprehensive understanding of this phenomenon are associated with the aim of helping improve casting technologies. A large number of reviews (see Refs [8,86,89]) can be found in this topic.

Generally, the formation of hot tearing can be attributed to: (a) lack of liquid feeding in the mushy zone, and (b) shear or tensile stresses transferred to the dendritic network [3,8,86,90]. It forms at the last stage of solidification with high solid fraction (above 85% - 90 %) [45]. As discussed in section 2.2.1, at this stage, the solid network is capable of transmitting stress, and the mushy zone is vulnerable to deformation. The stress is mainly due to changes of
internal deformation condition upon cooling. The stresses can pull these dendrite arms apart quite easily [90], as the thin liquid films present between dendrites prohibit the formation of strong bonds between solid grains. Deep into the mushy zone where the permeability is quite low, hot tears would form on the condition that the liquid flow could not compensate the opening of the dendritic network induced by the tensile stress [90]. It is also well established that hot tearing propagates along the inter-dendritic liquid film, resulting in a bumpy fracture surface [8,45,86].

As described above, hot tearing is a defect that has been strongly linked to the deformation behavior of the mushy zone, especially the tensile stress at the last stage of solidification. The connection has been extensive investigated experimentally for some time and it will continue with the help of emerging techniques. It is more important to predict at which conditions (e.g. composition and casting process parameters) hot tearing tends to occur. A number of models and criteria have been developed in this respect, including solidification interval [86], accumulated stress [21], strain [91], strain rate [90], liquid pressure drop [92]. However, those models are restricted to predict its formation over a range of casting conditions. The thermal history, shrinkage, constitutive behavior of semi-solids and the liquid flow resistance of the solidifying structure are all required to be considered to develop a robust hot tearing criterion.

2.4 Measurement methods of semi-solid mechanical properties

Prior studies have quantified the mechanical behavior of semi-solid alloys through a range of mechanical tests, including rotation viscometer [18,49,50,93], compression [19,94,95], tensile loading [96–99], direct shear [100], extrusion [101] and indentation [102,103]. Usually, two varieties of semi-solids can be applied: (I) those continuously cooled from liquid state; and (II)
those isothermally held after partial remelting or partial solidification. These mechanical tests are described in this section as follows.

2.4.1 Rotation viscometer

Rotation viscometer is commonly used to measure viscosity of slurries. It has been adapted to determine the viscosity of semi-solid metallic alloys [6,18,49,93,104–108]. This method is more suitable for low viscosity and hence low solid fraction (e.g. \( f_s < 50\% \)). The key to rotation viscometer is the construction of an inner cylinder (or a fan) and an outer cylindrical cup. The semi-solid sample is contained in the annulus between the two cylinders. Either the inner cylinder rotate and keep the outer cup stationary, or the outer cup rotate but hold the inner cylinder stationary. The torque is measured from the rotating part. Two different rotation modes can also be used to determine the time-dependent viscosity: (a) constant shear speed and (b) shear rate jump. This instrument has been employed to measure viscosity of various semi-solid alloys (e.g. Sn-Pb alloys [18,106], Al-Cu-Mg alloy[104], SiC reinforced Al composite [107,109]) since the 1970s.

The usage of a rotation viscometer has found a number of interesting results: (1) the viscosity of a semi-solid metallic alloy is dependent on the solid fraction and decreases with increasing shear rate (shear-thinning), similar to colloidal suspensions [18]; (2) the viscosity of a semi-solid alloy is of the same order of magnitude as that of a suspension of interacting particles and higher than that of a suspension of non-interacting particles [105]; (3) the viscosity is dependent on the morphology of the solid grains, e.g. the dendritic structure shows higher resistance to shear than globular structure [38,106]; (4) shearing dendritic structure at semi-solid region could bend or even break up the dendrite arms, as observed from the final deformed and solidified samples [106]; (5) the viscous response of semi-solid alloys can be
shear-thickening under conditions when the internal structure of the mushy zone was stationary[110].

Rotation viscosity has also been used to investigate the dendritic coherency point (discussed in section 2.2.1) [51,111], which is based on the fact that the viscosity of semi-solid alloys would increase significantly when the solid fraction is increased over the dendritic coherency point. For instance, Dahle and Arnberg [47] used a four-vane rotation viscometer to clarify the strength development of the mushy zone of various alloys (Al-7wt%Si-Mg, Al-11wt%Si-Mg, Al-4wt%Cu, and Al-5wt%Mg).

2.4.2 Parallel plate compression

Parallel plate compression has been extensively utilized due to its ease of implementation and close resemblance to many key industrial processes [17,94,112–118]. Properties such as the yield stress and viscosity of a semi-solid alloy can be directly measured. Compression tests are suitable for higher solid fraction (>50%). Two types of compression can be used: (I) the semi-solid sample is squeezed at a constant load and the strain versus time is measured; and (II) the sample is compressed at a constant displacement rate while the load vs. time/strain is measured.

The strain-time curve determined by Mode (I) semi-solid compression can be further mathematically treated to resolve the viscosity of the tested semi-solid alloys, as shown in [73]. The effects of grain morphology [17], grain refiner [73], pouring temperature for the bullets [119] on the rheological response of semi-solid alloys have been investigated. Different grain structures (equiaxed, columnar dendritic, and globular) present distinguished strain-time curves as well as different level of damage in the final deformed samples [17]. Non-dendritic structures show no sign of cracking during compression while dendritic structure is shown to be more susceptible to damage formation [17]. Samples which are grain
refined with fine globular grains are easy to flow and deform, thus have lower apparent viscosity than its counterpart without grain refinement [73].

Mode (II) compression can be used to measure the yield stress of semi-solids, and has been used to assess the effect of strain rate [19], sample-piston friction [120], solid fraction [120] on global mechanical properties. Unsurprisingly, higher strain rates show higher yield stress [19]. Breakdown of dendrite network and serious segregation of liquid from the semi-solid were observed in the deformed samples [19]. Pinsky et al [120] studied the effect of solid fraction and friction condition between the sample and compression piston. The maximum stress decreased with the increase in liquid fraction [120]. Higher friction between the sample and piston leads to a higher degree of liquid segregation.

2.4.3 Tensile test

Semi-solid tensile test is a commonly-used deformation method in solidification, as it can be used to determine the hot tearing tendency of metallic alloys [16,34,45,98,121], and thus can aid in predicting solidification defects. The tensile tests are carried out at high solid fraction (e.g. >90%). The stress-strain curves of semi-solid tensile tests are very scattered due to the fact that various sample geometries, testing methods, experimental conditions were used by different research groups. A thorough review of semi-solid tensile properties of Al alloys can be found in Refs [45,79]. Semi-solid tensile tests have also been used in steels [122], Ni-based superalloys [123] and Mg alloys [124]. One interesting example is described below.

Phillion et al. [96] observed damage and strain evolution using X-ray tomography during semi-solid tensile loading of DC cast AA5182 alloy at 528 °C (solid fraction about 0.98 %). The results indicated that porosity initiation and coalescence play an important role in the fracture behavior of semi-solid deformation. In addition, hot isostatically pressed (HIP) samples were compared with as-received ones, showing that HIP increased the semi-solid
tensile stress and ductility, as it reduced the volume fraction of porosity [96]. This study, although used an interrupted *in situ* experiment (the tensile deformation was stopped at certain strains, then the deformed sample was cooled down and scanned by X-ray tomography), shows the key advantages of X-ray tomography (non-destructive nature and three dimensional visualization). X-tomography will be introduced in section 2.5.3.

![Figure 2.9](image)

*Figure 2.9 Evolution of damage and porosity during semi solid tensile deformation, after [53].*

### 2.4.4 Indentation

Indentation can be potentially used as an in-line quality control tool for semi-solid processing as it is an easy method for simulating the mechanical properties of semi-solid alloys. This method can produce highly localized deformation around the indenter which has been demonstrated where ductile metals were indented at room temperature [125]. Bigot *et al.* [102] proposed an indentation test to determine the mechanical behavior of semi-solid globular and dendritic alloys. It is found that a globular semi-solid Sn-15%Pb alloys needs less stress to deform than a dendritic one. Very high peak load (500-700 N) was measured during their indentation tests. Ferrante and Freitas [103] also used the indentation to measure the force versus penetration curves of various grain structures at a solid fraction of 86%. Although only two publications are available in the public domain [102,103], it still shows that indentation presents great potential to become a standard method to probe semi-solid mechanical properties, e.g. viscosity.
2.4.5 Other methods

Although rotation viscometers, compression and tensile tests have been widely used to measure the semi-solid alloys’ global mechanical properties, researchers developed a number of other deformation methods, for example, direct shear [100] and extrusion [101], which have not been widely applied as yet. A direct shear device (Figure 2.10(a)) was designed to study the shear behavior of semi-solid Al-Si-Cu alloys by Sumitomo et al. [100]. The shear strength as a function of solid fraction and alloy composition was determined as shown in Figure 2.10(b), which indicated two transition points, namely, coherency and maximum packing solid fraction. The shear strength increased rapidly after the solid fraction reached the maximum packing solid fraction. The result also showed that the coherency point and maximum packing solid fraction are related to alloy compositions.

Figure 2.10: (a) Schematic of the direct shear rig, and (b) shear strength versus solid fraction for AlSi₄Cu₁, AlSi₇Cu₁, AlSi₄Cu₄ and AlSi₇Cu₄ after [100].
Ludwig et al. [126] used several different experimental set-ups for testing aluminum alloys during solidification, with the aim to study the rheological behavior of different semi-solid alloys, e.g. shear and tensile devices and a drained compression setup (see Figure 2.11). The rheological behavior of Al-Cu alloys [126] and 5182 alloy [127] has been carried out at constant temperatures and different stain rates. Drained compression is essentially an extrusion process which can drive the liquid to flow out of the mushy zone (Figure 2.11(b)). Gebelin et al [115] also used a variety of deformation methods (compression, extrusion tension and filtration) to measure the viscosity of an AZ91 Mg alloy.

![Figure 2.11](image.png)

Figure 2.11: Schematic diagram of the (a) translation shear apparatus, (b) drained compression device, and (c) tensile experimental set, after [126].

The measurements of semi-solid mechanical properties through different deformation methods are essential and have been used to develop and verify continuum constitutive models (which will be discussed in section 2.6.1). However, microstructural analysis carried out on the deformed samples (‘post-mortem’) has failed to simultaneously link the mechanical properties with microstructural changes and failure mechanisms, thus these studies were not able to strongly support semi-solid deformation models. To facilitate this link, in situ observations of semi-solid deformation coupling with measurements of global mechanical properties are necessary.
2.5 In situ studies of semi-solid deformation

The aforementioned experimental observations of microstructural changes were conducted usually using 2D metallographic sections acquired after deformation (‘post-mortem’). Although the post-mortem microstructure analyzes have contributed greatly to understand the problem, it is still far from satisfactory. Without ‘visualizing’ the complex interaction of various phases during semi-solid deformation, the assumptions for which the microstructural models were based cannot be validated. This can only be realized through in situ experiments. However, in situ imaging of solidification microstructure in metallic alloys is extremely difficult because of the opaque nature of metal samples and the need for precise control of the temperature. A range of techniques from transparent organic materials [22], to X-ray imaging methods [15] combining mechanical loading set-ups have been used to this end and are reviewed in this section.

2.5.1 Analogous transparent organic materials

A major breakthrough in the studies of solidification was the introduction of transparent organic alloys (e.g. Succinonitrile (SCN)) in the 1960s by Jackson and Hunt [128]. Since then, SCN has been broadly applied in real-time observations of solidification by optical microscopy, owing to it being transparent to visible light, as well as its similarity to metallic alloys when solidifying [129–132]. It has been mostly used to study the morphology of solidifying structure under a constrained temperature gradient (directional solidification) [129,132]. One of the examples is shown in Figure 2.12 [132]. There are also a few studies visualizing the influence of deformation on the mushy zone structure [22,23].
Figure 2.12 Steady-state dendritic patterns formed at the crystal-melt interface of a binary succinonitrile alloy during directional solidification, after [132].

Farup and Rappaz [22] used succinonitrile-acetone (SCN-ACE) alloy solidifying in a directional solidification cell (Figure 2.13). A wedge was forced into interdendritic liquid region during solidification (Figure 2.13). The experimental results show fundamental insights into hot tear formation mechanisms. It was found that hot cracks always opened at grain boundaries, as illustrated in Figure 2.14. The results highlight that at lower solid fraction, the opening was healed by interdendritic liquid. At higher solid fraction, hot tears nucleated from pre-existing pores or directly formed in the interdendritic liquid.

Figure 2.13: Schematic drawing of the experimental set-up in bottom view, after [22].
Figure 2.14: Sequence showing a dendritic network opening induced by pulling the columnar dendrites with a pulling stick (‘A’), after [22].

Shen and Beckermann [23] constructed a test cell with the use of SCN-ACE alloy, where the directionally solidifying columnar dendritic mushy zone was compressed by a cooling plate towards the growth direction of dendrites. Deformation has been shown to affect fluid flow significantly.

The use of transparent alloys has provided benchmark data to advance modelling. However, the organic systems cannot be completely analogues to metals and alloys. Firstly, transparent alloys are not completely identical to metallic systems in terms of most physical properties determining the solidification processes (e.g. thermal conductivities, freezing temperature and partition coefficient etc.) [133,134]. In addition, transparent alloys may have different mechanical properties from their metallic counterparts, when deformed within the semi-solid region.
2.5.2 X-ray radiography

Another technique which has been used for many decades for in situ observations of the solidification is X-ray radiography [87,133,135–137]. This method can achieve high spatiotemporal resolutions, resulting in time-resolved 2D imaging. In addition, the solid/liquid interface can be reliably resolved due to high spatial resolution, providing that the alloy system under investigation comprises segregation with good X-ray transmission contrast. It has been recently combined with thermo-mechanical loading methods to study microscopic responses during semi-solid deformation [15,138,139]. Phillion and Lee et al. [15] designed a tensile tester combined with synchrotron X-ray radiography at Diamond Light Source, UK (see Figure 2.15), which allow real-time observation of semi-solid tensile deformation. Three stages for the damage formation have been confirmed by their study: (1) strain localization, (2) damage nucleation and growth, and (3) final rupture, as illustrated in Figure 2.16. In addition, load and strain relationship at various temperatures was measured simultaneously during X-ray imaging in their study.

Figure 2.15 The semi-solid tensile tester, after [15].
Figure 2.16 Upper: Sequences showing the void evolution during semi-solid tensile; Lower: schematic showing three stages during semi-solid deformation, after [15].

Gourlay and co-workers [24,138,140–143] designed a direct-shear cell which was coupled with radiography at the Spring 8 synchrotron, Japan (Figure 2.17). Al-15wt%Cu globular grains with 48% solid volume fraction and dendritic grains with 30% solid volume fraction were both tested [138]. In addition, a semi-solid (various solid fractions) carbon steel (Figure 2.18) with globular grains was also used [140,143]. Their experiments were dedicated to clarify that semi-solid alloys can be considered as granular materials. They observed the rearrangement of grains like quasi-rigid particles resulting in dilation during shear.

Figure 2.17 Schematic the semi-solid shear cell, after [138,141].
Figure 2.18 Radiograph of direct shearing a semi-solid carbon steel, after [144].

Zabler et al [145] developed a rig which injected a semi-solid globular Al-Ge alloy through a thin channel while imaging the process with high speed radiography at ID 15A at European Synchrotron Radiation Facility (ESRF). They studied the flow behaviour of solid particles and particle clusters conjointly with the liquid flowing through these thin channel geometries.

X-ray radiography combining with semi-solid deformation could deliver dynamic information on materials flow and damage formation. However, the result may not represent 3D bulk behaviour and properties. Firstly, the usage of thin sheet samples (usually 200-400 µm) can cause superficial artefacts. Second, the strength and weight of a thin section of dendrite can be much lower than those of a whole dendrite, which means thin-section dendrites are easy to break-up and flow when they are mechanically deformed.

2.5.3 High speed X-ray tomography

It is the earnest wish of material scientists to acquire real time information of microstructure and damage evolution during materials processing in three dimensions. X-ray tomography is a non-destructive, three dimensional characterization method that has been applied in a number of fields [146–151]. It has been improved tremendously with the advancements in X-ray source and detectors over the last few decades [26]. More importantly, recently, researchers are combining heating furnace, and/or mechanical testing rig with X-ray tomography to perform in situ experiments. In this section, an introduction to (high speed) X-
ray tomography will first be presented and then a few applications of high speed X-ray tomography to study solidification and semi-solid deformation will be reviewed.

X-ray attenuation describes the proportion of X-rays scattered or absorbed as they pass through the sample, which primarily relates to X-ray energy, the density and atomic number of the sample. In the case of a monochromatic X-ray source, if N photons enter the material, then the number of photons exiting, I, is given as a function of the incident intensity $I_0$, following by the Beer-Lambert Law:

$$I = I_0 e^{-\mu_0 x}$$  \hspace{1cm} (2.11)

where $\mu_0$ is the attenuation coefficient of a homogeneous material.

The resultant grey level images (named as radiographies or projections) are then used to reconstruct a 3D representation of the structure, usually using a filtered back-projection method [152]. Two different sources are generally used to perform X-ray tomography, namely, laboratory based source (divergent/cone beam) and synchrotron source (parallel beam).

Synchrotron sources deliver many benefits over laboratory based X-ray sources and these include:

1. A very high flux at small beam size which leads to higher signal-to-noise ratio in a short time interval.
2. A (partially) coherent beam can be set up for phase contrast and holo-tomography.
3. The high energy beamlines at the third generation synchrotron sources, for instance, I12 at Diamond Light Source (DLS) [153] and ID19 at European Synchrotron Radiation Facility (ESRF) [154], allow high density materials with engineering applications, such as Ni-based superalloys and steels to be imaged.
4. The development of third generation X-ray source enable the capture of 3D images in high spatial (<50 nm) and/or temporal resolution (<1s). Commonly, X-ray tomography with high temporal resolution is referred as high speed (fast or ultra-fast) X-ray tomography.

Recent years have seen a rapid increase in the development and usage of high speed synchrotron X-ray tomography. Here the high speed is referred to as collecting a tomography at an acquisition time of seconds (commonly less than 30 s but typically less than 10 s). One of the important aspects of high speed X-ray tomography is its capacity to collect time-resolved (or time lapse/real time) 3D information. This requires that the structural changes of the sample during the acquisition time of the tomography should keep at a minimum level [155]. Otherwise, the reconstructed tomography can end up with motion blur (movement artefacts).

The first publication using high speed X-ray tomography was in 2005, where a solidification sequence of an Al-Cu alloy has been visualized in situ [156]. Nowadays, high speed X-ray tomography has been routinely available at a few synchrotron sources (Figure 2.19). The temporal resolution can even achieve at less than 1 s. With more than a decade of development, substantial progress has been achieved as a result of continual advancement of X-ray imaging techniques, together with the development of custom-build environmental cells. It has been proved to be a very promising tool and has been used to study a range of complex microscopic interactions, for instance, solidification and coarsening of melt metallic alloys [66,156–160], sintering of metallic powders [161] and fluid flow through rocks [162].
High speed X-ray tomography has been coupled with a few semi-solid deformation tests before and during undertaking of this thesis. Semi-solid tensile tester was the first coupled with high speed X-ray tomography by Terzi et al [25,27]. This study showed the complex interactions of the three phase structure (globular-like solid grains, solute-enriched liquid, and gas porosity). The accumulation of liquid at the strained region and formation of cracks had been observed in situ (Figure 2.20). A semi-solid extrusion experiment has been performed combining high speed X-ray tomography at beamline I12 in Diamond Light Source [163]. It is identified that with suitable choice of grain microstructure (coarse globules) with respect to the X-ray imaging system, it is possible to process the data to assess the grain movement, and its quantitative evolution. The two studies both used a microstructure with a few large globular-like grains across the sample. Whether naturally formed dendrites behave in the same manner or not poses an open question. In addition, how the semi-solid alloy behaves in different deformation conditions is not clear and has not been directly examined. It is

Figure 2.19 The comparison of high speed X-ray tomography, after [26].
recommended to use complex dendritic structures and complex deformation conditions for further studies.

Figure 2.20 Cross-section slices from X-ray 3D volumes showing the semi-solid tensile deformation at (a) \( t = 496 \) s, (b) \( t = 729 \) s and (c) \( t = 1215 \) s, after [27].

In conclusion, the results from the in situ experiments have facilitated new insights into semi-solid mechanics at a microstructural level. The non-destructive nature and fast acquisition of high speed X-ray tomography is a unique and powerful tool, overcoming some limitations of X-ray radiography and analogous organic materials. The importance of semi-solid deformation processes therefore demands considerably more attention and more use of high speed X-ray tomography to further clarify semi-solid deformation mechanisms.

2.6 Semi-solid deformation models

Although this thesis focuses on uncovering semi-solid deformation mechanisms experimentally, it is of vital importance to introduce the prevailing theories and models that are used to describe semi-solid deformation. It is hoped that the experimental results obtained in this thesis can be used to prove or disprove those proposed theories, as well as provide validation cases for the further development of advanced models.
2.6.1 Continuum deformation models

There are a variety of continuum models to characterize the constitutive responses exhibited by semi-solid alloys. Most of them originate from empirical relations fitting with experimentally measured global mechanical properties.

A classic power law model (i.e. Eq. 2.12) has been widely applied to describe the rheology of solid-liquid slurries [17,105].

\[ \mu = m \dot{\gamma}^{(n-1)} \]  

where \( \mu \) is the viscosity, \( \dot{\gamma} \) is shear rate and the values of the coefficients \( m \) and \( n \) can be derived experimentally and is believed to be a function solid fraction. For example, Laxmanan and Flemings [17] established a relationship between viscosity, shear rate and solid fraction in the range of 0.15 to 0.6, for a semi-solid globular Sn-15wt.%Cu alloy (Eq. 2.13).

\[ \log \mu = 1.02 + 8.94 f_s + 1.78 f_s \log \dot{\gamma} - 1.39 \log \dot{\gamma} \]  

Kumar et al. [106,110] developed an internal variable model combining the macroscopic constitutive behavior with the microstructure kinetics. They categorized the factors influencing the semi-solid constitutive behavior into two subsets: (I) internal variables including the level of agglomeration, fluid phase viscosity and particle properties; (II) external variables, such as temperature and shear rate. Two sets of relations representing the flow behavior of the semi-solid and the evolution of the internal variables were established.

The flow equation:

\[ \tau = \dot{f}(\dot{\gamma}, T, s_{1...k}) \]  

And the internal variable equation:
\[
\frac{d s_p}{dt} = \hat{\gamma}_p(\dot{\gamma}, T, s_{1...k}), \quad 1 \leq k \geq p
\]

\(s_{1...k}\) is the internal variables. Although there are quite a few internal variables, Kamar et al. [110] assumed that the degree of agglomeration plays a vital role, hence they chose only to consider this factor. Their calculation fits reasonably well with the measured semi-solid properties.

Some others have considered semi-solid alloys as porous solid networks saturated with liquid [20,126,164,165]. This theory is based on the assumption that the solid grains are connected (formation of bridges/bonds between grains), which means it may be only applicable at high solid fraction. The solid region is mostly described by visco-plastic constitutive equations, while the liquid is governed by Darcy’s law [20,126,164,165]. However, Favier and Atkinson [112] argued that pure visco-plastic models are not adequate and proposed that the solid phase can be regarded as an elastic porous skeleton saturated with liquid, which indeed showed better prediction of the global mechanical behaviour for semi-solid alloys of high solid fraction.

Semi-solid alloys have also been treated as a suspension which means the solid particles are of low cohesion and dispersed in a liquid matrix [166–169]. Based on this consideration, Chen and Fan [166] derived a model describing the rheological properties of semi-solid alloys under a simple shearing flow condition. Through the so-called effective solid fraction (\(\phi_{eff}\)) which takes the degree and evolution of agglomeration into account, the viscosity and shear stress can then be predicted (e.g. Eq. 2.16).

\[
\mu = \mu_0 (1 - \phi_{eff})^{-5/2}
\]

\[
\phi_e = (1 + \frac{n-1}{n} A)f_s
\]
where $A$ is a parameter related to the packing mode, and $n$ is a structure parameter defined by the average number of grains in the agglomerate.

Zavaliangos [170] claimed that the solid phase in the mushy zone has two distinct limits: (a) a fully cohesive porous solid and (b) a cohesionless granular material. He introduced the degree of cohesion interpolating between these two limits. The cohesion depends on the wetting of the contacts between solid particles, which appears a rather difficult parameter to determine experimentally. Albeit that this model only provides a general description of semi-solid mechanical properties, they argue that semi-solid can be considered as either cohesive porous median, cohesionless granular or a state in between depending on the conditions of semisolid itself and imposed deformation. This concept provides a possible mechanism to understand the changes of microstructure during semi-solid deformation, e.g. (a) the destruction of bridges between grains due to shearing, and (b) the increased level of cohesion when adjacent grains move to a relatively favorable orientation [170].

In conclusion, various constitutive equations have been developed based on different assumptions, with a qualitative match with experimentally determined stress/strain or rheological curves. Most of those assumptions based on assumptions of microstructural level e.g. agglomerate/deagglomerate, and grain-grain bonds, which are yet to be validated by experiments.

### 2.6.2 Granular deformation models

Although continuum deformation models could be used to predict the macroscopic mechanical properties, they cannot resolve most of the phenomena occurring at the microstructure scale, e.g. grain to grain interaction, liquid flow, and strain localization. Recent years have seen a few attempts to incorporate grain to grain interaction and/or liquid flow into semi-solid deformation simulations [3,95–98].
Phillion et al [142] developed a three phase simulation framework with direct finite-element techniques: a 2D solid grain based on Voronoi tessellation, liquid film surrounding the grains, and globular pores decorated at liquid channels. They treated the solid phases as elasto-plastic materials and liquid phase as a perfectly plastic material with low yield stress. Although these assumptions might not be physically correct, these simulations still showed how microstructures (grain size and void fraction) can influence the macroscopic constitutive behavior. Rapaz and co-workers [27,92,171,173] proposed a 3D hydromechanical granular semi-solid deformation model incorporating solid phase deformation, liquid flow and failure mechanism. The mechanical behavior of the solid phase was assumed to be elasto-viscoplastic, while the liquid was modeled as a simple Poiseuille flow. The failure/hot tearing started to initiate when the liquid pressure reached a critical value. The simulated results from this model also compared with an in situ X-ray tomographic observation of the tensile test [25] with excellent correlations in both the prediction of damage formation and liquid flow tendency. Yuan et al [172] used a discrete element method (DEM) to model grain rearrangement during semi-solid direct-shearing of equiaxed and globular Al alloys. DEM is widely used to simulate granular deformation with a wide range of applications [174–176]. The grains generated by a solidification model [177] were mapped by circular disks, then integrated into a DEM package to run the deformation simulation. Those grains were considered as cohesionless rigid bodies where viscoplastic deformation and liquid flow were ignored in their model. Using the model, the influence of grain morphology and solid fraction on dendrite coherency was systematically studied.

The intricate models of direct finite element [172], discrete element [178] and a combined finite element/discrete element method [92,173], proved to be well suited to simulate semi-solid deformation at a small scale (tens or hundreds of grains). It is anticipated that DEM
based solidification models at a scale of a cast can be developed incorporating the effect of deformation.

### 2.6.3 Grain-scale deformation models

As discussed in section 2.3.1, the deformation imposed on the mushy zone can lead to significant modulation of grain structure. It is therefore of importance to simulate the deformation response of solid grains at the scale of a grain during solidification, e.g. how strain and stress localize in a single grain during semi-solid deformation. Fuloria and Lee [179] simulated inelastic deformation of a single columnar dendritic structure of various solid fraction. They found a flow stress relation based on the simulated results:

\[
\sigma = K \times \varepsilon^m \times \dot{\varepsilon}^n \times F(f_s), \quad 0.1 < f_s < 0.9 \tag{2.18}
\]

\[
F(f_s) = A_1 + \frac{(A_2 - A_1)}{1 + 10^{(0.7172-f_s)p}} \tag{2.19}
\]

where \(A_1, A_2\) and \(p\) are parameters related to grain morphology.

Yamaguchi and Beckermann [180–182] coupled a phase field solidification model with a material point deformation method to simulate deformation of growing single and multiple dendrites during solidification. Not only could this model simulate stress and strain accumulation and distribution during grain growth coupled with deformation, but also the evolution of crystal orientation (Figure 2.21).

![Figure 2.21 Predicted phase-field, von Mises stress, equivalent plastic strain and crystallographic orientation angle during compression of a single growing dendrite with 15% compression, after [182].](image-url)
In conclusion, theories and models covering the underlying semi-solid mechanisms have advanced considerably over the last couple of decades. Recent developments of those advanced microstructural models (e.g. [27,172]) are guided or verified by the latest in situ observations of semi-solid deformation. To further develop existing or new semi-solid rheological theories and models, there is a critical need to design new in situ semi-solid deformation experiments using the state-of-art high speed X-ray tomography.

As illustrated in the literature, deformation imposed in the casting can have significant impacts on solidified structure, refining grains or causing defects including segregation band and porosity. However, how their formation correlated with imposed deformation is not fully understood. A few in situ semi-solid experiments, especially the latest high speed X-ray tomography were developed which shows the advantage of in situ studies (revealing the dynamics of two phase structure (Chapter 2.5)). There are also arguments how the semi-solid shall be treated in the deformation models (see Chapter 2.6). Those models need to be validated or disproved ideally through in situ investigation. This thesis, written in a way that each chapter is a journal article format, focuses on the combined use of high speed X-ray tomography and thermo-mechanical test to study semi-solid deformation under different configurations, namely, uniaxial compression (Chapter 2), extrusion (Chapter 3) and indentation (Chapter 4). In addition to that, precise tomographic quantification and imaged based modeling have been used, uncovering key mechanisms governing how semi-solids response to imposed deformation and providing benchmark cases for semi-solid micromechanical models.
Chapter 3. Semi-solid compression*

The microstructural and mechanical response during the deformation of semi-solid mixtures is termed semi-solid mechanics, which is concerned with both materials processing (e.g. metallic component fabrication [6,179]) and natural phenomena (e.g. magma flows [183,184]). For example, during the casting of aerospace or automotive metallic components, the thermal contraction and/or imposed deformation during solidification influences the microstructure and defects formed (e.g. grain size [9], porosity [185], segregation [8] and hot tearing [179,186]). In many industrial processes where deformation is imposed, such as semi-solid processing and twin roll casting, this effect is particularly strong [9,41]. Therefore, an improved understanding of the response of a solidifying structure to deformation is important when designing a range of manufacturing processes.

Semi-solid systems are conventionally treated as homogeneous media, described using governing laws based on a continuum approach [20,127]. However, this treatment cannot account for localized phenomena, such as strain localization, which leads to defects such as shear banding or void formation [92]. A granular mechanics approach has been proposed by some authors to link the microstructural evolution to the semi-solid responses due to deformation, by treating solid grains as particulate suspensions [9,18,92,142,172]. For example, Spencer et al. [18] discovered that a semi-solid metallic alloy’s viscosity depends on the solid fraction and decreases with increasing shear rate (shear-thinning), analogous to colloidal suspensions. Tzimas and Zavaliangos [187] discussed the occurrence of dilatancy in

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semi-solid equiaxed alloys during compression and more recently, Gourlay et al. [6] reported that partially solidified alloys exhibit Reynolds’ dilatancy under shear leading to strain localization. Dilation is also an important feature of saturated granular materials. The previous studies suggest that the size and morphology of solid particles and the liquid fraction influence the occurrence of granular phenomena in semi-solid alloys [49,188]. Although prior studies have shown some aspects of granular behavior, grain-grain interactions and liquid flow lead to various other localized phenomena at the microstructural scale, such as flow of solid particles [145,189], agglomeration/deagglomeration [41,112], visco-plastic deformation of grains [179,181,190] and formation of damage [22,96,191]. Whether such behavior can be governed by granular mechanics needs to be validated through experiments.

Commonly, constitutive equations have been used to describe the mechanical behavior of semi-solid alloys via continuum analysis of a range of tests, including: tensile loading [96–99], compression [19,94,95], direct shear [100], rheometry [18,49,93] and indentation[102]. It is worth noting here that compression has been extensively used due to its ease of implementation and close resemblance to many key industrial processes. Additionally, properties such as the yield stress and viscosity of a semi-solid mush can be directly measured [19,94,95]. However, in most of these studies, the effects of deformation on microstructure were quantified only using post mortem analysis, limiting our understanding of any time-dependent kinetics. To understand and quantify the underlying kinetics or microstructural dependent interactions, simultaneous measurement of the mechanical properties of semi-solid alloys and direct quantification of microstructural evolution with time is necessary.

A few recent studies have reported direct observation of granular shear deformation in semi-solid Al-15wt%Cu alloys [189] and low carbon steel [144] using X-ray radiography, but did not measure the macroscopic mechanical behavior. However, these 2D studies did provide
direct evidence of local dilatancy, induced by rearrangement of grains under shear deformation in metallic systems [138,189,192]. Unfortunately, these radiographic observations require a very thin sample thickness, and may not represent the 3D bulk behavior due to restricted out-of-plane motion and the friction of particles along the sample container wall.

High speed X-ray tomography can now overcome many of the limitations of radiography, resolving real-time 4D (3D plus time) information [26,163,193,194]. This technique has been used by several authors to quantify microstructure and defect formation during solidification [158,195,196] and under tension [27,186,197]. In this chapter, we present the first in situ 4D quantitation of semi-solid compression of equiaxed dendritic grains. Using a bespoke thermo-mechanical rig designed for X-ray tomography, both the macroscopic mechanical behavior, and the evolution of microstructure and damage are simultaneously measured. The microstructural dynamics can then be correlated with the true stress and strain measurements, providing fundamental understanding of the responses of partially solidified alloys to the imposed loading. We demonstrate that this methodology can provide unique advantages when developing and validating semi-solid constitutive models, elucidating the behavior of granular semi-solid systems and the nature of underlying granular deformation mechanisms.

3.1 Experimental methods

3.1.1 Materials

An Al-15wt.%Cu alloy was selected for two key reasons: 1. to achieve a solid fraction typical of widely used commercial alloys (e.g. A356 [198]); and 2. its good X-ray attenuation variation between the primary phase and interdendritic liquid. The latter is due to the higher electron density of copper and its low partition coefficient, preferentially segregating into the
interdendritic liquid. Cylinders 3 mm diameter and 4 mm high were wire electro-discharge machined (EDM) from 2 kg cast cylinders. Their microstructure was equiaxed dendritic, with a grain size of ~600 µm.

3.1.2 Testing apparatus and procedures

Semi-solid compression tests were performed using the bespoke P2R mechanical test rig [186] designed for *in situ* X-ray tomographic experiments, with air bearing continuous rotation built into the load train. This allows simultaneous tension, compression and/or torsion during tomography, with 100 nm motion and 0.1 N load measurement precision. A bespoke PID controlled resistance furnace with an X-ray transparent window [163,186,199] was mounted on the mechanical rig, and the entire thermo-mechanical setup was integrated into the I12 beamline at Diamond Light Source.

The experimental setup is shown in the insert of Figure 3.1. The specimen was placed at the center of a boron nitride holder (inner diameter of 7 mm, wall thickness of 1 mm), to ensure that the sample was secure and the deformation was unconstrained. A pre-load of 3 N was applied to stabilize the sample during rotation for tomography. The specimen was heated at a rate of 40 °C/min to 555±2 °C, where it is semi-solid with a liquid fraction of 30%±2%, and held isothermally for 10 min during which slight coarsening occurred. (Note, the liquid fraction was determined by image analysis of tomographic scans, but compares well to the equilibrium phase diagram). The sample was then compressed at a displacement rate of 5 µm/s (an initial strain rate of about 1.25×10⁻³ s⁻¹). After 20 s, continuous high speed X-ray tomographic imaging was initiated, and 24 datasets were continuously captured over 96 s (i.e. one tomogram every 4 s of 720 images collected over 180°).

A monochromatic X-ray beam with photon energy of 53 keV was used in the experiment. A Phantom V7.3 high speed camera (Vision Research, USA), together with a LuAg:Ce single
crystal scintillator was used, offering a field of view of 9.8 × 7.3 mm$^2$ and a voxel size of 12.25 µm. An exposure time of 5.5 ms was used.

![Figure 3.1](image)

Figure 3.1 (a) Picture of P2R compression rig at the I12 beamline in Diamond Light Source with furnace and the insert shows the schematic of the set up: (1) compression (top) ram (2) specimen (3) boron nitride holder; (b) A solidworks model of P2R.

### 3.1.3 Image reconstruction and quantification

A filtered back projection algorithm was used to reconstruct the 3D tomographic datasets, including ring artifact removal [152]. Each reconstructed 3D dataset was 800 × 600 × 600 pixels. Image process and analysis was performed using Avizo 7.0.1 (Visualization Science Group, France), ImageJ (U.S. NIH Bethesda, USA) [200] and MATLAB 2012b (The Mathworks Inc., USA). The first stage was a 3D median filter algorithm to reduce noise, followed by registration using a 3D affine registration approach. A longitudinal section of an individual tomogram is shown in Figure 3.3, where the $\alpha$-Al dendrites are dark grey, and the Cu enriched interdendritic liquid is light grey. Global thresholding values were used to segment the images into solid, liquid and void. Voids smaller than 27 voxels were ignored. The local thickness of the liquid channel was determined using BoneJ, an ImageJ plugin.
The channel thickness is measured as the diameter of the greatest sphere that fits within the liquid channel and which contains the measuring point [201].

The length ($l_i$, where $i$ is the scan step), maximum cross-section area ($A_i$) and volume ($V_i$) of the sample are measured from the 3D tomographic volume, and used to calculate the true axial ($\varepsilon_l = \ln\left(\frac{l_i}{l_0}\right)$), and lateral ($\varepsilon_a = \ln\left(\frac{A_i}{A_0}\right)$) strains.

### 3.1.4 Digital Volume Correlation

Digital volume correlation, DVC (Davis Strain Master, Version 8.1), was used to provide a full-field displacement field and 3D strain map. DVC tracks the patterns of grey value within small subvolumes in a digital 3D image, calculating their motion between frames, and hence displacements and strains [202]. Cuboidal subvolumes of 24 pixels edge length, were chosen as a balance between being large enough to contain a unique pattern for matching, whilst being small enough to avoid affine straining over a deformation increment [203]. Using a 50% overlap and 4 passes, a displacement matrix grid of 12 pixels or 147 µm was obtained. Displacement vectors with a correlation coefficient less than 0.5 were deleted.

Figure 3.2 schematically illustrates the application of digital volume correlation for the determination of the strain field on the time-revolved X-ray tomographic datasets. The displacement fields ($u_m$, $m = i, ii, iii \ldots$) were calculated from the nearby tomographic dataset (step 1→step 2, step 2→step 3\ldots). Then the measured displacements were accumulated (i, i+ii, i+ii+iii \ldots) to calculate the integrated displacement fields ($U_n$, $n = I, II, III\ldots$) in the data series. The integrated displacement fields were used to calculate the strain tensor ($\varepsilon_{ij} (i, j = x, y, z)$ by the finite difference method. The coordinates were displayed in, $z$ being the vertical axis in the direction of loading.
\[
\varepsilon_{ij} = \left(\frac{\partial U_i}{\partial j} + \frac{\partial U_j}{\partial i}\right) \quad (i, j = x, y, z)
\]

3.1

Figure 3.2 Schematic of the full-field strain measurement using digital volume correlation on time-revolved X-ray tomographic datasets.

The six unique components of \( \varepsilon_{ij} \) define a general 3D strain state, and can be transformed onto the octahedral planes, which are the 8 planes forming equal angles with each of the principal strain directions, and results in the octahedral normal strain \( (\varepsilon_n) \):

\[
\varepsilon_n = \frac{1}{3}(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})
\]

3.2

and the octahedral shear strain \( (\varepsilon_s) \):

\[
\varepsilon_s = \frac{2}{3} \sqrt{(\varepsilon_{xx} - \varepsilon_{yy})^2 + (\varepsilon_{xx} - \varepsilon_{zz})^2 + (\varepsilon_{yy} - \varepsilon_{zz})^2 + 6(\varepsilon_{xy}^2 + \varepsilon_{xz}^2 + \varepsilon_{yz}^2)}
\]

3.3

\( \varepsilon_n \) describes the volume change, equivalent to 3 times the volumetric strain, while \( \varepsilon_s \) is the maximum value of the shear strain on any plane. Both components are independent of the orientation of the coordinate system.
3.2 Results and discussion

3.2.1 Dilation during semi-solid compression

Initially (Figure 3.3(a) and (f)), there is a uniform distribution of interdendritic liquid throughout the sample with only minor microstructural variations. As compression proceeds ($\varepsilon_l=-1.2\%$), the sample shows typical barreling (Figure 3.3(b)) due to friction at the plattens; the $\alpha$-Al equiaxed dendritic grains separate (Figure 3.3(c-d)) with a corresponding increase in liquid volume fraction (Figure 3.3 (f)-(g)). We also observe void nucleation and growth with increasing strain (Figure 3.4), showing the complex, heterogeneous interaction and deformation of the three-phases (solid dendritic grains, interdendric liquid and voids) under compression, which will be quantitatively described below.

Grain movement during semi-solid compression is observed by tracking markers at centroids and dendrite tips of the grains. The motion of these markers was quantified at axial strain intervals of approximately 4%, and tracked both in the longitudinal and transverse slices, using the bottom ram as the frame of reference (Figure 3.4 (a) and (b)); grains just below the top ram (upper part) move vertically downward, while grains in the middle region move simultaneously downwards and outwards due to barreling. The extent of motion in a single transverse mid-height plane is illustrated in Figure 3.6a; the radial motion of grains is significantly greatest at the central region (Figure 3.6(b)). As grains move, they also undergo small rotations.
Figure 3.3 (a)-(d) Series of longitudinal slices from 3D volume of semi-solid Al-15wt.%Cu alloy with solid fraction of 70 ± 2% under semi-solid compression; (f)-(i) mid-height transverse slices.

Figure 3.4 X-ray tomograms of semi-solid Al-15wt.%Cu alloy (fraction liquid of 30 ± 2%) at four stages of compression: (a) $t = 24$ s, $\varepsilon_l = 1.2\%$; (b) $t = 52$ s, $\varepsilon_l = 4.8\%$; (c) $t = 76$ s, $\varepsilon_l = 7.3\%$; and (d) $t = 116$ s, $\varepsilon_l = 12.6\%$. 
Figure 3.5 (a) and (b) Grain displacement vector (scale factor = 1) from $\varepsilon=-1.2\%$ to $\varepsilon=-12.6\%$.

Figure 3.6 (a) Grain motion in the transverse mid-height section; (b) average radial marker displacement versus height (st1 at $t=24$ s ($\varepsilon=-1.2\%$), st2 at $t=52$ s ($\varepsilon=-4.2\%$), st3 at $t=84$ s ($\varepsilon=-8.3\%$) and st4 at $t=116$ s ($\varepsilon=-12.6\%$)).

The gradual change in the cross-sectional area of the sample is quantified in Figure 3.7, with
the corresponding increase in intergranular liquid fraction in Figure 3.8; there are fluctuations in the fraction liquid due to variations in grain and dendrite morphology, with periodicities at both of these length scales. The distribution of local thickness of the liquid channels in the final stage of deformation was measured via the medial axis method [201] and plotted in the inserts of Figure 3.6b. Figure 3.10 (a) and (c) compare the local thickness of the liquid channel in a region of interest (1.47 mm³ extracted from the central region of the sample, marked using the black box in Fig. 4(b)) at $\varepsilon_l = -1.2\%$ and at the final step of the deformation ($\varepsilon_l = -12.6\%$). The inter-granular channels are initially thin and highly tortuous (Figure 3.10(a)), with regions thicker than 73 µm only at the grain triple points (Figure 3.10 (b)). By the final stage of deformation (Figure 3.10 (c)), the liquid channel thickness has increased dramatically in the central region, with interconnected regions thicker than 73 µm between almost all grains (Figure 3.10 (d)). This is quantified in Figure 3.11(e) for the two strains as the distribution of liquid channel thickness, with maximum channel thicknesses of ~98 µm and ~145 µm, respectively. This observation is an example of dilatancy, first described by Osborne Reynolds [204]. The grains were naturally packed initially, and when pushed, they rearranged themselves to accommodate strain, widening the interstices in between.

![Figure 3.7 Variation in transverse cross-sectional area of the sample along deformation axis.](image-url)
Another phenomenon that is less well reported was also observed. The dilation of the grains in the central region introduces a zone of low liquid pressure, which draws in interdendritic liquid from surrounding regions (Figure 3.8). This flow locally increases the average Cu concentration via mass transport, reducing the local equilibrium melting temperature and leading to remelting of the solid grains. Although such remelting is best known in solute plumes/freckles due to thermal solute convection [205], observations in semi-solid tension via in situ X-ray radiography [15] and tomography [25] on Al-Cu alloys have indicated it could occur during deformation. This local change in composition will further contribute to
the increase in liquid fraction in the central, dilated region. Hence dilatancy in semi-solid alloys is different from those in insoluble soils, as localized changes in thermodynamic equilibrium may cause greater localization of shear banding. This effect may be significant in other fields such as volcanology, where an igneous magma is also a granular system that has a solid fraction that is dependent on local composition; deformation of a magma mass may cause the onset of dilation, producing zones of higher liquid fraction within the strain localized region [183] where remelting might also occur.

Dilatancy not only causes the increase of liquid volume fraction in the deformed region, but also leads to the formation of voids/damage (Note we use the term damage interchangeably with voids, as the linkage of them leads to loss of mechanical integrity). Three distinct stages of void evolution were identified in Figure 3.13 and Figure 3.14: I. initial pore shrinkage, II. an incubation period, and III. final rapid growth leading to cracking.

Figure 3.10 (a) and (c) Liquid channel local thickness in central region for the subvolume box in Figure 3.9 at $t=24s$ ($\varepsilon_l=-1.2\%$) and $t=116 s$ ($\varepsilon_l=-12.6\%$), respectively. Liquid channels with local thickness larger than 73.5 $\mu$m were rendered in (b) and (d).
During stage I, the initial, 0.11% pre-existing porosity (Figure 3.14(a)) shrinks as a result of both compressive strain and liquid feeding into the pore space, and one single pore as an example is shown in Figure 3.14(b)). Interestingly, Terzi *et al.* [27] also observed this during the initial stages of semi-solid tensile loading. During stage II, existing voids grow slightly along dendritic boundaries ((b)-(d)), and a few new ones are initiated. Two competing factors arise controlling the formation of voids in the mush zone - (i) tensile strain imposed on the liquid channel trying to open up a void, (ii) the liquid feeding tending to feed the dilated space. Thus, when a critical amount of tensile strain is imposed on the liquid channels and the liquid fails to feed the dilated space, voids grow or are nucleated, resembling hot tear formation [15,90]. During the last stage (Stage III), voids both rapidly grow and coalesce (starting at an axial strain of -6.4 %), as seen in (d)-(f). The damage penetrates into the sample ((d)), and extending in the vertical direction ((f)-(g)). Damage formation in a semi-solid depends on a balance between strain rate and fluid flow. At high strain rates, damage forms faster as liquid flow is insufficient to feed the newly dilated interdendritic space.
Figure 3.12 (a) to (f) Evolution of voids (coloured according to its size) during semi-solid compression at $f_s = 70\%$ of Al-15wt%Cu sample (transparent grey) at deformation rate of 5 µm/s at an axis strain of (a) 0, (b) -1.2%, (c) -2.6%, (d) -4.2%, (e) -8.3%, (f) -12.6%.

Figure 3.13 Variation of void fraction in area along deformation axis.
3.2.2 Bending and fragmentation of grains

Although dilatancy was the predominant mechanism for accommodating strain, other deformation mechanisms were also observed. Due to the irregular dendritic morphology, some grains interlocked, causing deformation within individual grains (termed intra-granular deformation), specifically bending and fragmentation of the primary dendrite stems.

An example of this is shown in Figure 3.15(a) to (c); seven grains are labeled A-G to explain the breakage of C, bending of F, and the associated force chain. When dendrite F is pushed down, its stem fragments near one tip (Figure 3.15(b)). Load is transmitted to A and B from the grains above, pinning them against C, eventually shearing the dendritic stem of grain C.
(Figure 3.15 (f)-(g)). This breakoff may also have been added by local remelting although the tomography resolution was insufficient to quantify this.

Figure 3.15 Zoomed longitudinal slice showing the bending and breakage of dendrites at time (a) 24 s, (b) 84 s (c) 116 s; (d) 3D rendering of α-Al grain marked ‘C’ in images a-c at time 24 s, then at subsequent times (e-g) as the primary arm fragments due to loading from neighboring grainsto ‘A’ and ‘D’. (h)-(k) 3D rendering of α-Al grains ‘F’ at subsequent times as the primary arm stem bends due to slow loading from neighboring grains ‘E’ and ‘G’.

Looking now at grains E, F and G, a similar micro-mechanism is observed for ‘F’ leading to its eventual bending. These grains also form a micro shear cell, where E and G tend to shear F in anti-clockwise direction (Figure 3.15(b)). Under this configuration, the primary stem of grain ‘F’ bends ~15° without breaking (Figure 3.15(k)).
The current observations indicate that dendrites are sheared by their neighbors through a direct-shear mechanism; the shear force imposes a sufficiently high bending moment on the dendrite arms to bend and ultimately break off portions of these equiaxed dendritic grains. Throughout the sample, many bent dendrites were observed, but break-off was less prevalent. Further detailed studies are required to quantify the loads and stresses responsible for breaking individual dendrites.

For this particular solid fraction, dendritic morphology and strain rate, the predominant mechanisms, in order of prevalence, are: (1) dilatancy; (2) grain deformation and (3) dendrite fragmentation. Granular models, particularly discrete element models of semi-solid materials, commonly consider the solid grains as rigid particles and treat particle-particle deformation as elastic deformation [172]. However, the present study suggests that the deformation of semi-solid equiaxed dendritic structures is considerably more complex.

3.2.3 Stress and strain measurement

In this section we correlate the granular flow behavior and microstructural evolution to the measurement of the mechanical properties (stress and strain) during semi-solid deformation. During deformation, displacement and load values were recorded at every 1 ms, as plotted in Figure 3.16. (Note the load fluctuates by 20% with a periodicity that is equal to the rotation speed resolution for tomography due to slightly non-concentric loading, therefore a weighted moving averaged load was used). The resulting stress-time curve (Figure 3.16 has a maximum stress of about 1.4 MPa at t=68 s (axial strain of -6.4%), corresponding to the initiation of rapid damage formation (Stage II to III), and slowly decreases afterwards. The total work done on the sample is mainly consumed by: (i) grain rearrangement, (ii) deformation within grains (iii) grain fragmentation, (iv) liquid flow through restricted channels and (v) formation and growth of voids.
Figure 3.16 Load and true stress curve versus time during semi-solid compression.

The axial and lateral strains versus time (Figure 3.17) monotonically decrease and increase respectively during compression, as expected. The axial strain as calculated by DVC ($\varepsilon_{zz}$) is also plotted in Figure 3.17, correlating well to the traditional measurement ($\varepsilon_l$). The increase in standard deviation of $\varepsilon_{zz}$ indicates that the strain becomes more inhomogeneous during the course of the deformation, shown even more clearly by the DVC results (Figure 3.18).

Figure 3.17 Axial strain, lateral strain, and $\varepsilon_{zz}$ with its standard derivation versus time.

During the initial stages of deformation, the specimen predominantly exhibits relatively homogeneous negative normal strain, indicating compaction within the sample (Figure 3.18 (a)). As deformation progresses, positive normal strain (corresponding to dilation) occurs
locally within the central region (Figure 3.18 (b)), accumulating with increasing deformation (Figure 3.18 (c)-(d)). The region with the highest positive normal strain (>3%) is mainly located in the central region of the specimen (Figure 3.18 (d)). The octahedral shear strain tends to concentrate into a band (Figure 3.18 (e-h)), with the highest shear strain exceeding 12% (Figure 3.18 (h)). This suggests that the shear localization occurs as deformation increases, indicating the presence of dilatant shear bands.

Figure 3.18 Development of octahedral normal strain; and (e)-(h) shear strain during semi-solid compression on the med-longitude cross section: (a) and (e) $\varepsilon_l = 0$ to -1.2%, (b) and (f) $\varepsilon_l = 0$ to -4.3%, (c) and (g) $\varepsilon_l = 0$ to -8.3%, (d) and (h) $\varepsilon_l = 0$ to -12.6%.

The distribution of the principal strain ($\varepsilon_1$) as measured by DVC is shown on the outer surface and cross-section of the specimen in Figure 3.19, at four stages of compression. The iso-surface of $\varepsilon_1$ equal to 20% is rendered in red in the other half of the specimen, showing the localisation of the strain into shear bands. The highest strain levels are first observed in the regions where the interdendritic channels widen between grains, followed by subsequent void / damage formation, often connected to the surface of the sample (compare Figure 3.19d & Figure 3.19d). Going through the successive compression steps, initially the strain is homogeneous (Figure 3.19a, axial strain 3.6%). However, when compressed to $\varepsilon_l = 7.3%$,
localisation starts to occur as the grains contact, rotating and shearing (Figure 3.19b), although $\varepsilon_1$ larger than 20% is still negligible. When compressed to $\varepsilon_l = 10.2\%$ (Figure 3.19c), dilatant effects become even stronger, and there is a considerable increase of the fraction of $\varepsilon_1$ larger than 20% with the on-set of what appears to be shear banding. At the final stage of deformation ($\varepsilon_l = 12.6\%$), $\varepsilon_1$ is connected to the edge of the cylindrical mask, with strong bands of localised strain (Figure 3.19d).

![Figure 3.19 3D rendering of the first principal strain during semi-solid compression (red iso-surface of $\varepsilon_1=20\%$) at axial strains of: (a) 3.6%; (b) 7.3%; (c) 10.2%; and (d) 12.6%.](image)

### 3.2.4 Effect of grain structure

Grain structure has been found to influence the rheological properties of semi-solid alloys [73]. However, little is known about the cause of this influence. In order to clarify this effect, two extreme points were chosen: a sample with large dendritic grains (LD, grain size ~ 700 µm) similar to the afore-discussed sample, and another sample with fine globular grains (FG, grain size ~ 60 µm). They were heated to 560 ± 2 °C ($f_s = \sim 70\%$), isothermally holding for 20
min, then compressed at 1 µm/s. High speed X-ray tomography was used during the compression.

The vertical slices extracted from the 3D volume show the difference between the two samples (Figure 3.20 and Figure 3.22 for LD and FG, respectively). LD shows signs of dilatancy at the initial stage of deformation (Figure 3.20(b)): the enlargement of liquid channels, which continued through the course of deformation. At a latter stage, damages started to form from the periphery and propagated. The final compressed structure is shown in Figure 3.21. This is similar to the afore-discussed case of the same large dendritic structure (Section 3.2.1). However, the dilatancy effect is much less pronounced in the FG sample. As shown in Figure 3.22, through continuously compressing the sample, there is no obvious opening of liquid channels between grains and no formation of damages during the course of deformation.

In addition to the obvious difference in the level of dilatancy between the LD and FG samples, the measured force is clearly distinct between the two samples (Figure 3.24). The force measured during semi-solid compression of LD sample is more than 15 times larger than the FG counterpart at the same strain level. For instance, at a displacement of 900 µm, the force of LD is ~10 N, while that of FG is only ~0.6 N. This demonstrates the superior flow characteristics of the FG sample.

The refined globular semisolid sample deforms and flows more easily than the large dendritic one and shows less susceptible to dilatancy, indicating that the grain structure (size, morphology and distribution) controls the interdendritic volume fraction and the level of dilatancy, hence, in the end, determines the rheological properties and severity of damage as a result of semi-solid deformation.
Figure 3.20 Series of longitudinal slices from 3D tomographic volume of semi-solid dendritic structure at various compression stages (displacement rate is 1 µm/s).

Figure 3.21 Enlarged view of the compressed structure at $d = 828$ µm
Figure 3.22 Series of longitudinal slices from 3D tomographic volume of semi-solid fine globular structure.

Figure 3.23 Enlarged view of the compressed FG sample at \( d = 892 \, \mu m \)
Figure 3.24 Load-time curve of the semi-solid specimens with (a) large dendritic (LD) grains; (b) fine globular (FG) grains.

### 3.3 Summery

High speed synchrotron X-ray tomography was used to quantitatively analyze the compressive deformation of semi-solid equiaxed dendritic Al-15wt%Cu at ~30 % liquid fraction. The results confirm the behavior of this semi-solid structure as a granular system, highlighting how the deformation is accommodated not only by granular flow and associated dilatancy, but also by the formation of micro-shear cells resulting in intra-granular bending and breakage of dendrites. These mechanisms lead to defect formation, causing segregation.
(which may increase dilatant shear band formation) and damage formation. Three distinct stages of void evolution were identified: I. initial pore shrinkage, II. an incubation period with slow pore growth, and III. final rapid damage growth, both from the surface and via coalescence of internal and externally connected voids. These stages are directly linked to granular behavior of the solid phase.

Using digital volume correlation, analysis of the full-field strain evolution showed regions of high strain concentrations corresponding to the formation of shear bands at a granular/microstructural level.

It is also shown that grain structure plays a significant role on the deformation responses of semi-solid alloys. The fine grained semi-solid alloy shows much less tendency to dilatancy and less resistant to flow than the large dendritic semi-solid alloy.
Chapter 4. Semi-solid extrusion*

Deformation-induced melt flow is a key phenomenon in a wide range of processes from metal casting [8,36] to volcanology [184,206,207]. During casting, deformation of semi-solid alloy can influence liquid flow, resulting in segregation, degrading the mechanical properties of the final product [8,77,79,83,208]. The deformation could be caused by thermal contraction or deliberately applied external forces, for example in high pressure die casting and squeeze-casting. Similarly, semi-solid magmas, when transported from the Earth’s mantle to the surface, are deformed by convection and/or tectonic plate spreading, which induces melt migration and segregation [184,206,209]. Therefore, an improved understanding of liquid flow through a deformable granular or porous medium is important for optimizing casting process [77,210] and predicting natural disasters like volcanic eruptions [184,209].

In solidification processing, deformation in the semi-solid can induce a range of defects, including extrusion segregation in squeeze-casting [36] and surface exudation in direct-chill casting (as shown in Figure 2.7(a))[83]. Although several prior investigations have identified deformation-driven melt flow as a possible mechanism of such defects [36,83,211], the influence of stress on a semi-solid alloy and the melt flow through the equiaxed microstructure are not clearly understood. Many models have been developed to predict the formation of those defects, based on the proposition of the mushy zone as a sponge saturated with liquid [8,81,83,210]. However, currently there are no direct validation techniques that capture the kinetics incorporated in this hypothesis; in situ synchrotron tomography is one possible solution.

Recently, as discussed in Section 2.5.3, high speed X-ray tomography has been utilized to perform four dimensional imaging (4D, i.e. 3D plus time) of the pore-scale fluid flow [162], solidification [66,156,212], and the influence of deformation on semi-solid alloys [25,186,213]. Tensile and uniaxial compression tests have been used previously with the help of 4D imaging to study semi-solid deformation; these were mainly focused on the formation of damage (hot tearing) as a result of the granular response of the mushy zone [25,213,214]. In this chapter, we describe the application of an indirect extrusion cell to study the rheological behavior of semi-solid alloys and the mechanisms responsible for the liquid migration induced by deformation. Such an indirect extrusion cell can also be used to study how extrusion segregation and exudation form, since it mimics their forming conditions.

4.1 Materials and Methods

The sample was equiaxed dendritic Al-15wt%Cu. A cylindrical specimen of 2.9 mm in diameter by 2.9 mm long was prepared using wire electro-discharge machining. It was placed in a boron nitride holder with an inner diameter (ID) of 3 mm and outer diameter (OD) of 5 mm. An alumina tube (1.5 mm ID and 3 mm OD) was placed on top of the specimen forming an indirect extrusion cell (Figure 4.1). The entire extrusion set-up was enclosed within a resistive furnace [213], mounted on a bespoke mechanical testing rig with inbuilt rotation (P2R [186,213]).

The experiment was conducted using 53 keV monochromatic X-rays on the I12 beamline at Diamond Light Source, UK. A high speed X-ray imaging system was used, consisting of the beamline’s custom-built imaging modules coupled to a CMOS camera (Miro 310M, Vision Research, USA). The imaging system provided a field of view (FOV) of 5.12 \times 3.2 mm and 4 \mu m pixel size. The sample was positioned so that the top half of the billet and extrudate was in the FOV. The sample was heated to 560±2 °C in 15 min, and then held for 10 min for
thermal homogenization. Subsequently, the top ram was moved down at 1 µm/s, forcing the alumina tube downwards while measuring loads.

![Schematic illustration of the extrusion cell, built inside a resistance furnace: 1-alumina tube; 2-specimen; 3-boron nitride holder.](image)

Figure 4.1 Schematic illustration of the extrusion cell, built inside a resistance furnace: 1-alumina tube; 2-specimen; 3-boron nitride holder.

Seven tomograms were captured, each comprising of 900 radiographs, collected within 9 s at 45 s intervals. A filtered back projection algorithm was used to reconstruct the data to generate a tomography [152], which was converted to 16 bit dataset. Noise reduction was performed using a 3D median filter, followed by an anisotropic diffusion filter by Avizo 8 (FEI VSG, France). The result is shown in Figure 4.3 (b) together with the original image (Figure 4.3(a). The histogram after noise reduction is shown in Figure 4.2, revealing the consistence of the gray level of the reconstructed volumes. Liquid phases were segmented by Ostu method [215] using MATLAB 2012b (The Mathworks Inc., USA). The resulting segmented image is shown in Figure 4.3 (c). Varying the value calculated by this method by ±50 was used to determine the errors. Avizo 8 (FEI VSG, France) was used to visualize the 3D data.
Figure 4.2 Histograms of the collected tomographies (colours represents different tomographies).

Figure 4.3 Image processing steps: (a) the original reconstructed image, (b) filtered image, (c) segmentation of the $\alpha$-Al (white) and interdendritic liquid (black).
4.2 Results

Figure 4.4 (a) to (c) display the resulting 3D views of the specimen under extrusion at the displacement of 0, 162 and 324 µm. The corresponding 2D longitudinal slices are shown in Figure 4.4 (d) to (f). The dark grey dendrites are the α-Al grains, while the Cu-enriched liquid is light grey. Note a small amount of liquid segregated into the tube on top of the sample (Figure 4.4(d) at d = 0 µm). This small amount of extrudate was due to the stress caused by thermal expansion during heating. The subsequent response of the mush to the applied deformation is shown in Figure 4.4e (162 µm) and Figure 4.4f (324 µm). As deformation progressed, more melt flowed into the alumina tube from the semi-solid specimen. The liquid channels under the wall of the extrusion tube closed in response to the deformation (zone D in Figure 4.4e to 2f). The evolution of the extruded liquid (Figure 4.5) displays the characteristic profile of laminar flow in a pipe. We can also observe the closure of pre-existing porosity (Figure 4.6) due to the compressive strain.

In addition to making the above qualitative observations, we performed a detailed, time-resolved quantification of the extrusion. From $d = 0$ to 324 µm, the volume of the expelled liquid in the tube increased from ~0.2 to ~2 mm$^3$ at an almost constant rate of ~0.0055 mm$^3$ per µm displacement. The extruded liquid volume increases at the same rate as the volumetric displacement (~0.0053 mm$^3$/µm) of the alumina tube. The liquid fraction in the billet (lower part of the specimen) decreased from 26.7±2.8% to 15.1±2.1%, indicating densification of the mush (Figure 4.7(b)). The extraction of the liquid by compression of the solid skeleton can be understood by considering the mush to be a saturated sponge, consisting of two phases (the solid grains and the liquid phase). This observation is contrary to the shear-induced dilation observed during uniaxial semi-solid compression of equiaxed dendrites [213] and globular grains [214], where the liquid channels locally open rather than close.
Figure 4.4 Semi-solid extrusion at displacement rate of 5 µm/s at three degree of deformation: (a), (b) $d = 0$ µm, (c) and (d) $d = 162$ µm, (e) and (f) $d = 324$ µm. (a), (c) and (e) 3D view of the semi-solid extrusion; (b), (d) and (f) 2D longitude slices (the deformation axis is vertical),

Figure 4.5 3D profile of the extruded liquid,
Along with liquid being expelled, a small amount of solid phase was pushed into the die cavity (Figure 4.4 (e) and (f)). The peak height of extruded solid increased gradually (Figure 4.7(a)). A magnified view of the extruded grains is shown in Figure 4.8 (a) and (b). Those grains located near the extruder inlet were free to move and appear to be sheared by the grains below, leading to dilatant translation and rotation (e.g. the grain A moved down ~0.3 mm and rotated ~7° in anticlockwise direction, while grain B underwent ~12° clockwise rotation). Consequently, the liquid-filled interstitial space increased slightly (Figure 4.8(b)). Buoyancy force might also play a role in the grain movement as the Cu-rich liquid is denser than the α-Al solid. The movement of grains due to deformation and associated changes of
interdendritic liquid will cause both compositional and microstructural variation in the final component.

Figure 4.8 (a) and (b) Magnified view showing the grain movements near the extruder inlet.

Determining the mechanical response of the mush requires knowledge of the strength of the dendritic/globular \( \alpha \)-Al network and the resistance of the liquid flow. Although calculating the strength of \( \alpha \)-Al network would require complex simulations, we can use the 3D geometry of the liquid network to directly determine the permeability, or resistance to the flow of the interdendritic liquid. This was done by solving the Navier-Stokes equations on a subset of the mush at each time step. A subvolume of \( 2 \times 2 \times 0.8 \) mm was extracted from the central region of the sample within the billet. Avizo XLab flow simulation code (FEI VSG, France) was used for the simulations (conditions detailed in Ref. [66]). The simulation is also compared with the Carman-Kozeny permeability relationship [56]:

\[
K = \frac{f_l^3}{k_C S_V^2}
\]

where \( f_l \) is the liquid fraction, \( S_V \) is the surface area of the solid per unit volume of sample measured directly from the 3D data, and \( k_C \) is set to 5 as suggested by Duncan et al [59]. The simulated permeability decreased monotonically from \( \sim 2.4 \) to \( \sim 0.5 \) \( \mu \)m\(^2\) during the 324 \( \mu \)m of extrusion (Figure 4.7(b)). Although there is disparity between the simulation and Carman-Kozeny equation, this disparity is still within the scatter of previous work [64]. The continuous decrease of permeability shows the extrusion continued to compress the solid skeleton, increasing the flow resistance and blocking further flow of the interdendritic liquid.
The force measurement (Figure 4.9) provides additional information on the mechanical response of the semi-solid specimen. The load linearly rose from 9.7±1.6 N at d=54 µm, to 35.5±2.5 N at 324 µm. The load increase rate is roughly linear at 0.1 N/µm. It is likely that further densification of the mush will significantly increase the stress as observed by Ludwig et al [126]. Note although the measured force is a combined response of liquid flow and solid deformation resistance of the mush, it is expected the liquid flow resistance is minimal as compared to the mechanical load of α-Al network.

![Force development during semi-solid extrusion](image)

Figure 4.9 The force development during semi-solid extrusion (the periodicity fluctuation of force is due to sample rotation).

Though the measured bulk properties (force, liquid fraction, permeability and expelled liquid volume) are linear with time, the deformation is inhomogeneous. This has been quantified by determining the liquid fraction within different regions (A and B in Figure 4.10 (a) insert) in the billet. Figure 4.10 (a) reveals that the liquid fraction of Region A decreased faster than that of B. At the initial stage of deformation (d = 0 µm, Figure 4.10 (b) and (d)), the liquid flowed through a complex network, which was homogeneously distributed and well connected with few isolated liquid pockets. During the extrusion, a considerable rise in the number density of isolated liquid pockets was observed from ~224 to ~896 mm$^{-3}$ in Region A, while Region B showed a marginal increase (~320 to ~448 mm$^{-3}$). At the final stage, more liquid pockets were observed in Region B than in A at 324 µm (Figure 4.10 (c) and (e)). Compressive deformation narrowed the liquid channels and closed them at their throats. The
inhomogeneous nature of deformation is due to the fact that the propagation of compression in granular medium is strongly dependent on the microstructure and tends to follow the percolating pathways [216].

Figure 4.10 (a) Liquid volume fraction in Region A and B vs. displacement; (b) to (e) 3D view of liquid channels and droplets coloured according their volume.

4.3 Discussion

This study of semi-solid extrusion and associated fluid separation provides important insight into the kinetics of deformation-driven liquid flow during solidification. It also provides an experimental validation for solidification segregation and rheological models [81,210,217]. Deformation-induced segregation is a known problem in casting [8,36]. For example, during
direct-chilled casting, the deformation (caused by thermal contraction) drives interdendritic liquid from the mushy zone into the ingot surface. This produces surface segregation and exudation (Figure 2.7) [83]. The extrusion set-up mimics such conditions and supports the hypothesis that it is the strain imposed on the solid network that causes the closure of the permeable channels and leads to the extraction of liquid from the mush.

In addition, this study demonstrates that under compaction the semi-solid alloys can be treated as a deformable porous medium saturated with liquid, although under tensile [25] or shear conditions [24,213], it can be considered as granular material. Both compaction and shear can significantly influence the redistribution of the fluid, and the modulation of permeability. This study shows constrained compressive stress densifies the solid skeleton and expels liquid from the mush; while prior investigation [213] reveals that shear-induced dilatancy can draw liquid from the surrounding neighborhood into the dilated spaces between the grains.

Additionally, deformation-driven liquid flow is applicable to a wide range of other systems including natural flows, e.g. the migration of silicate liquid in the deep region of the Earth [206,207]. However, despite its importance, the effect of stress on the melt extraction is not fully understood. Our experimental method permits the direct acquisition of quantitative kinetics of mush deformation and can provide unique insights into such complex processes.

4.4 Summary

A novel technique combining high speed synchrotron X-ray tomography and mechanical deformation was developed to measure the influence of microstructure on the rheological behavior of semi-solids. The potential of the technique has been demonstrated by observing and quantifying the rheology of a semi-solid equiaxed dendritic Al-15wt%Cu alloy. The real time 3D quantification of semi-solid extrusion provided new insights into the behavior of a
mush, as follows: the strain distribution is very inhomogeneous due to the sponge-like compression of the partially coherent equiaxed dendritic solid; the strain is mostly accommodated by inter and intra-grain compaction, with only a small amount of granular flow; the interdendric liquid is driven out of the semi-solid mush and forms an extrude; and the permeability of the compacting mush approximately follows a Carman-Kozeny relationship. These microstructural level observations can be directly used to develop and validate segregation and rheological models.
Chapter 5. Semi-solid indentation*

Indentation is one of the standard methods to probe local plastic deformation and characterize hardness and other material properties of solid samples [218–221]. This has also been employed to investigate high temperature properties such as creep [222,223], and to obtain deformation response of granular ensembles [224,225]. Granular media (e.g. sands, magma, sugar etc) consists of solid grains and pores filled with gas or liquid [226]. The behavior of a granular material under indentation by a rigid body is of vital importance with a wide range of applications, including soil-machine interaction [227], the deformation of volcanic edifices by viscous magma [224], and powder sintering fabrication [228].

It is well known that indentation can generate highly localized deformation zone in the vicinity of the indenter [125,218,229]. However, this zone is determined mostly from the final-deformation states, providing inadequate information on the dynamics. This dynamic process includes two important aspects: (1) the change of microstructures induced by the localized deformation; (2) the evolution and distribution of the strain fields as the deformation proceeds. A considerable challenge is presented when attempting to map out the very complicated time-dependent evolution of microstructures, material flow and stress/strain fields induced by the indenter, in particularly, in situ and in three dimensions (3D). This is mainly due to the opaque nature of the sample. Particle tracking methods based on optical images (which is 2D) [230–232] and nondestructive magnetic resonance techniques [226] have been developed to overcome the difficulties, which can provide material flow gradient and/or strain evolution, but limit to determine the dynamics at a microstructural scale.

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Here, we report that the combined usage of high speed synchrotron X-ray tomography and digital volume correlation (DVC), to readily resolve the microstructural evolution during the indentation in 3D, alongside evolving strain fields. The technique has been established by indenting a dense semi-solid dendritic alloy with liquid fraction of 28%, which has been proved to be of granular-like [213]. The use of such a sample not only demonstrates the advantage of the technique (can be used in a complex two phase granular system at elevated temperatures), but also shed new insights into the mechanics of granular materials, especially semi-solid alloys. The behavior of semi-solid alloys under deformation has attracted renewed attention recently (e.g. [6,172,181,213,214,233]). The complex interactions of solid-solid and solid-liquid have shown characteristic granular behavior (e.g. dilatancy and strain localization) [213,214]. It is thus important to further advance our understanding on how the granular semi-solid alloy responds to localized deformation under an indentation loading. It is also of practical relevance for advanced casting techniques because mechanical forces are not only imposed on the surface of but also penetrated deep into the casting components [208].

5.1 Materials and Methods

A cylindrical (3 mm diameter and 3 mm height) of Al-15wt.%Cu sample was used and placed inside a Boron Nitride holder with inner diameter of 3 mm and wall thickness of 1 mm. We used a purpose-built thermo-mechanical rig with a split open resistance furnace (Fig. 1) [213]. A 30° conical indenter with a flat punch made of alumina (the tip is flat with a diameter of ~250 µm) was used. The indenter was placed just on the top surface of the sample before heating the furnace up.

The beamline and X-ray imaging system is the same as Chapter 4 (please give section). The sample was heated to 560±2 °C with liquid fraction of 28% and held for 10 min. The deformation was then started at a speed of 5 µm/s. High speed tomographic scans were taken
at an interval of 9 s, acquiring one tomogram with 900 projections in 9 s over a sweep of 180°. Nine datasets were recorded in total during which the indenter travelled a distance of 720 µm. Another single tomogram was acquired at the end of the deformation process, for a total penetration of 1500 µm. Then the sample was cooled down (at a cooling rate of ~1 °C/s) with the furnace turned off. A final tomogram was recorded when the sample reached room temperature, together with dark and flat field images.

3D image reconstruction procedures described in section 4.1 of Chapter 4 were employed for this experimental dataset. A 3D medium filter followed by non-local diffusion filter using Avizo 8 (FEI VSG, France) was used to reduce the noise. The liquid and pore phase were segmented from the solid using the Otsu threshold method [215]. The thickness of the liquid channel was measured using BoneJ [201]. The movement of individual grains at a representative plane was determined via an image registration scheme. Due to the complexity of the dendritic structure and limited contrast to clearly identify the boundaries between two different dendrites, automatic isolation of the individual dendrites could not be performed. Instead, a manual separation using careful visual observations were carried out. A registration algorithm with affine transformation and an iterative optimization (Avizo 8, FEI VSG, France)
was then applied to track the movement (translation and rotation) of the same dendrite from an indentation depth of \( I = 0 \) to 720 \( \mu \text{m} \).

DaVis Strain Master Version 8.1 was used to measure the strain fields. The procedure of DVC has been described in Chapter 3 (Section 3.1.4). Here, a subset of 64 \( \times \) 64 \( \times \) 64 pixels with 50\% overlap was used. The resulting spatial resolution of the displacement field is 32 pixels, corresponding to 128 \( \mu \text{m} \). The displacement fields between the nearby tomographic dataset were integrated through the data series to calculate the accumulated displacement fields \((u_i, i=x, y, z)\). The velocity was obtained using:

\[
V_i = \frac{u_i}{\Delta t} \quad (i = x, y, z)
\]

where \( u_x, u_y, u_z \) denote the displacement at \( x, y, z \) direction, respectively, \( V_x, V_y, V_z \) represent the velocity component at \( x, y, z \) direction.

The magnitude of velocity at different directions is then calculated from the velocity field, using:

\[
v_z = |V_x|
\]

\[
v_{xy} = \sqrt{V_x^2 + V_y^2}
\]

\[
v_{xyz} = \sqrt{V_x^2 + V_y^2 + V_z^2}
\]

where \( v_z, v_{xy} \) are the magnitude of velocity along vertical (\( z \)) direction (parallel to the indenter displacement direction), on \( x-y \) plane (perpendicular to the displacement direction), respectively. \( v_{xyz} \) is the magnitude of velocity.

The accumulated displacement fields were also used to calculate the strain tensor \((\varepsilon_{ij}, i, j=x, y, z)\) by a centred finite difference method using Eq. 3.1. The strain tensor was further decomposed to \((\varepsilon_n)\) and shear strain \((\varepsilon_s)\), respectively.
5.2 Results and discussion

Microstructural evolution during the process of indentation will be presented, together with the velocity fields and strain maps determined by Digital Volume Correlation.

5.2.1 Grain motion

Figure 5.2 shows a typical sequence of longitudinally sectioned slices from the high speed X-ray tomography recorded during the indentation of a semi-solid Al-Cu sample at a displacement rate of 5 µm/s. Four different penetration depths \( I = 0, 90, 360 \) and \( 720 \) µm) are shown in Figure 5.2 (a), (c), (e) and (g). The Al-rich dendrites are darker grey and the Cu-rich liquid is white, resolved due to their X-ray attenuation differences. The initial equiaxed dendritic microstructure is clearly distinguishable in Figure 5.2 (a). This microstructure was held isothermally at 560 °C throughout the experiment at \( f_s \) of \(~72\%\). After 90 µm of indentation (Figure 5.2 (c)), the grains directly below the indenter moved slightly with the indenter, then the surrounding grains were forced to move, buffered by the thin layers of intergranular liquid. This indicates that a force chain was built up through the contacts of the grains. With further deformation (Figure 5.2 (e) and (g)), the grains rolled over each other, which appeared to trigger both translation and rotation of individual grains.

For instance, Figure 5.2 (b), (d), (f) and (h) show the isolated individual dendrites, which were used to perform the image registration algorithm to measure the grain movement. Through tracking the 34 dendrites in this slice (Figure 5.2) from \( I = 0 \) to 720 µm, the translation magnitude (Figure 5.3 (a)) and rotation angle (Figure 5.3 (b)) were measured. Although the system considered is three-dimensional, this measurement in two dimensions still can be used to clarify how grains arrange themselves as a result of imposed deformation. Since the solid fraction was relatively high, and dendritic grains were relatively large, the solid particles rotated only by a few degrees. For example, although dendrite A was moved by a
magnitude of $\sim100 \, \mu m$, it only rotated for about $3^\circ$ in anti-clockwise direction. The fact that grains moved differently from each other reveals the inhomogeneous nature of deformation.

Figure 5.2 Microstructural development of semi-solid Al-15Cu ($f_\alpha = 70 \%$) indented at displacement rate of $5 \, \mu m/s$ at an indentation depth of (a) and (b) $I = 0 \, \mu m$; (c) and (d) $I = 90 \, \mu m$; (e) and (f) $I = 360 \, \mu m$; (g) and (h) $I = 720 \, \mu m$ ((a), (c), (e) and (g) grey level images; (b), (d), (f) and (h) coloured individual dendrites).
5.2.2 Dilatancy effects

In addition to the observation of grain movement, the channels between grains appeared to change significantly, as shown in Figure 5.2. The opening-up of liquid channel (LC) can be directly observed in Figure 5.2, which is further illustrated in Figure 5.4 (a) to (d) where the same transverse cross-sectioned slice is shown at different indentation depth ($I = 0, 90, 360$ and $720 \mu m$). Although LC between second dendrite arms appeared to remain unchanged, the inter-dendritic space enlarged with increasing indentation depth.

In order to quantify the dilatancy effect, a medial axis method [201] was applied to the segmented LC. The resulting channel thickness distributions (colored according to the thickness) are demonstrated in Figure 5.4 (e) to (h). Initially, LC is rather homogeneously distributed (Figure 5.4(e)). With 90 $\mu m$ of indentation, the thickness of LC near the indenter surface started to increase by a small amount at this strain level. With more deformation ($I = 360 \mu m$), the opening of LC became obvious and propagated deep into the sample which appeared to grow vertically downwards. When the indenter is pushed even further ($720 \mu m$,}

Figure 5.3 (a) Translation amplitude and (b) rotation angle of individual grains at $I = 720 \mu m$ (dendrites with white could not be measured).
Figure 5.4 (g)), large inter-connected pocket-like channels appeared (red color). This suggests that when the deformation transferred through grains, certain grains moved preferably, causing some interstices increasing continuously overriding others. These giant inter-connected channels can be regarded as percolating paths linking void space [234], which consequently changes the permeability in the system.

Figure 5.5 presents cumulative distribution of LC thickness. Note due to the resolution limitation of X-ray tomography, thin liquid films less than 1 pixel (4 µm) could not be detected. The following key inferences can be made from the measurement of channel thickness:

1. The average thickness of LC increased only slightly, from a width of 37 µm at \( I = 0 \) µm to 50 µm at \( I = 720 \) µm. This is owing to the fact that LC between second dendrite arms, which represented a large volume fraction, remained unchanged during deformation.

2. The volume fraction of LC with thickness larger than 60 µm increased significantly from 4.7% to 17.4% during the 720 µm deformation. In the meantime, the maximum liquid channel thickness increased continuously, from 111 to 180 µm.

Although the grains only rotated slightly (Figure 5.3 (b)), considerable opening of inter-granular spacing was found, which is due to the complex dendritic structure of the specimen. Tomoya et al. [140] also observed that the liquid-filled channels increased greatly within the shear plane although there were only a few degrees of rotation of the solid particles.

The continuous dilation of liquid channels observed in this study was attributed to the dilatancy effect. Dilatancy is one of the fundamental microstructural responses of deforming granular materials [183,235]. The inhomogeneous dilation of liquid channel suggests that the propagation of deformation might have to follow optimized paths (percolating pathway) then
the preferred moving of grains was induced. The occurrence of dilatancy in semi-solid dense alloys subject to localized indenting deformation is relevant and essential to our understanding of solid-granular interaction.

Figure 5.4 (a) to (d) Transverse slices at the sample height of about 360 µm; (e) to (h) section view showing the distribution of liquid channel thickness at: (a) and (e) $I = 0$ µm; (b) and (f) $I = 90$ µm; (c) and (g) $I = 360$ µm; (d) and (h) $I = 720$ µm.
Figure 5.5 Cumulative LC thickness distributions (T is the liquid channel thickness).

5.2.3 Liquid migration

The opening of a liquid channel induced a low pressure in the region which subsequently caused the migration of liquid to the newly-opened inter-dendritic space. Figure 5.6 shows the variations of liquid fraction along the deformation axis at various depth of indentation. The deformation axis was normalized by the height of the sample. The liquid fraction was averaged over every 80 μm. As shown in Figure 5.6, the liquid fraction increased (the accumulation of liquid) in the upper part of the sample with increasing deformation, while there was depletion of liquid in the lower region. This was due to the stress/strain state of the region. Different stress states can modulate the flow of interdendritic liquid [233]. When the mush is under compressive stress, the porous solid phase can be compacted with liquid being driven out. On the other hand, when the mushy zone is sheared, the solid particles rearrange themselves (dilatancy), and draw liquid from the surrounding regions to compensate the increased volume.
Figure 5.6 Variation of liquid fraction versa the sample height.

Figure 5.7 Semi-solid Al-15wt%Cu at subsequent stages of indentation: (a) $t = 0$ s, $I = 0$ µm; (b) $t = 18$ s, $I = 90$ µm; (c) $t = 54$ s, $I = 270$ µm; (d) $t = 126$ s, $I = 630$ µm.

A small region (Zone D in Figure 5.6 (a)) shows an increase of liquid volume at the beginning of deformation then a reduction. This is due to the fact that when the indenter was
still away from Zone D, the force was transmitted to the grains in Zone D, shearing them apart. As the indenter traveled closer to this zone, more compressive deformation was imposed, thus draining the liquid out. Figure 5.7 shows the 3D section view of the tomographic volumes. The LC marked by the green arrow was open initially, but closed at the latter stage (from Figure 5.7(c) to (d)). The changes of deformation conditions can alter liquid flow tendency. This observation is important to understand the responses of semi-solid alloys under dynamic deformation conditions.

5.2.4 Grain deformation

In addition to grain movements, we observed the deformation of grains near the indenter. The dendrites below the flat surface of the indenter show significant changes as the indenter moving down (Figure 5.8 (a) to (e)). A dendrite tip (A) just underneath the indenter’s flat surface was highly compacted with increasing solid fraction. Another tip (B) showed similar behavior although this happened later than the tip A. As shown in Figure 5.8 (e), it seems that A and B were compressed together. Such formation of a solidifying zone underneath the indenter is contrary to a dead-metal zone observed when deforming solid samples or hard particles using a flat punch indenter [236,237]. The dead-metal zone is due to the no-slip condition present on the indenter surface, which keeps the material below the indenter’s flat surface fixed undeformed [236,237]. Here, the observation of a deformed/solidified zone beneath the flat surface of the indenter might be due to particular properties of the mushy zone, which are different from the hard particles: (1) liquid can solidify to solid if there are variations in the environment, in this case, a local compression force; and (2) the porous solid phase is soft at such high temperature hence it can be compressed together like a sponge which drives liquid out. Therefore, local compression of the solid matrix might cause solidification (a liquid to solid transformation) and meanwhile could drive liquid out of the compressive zone.
A few grains also showed certain degrees of deformation. One example is shown in Figure 5.8 (f) to (j). This shows that the principal dendrite arm was bent significantly. The force was transmitted to this single dendrite from the indenter, not only driving the dendrite to move, but also bending it mechanically which might generate variations in its crystallographic orientations. The bend happened at the latter stage of indentation when the dendrites became more entangled and uneasy to flow. Although the bending of dendrites in the mushy zone has been experimentally observed in several systems [74,213,238], current models of semi-solid deformation during solidification cannot simulate this phenomena [179,181]. The models developed by Yamaguchi and Beckermann [180,181] simulated the change of crystallographic orientation angle of a solidifying dendrite due to deformation, hence their model may present the potential for numerically modelling of the dendrite bending phenomena under such deformation conditions.

Figure 5.8 (a) to (e): Change of microstructure below the indenter’s flat surface; (e) to (i) the bend of dendrites during semi-solid deformation.

### 5.2.5 Velocity and strain field

In addition to the probe of the microstructural responses due to the deformation, the associated strain localization was quantitatively mapped out.
Figure 5.9 (a) to (d) show the progression of the velocity field at the start of indentation up to a depth of 720 µm as a series of quiver plots. The size (scaled up by a factor of 1.8 for the purpose of visualization in Figure 5.9) and direction of the quivers represent the magnitude and orientation of the velocity, respectively. The regions are moving away from the indenter radially, with materials close to the indenter moving quicker than those further away. The velocity at the beginning of indentation deformation is clearly larger than those developed at later deformation stages.

Figure 5.10 shows the average velocity magnitude of the material during indentation. The average of $v_{xyz}$ started from 1.2 µm/s, then decreased rapidly to 0.9 µm/s at 270 µm of
indentation. Afterwards, the drop of $v_{xyz}$ slowed down. $v_{xy}$ and $v_z$ show a similar trend. The speed of indenter ($5 \pm 0.09 \mu m/s$) was measured by tracking the location of the indenter tip in the tomographies. It was very close to the displacement rate ($5 \mu m/s$), conforming that the speed of the indenter was constant throughout the deformation. The slow-down of material flow velocity was not due to the speed change of the indenter but because that the materials become more resistant to flow as deformation proceeds.

Figure 5.10 Average velocity ($v_{xyz}$, $v_{xy}$ and $v_z$) within the sample during indentation measured by digital volume correlation

The octahedral normal strain ($\varepsilon_n$) was imposed with the velocity field in Figure 5.9. The positive values of octahedral normal strain represent the dilation (volume expansion) of the sample, while the negative values indicate the shrinkage. At a small amount of deformation ($I = 90 \mu m$), a large region of dilation with less than 2% of positive normal strain was clearly shown in Figure 5.9 (a). Negative $\varepsilon_n$ was prevalent in the regions away from the indenter, indicating the compression of solid skeleton. At the indentation depth of 180 $\mu m$ (Figure 5.9 (b)), the scope and intensity of dilation increased. Figure 5.9 (c) shows the $\varepsilon_n$ distribution with even more deformation ($I = 360 \mu m$). The dilated region became highly localized, with $\varepsilon_n$ larger than 3 located just below the indenter and it seems that the dilated region also transits
to a location slightly away from the indenter (region A). Figure 5.9 (d) shows that rather than a continually enhancement of positive $\varepsilon_n$ below the indenter, the positive $\varepsilon_n$ (amount of dilation) decreased and a small amount of negative $\varepsilon_n$ (compressive) appears under the indenter’s flat surface. The dilation in region A increased significantly with $\varepsilon_n$ larger than 4.

Figure 5.12 (a) shows the normalized histogram of the octahedral normal strain. It showed the increasing level of both volume expansion and compression of solid skeleton as deformation proceeded, although dilative strain was more prevalent than the compressive component.

Figure 5.11 shows the octahedral shear strain ($\varepsilon_s$) map. Unlike the distribution of $\varepsilon_n$ (Figure 5.9), it seemed that shear deformation became more and more intensified below the indenter when the indentation proceeded. A region of over 30% of $\varepsilon_s$ beneath the indenter can be observed in Figure 5.11(d). This narrow region of shear localization was different from the observations commonly found in semi-solid direct-shear deformation where the localized shear band had a width of a few grains [138,188,189].

With the combined application of time-resolved tomography and digital volume correlation, the evolving deformation zone localized in the vicinity of an indenter when indenting a semi-solid alloy was clearly unveiled. Mapping strain in situ is of great relevance to a wide range of applications from metal formation [236] to food processing [239]. Murthy et al [236] assessed the evolving two dimensional strain during indentation of a Cu sample by particle image velocimeter (PIV) where the surface textures were tracked. Waitukaitis and Jaeger [239] again used PIV technique but tracked the motion of foreign particles added into a dense suspension. The method, tracking the microstructural features inherent to the time-resolved X-ray tomographies via DVC, provides an evolving velocity and strain mapping in 3D.
can be applied to study the mechanics of various materials in harsh environments (e.g. mapping strain *in situ* in the vicinity of a high temperature fatigue crack tip).

Figure 5.11 (a) to (d): Octahedral shear strain map: (a) $I = 0$ to $90 \, \mu m$, (b) $I = 0$ to $180 \, \mu m$, (c) $I = 0$ to $360 \, \mu m$, (d) $I = 0$ to $720 \, \mu m$.

Figure 5.12 The normalized histogram of (a) octahedral normal strain, (b) shear strain.
5.2.6 Influence of deformation on porosity

The indenter was further travelled to a depth of 1500 µm, then the specimen was cooled down with the furnace whilst observations were performed. Figure 5.13 (a) shows the distribution of pores after the sample was deformed with 1500 µm of indentation (post-deformed condition). A few pores can be observed. The irregular pores were those present in the original sample but remained after the heating and deformation processes. One globular pore can also be found, probably a gas porosity formed during the isothermal holding period. Figure 5.13 (b) shows the pores in the solidified sample. There were many more pores in the solidified sample than the post-deformed sample. The number of pores increased from 33 to 479, while the total volume of pores increased from 0.056 to 0.106 mm$^3$. Although in the solidified sample, most of the pores in Figure 5.13 (b) were small in size, there were a few larger pores with irregular shape. The formation of those large pores was due to not only the growth of the existing pores in the post-deformed sample (Figure 5.13 (a)), but also the initiation and growth of fresh pores (marked by black arrows in Figure 5.13 (b)).

When the sample started to cool down, the Cu-enriched liquid began solidifying. It is well known that shrinkage pores could occur due to the density difference between the solid and liquid [85,240,241]. In our case, it appears that the locations where newly formed porosity was correlated with the dilated inter-dendritic liquid channels. The imposed deformation induced an uneven dilation of inter-dendritic space with liquid flowing-in. Upon cooling, the liquid in the dilated region would solidify at the same rate as their narrower neighborhood. Hence the liquid in the narrow channels would complete solidification first. The liquid then would flow from the dilated channels to the neighborhood to compensate for the shrinkage, thus reducing the liquid pressure in the dilated space, and then inducing the formation of pores there. This can be one possible mechanism via which pores form within the shear band of high pressure die casted components and sheared partially solid alloys [6,192].
Porosity distribution of (a) sample at 560 °C at an indentation depth of \( I = 1500 \mu m \), (b) in the solidified sample (yellow for pore volume smaller than \( 6 \times 10^5 \mu m^3 \), red larger than \( 6 \times 10^5 \mu m^3 \)).

5.3 Summary

Indentation is a well-known procedure to measure mechanical properties from hardness to creep of solid materials at a continuum level. In this study, we perform indentation of semi-solid specimen having complex dendritic microstructures. Using a novel thermo-mechanical setup, combined with 4D imaging (3D tomography plus time) and digital volume correlation, indentation of semi-solid/granular materials not only revealed deformation mechanisms at a microstructure level, (e.g. grain movement, dilatancy and liquid flow), but also measured the evolution of velocity and strain fields.
Chapter 6. Conclusions and future work

6.1 Conclusions

High speed synchrotron X-ray tomography was combined with three mechanical tests, (compression, extrusion and indentation), to not only observe and quantify for the first time the microstructural responses of a semi-solid Al-Cu alloy to a range of applied deformations. The microstructural mechanisms and dynamics observed were then linked to the macroscopic mechanical behavior of these semi-solid alloys.

The first goal of these series of studies was to clarify that dense semi-solid alloys (e.g. at f_s around 70%) can be considered as granular media (solid particles saturated in liquid). Granular materials display many distinct behaviors from continuum materials, a key one being the occurrence of dilatancy, where compression leads to localized shear and tensile forces. This study quantified the occurrence of dilatancy and its impact on defect formation during both isothermal semi-solid compression and indentation. It proved that dilatancy induced by deformation (localized shear and tensile strain/stress) could lead to liquid flow and damage formation. Just like granules (sands, for example), branched dendritic grains rearrange themselves in response to the subjected deformation, leading to the opening of interdendritic spaces. In alloys, the easy-flowing interdendritic liquid can initially migrate into the dilated interstices, but at a time when liquid feeding was no longer able to compensate the growing dilation, voids/damage form, tearing the liquid apart, and propagating along the dilated liquid channels. After solidification, shrinkage pores also tend to form at the dilated
interstices. This is schematically shown in Figure 6.1. The results also demonstrated that the extent of dilatancy is dependent on grain morphology; it is greater in the large dendritic specimen than in a fine globular one.

![Figure 6.1](image)

Figure 6.1. Schematic showing how shearing of semi-solid can induce dilatancy, leading to flow of interdendritic liquid, formation of damage and shrinkage porosity when the semi-solid alloy cooled down.

The flow of solute-rich liquid through equiaxed dendritic structures under deformation was also studied, demonstrating how the flow can be modulated by the state of deformation. In equiaxed dendritic structures, shear strain can cause dilatancy, which drives the interdendritic spaces open, thus increasing the permeability. Compressive stress/strain, on the other hand, can compact the solid phase (quite like a sponge), closing both intra-dendritic and inter-dendritic spaces with reduced permeability and subsequently squeezing the liquid out (Figure 6.2). This is the underlying mechanism responsible for the formation of liquid extrudate during squeeze casting.

The equiaxed dendritic grain structures behaved not only as granular materials, but changes in the intra-granular structure was also observed during semi-solid deformation. The individual grains underwent intra-granular deformation, leading to compaction, bending and fragmentation of dendrites as the grains interlocked, showing that deformation responses of semi-solid equiaxed dendritic structures is complex.
The full-field evolution of strain across the sample was measured in the semi-solid compression and indentation experiments via digital volume correlation. Compressive, dilative and shear strain were individually mapped out. Localization of different strain components was found and the transition between those zones was distinguished.

6.2 Suggestions for future research

6.2.1 Semi-solid deformation at high solid fractions and high strain rates

The three semi-solid deformation methods developed in this thesis have only been applied to one type of grain structure with a similar liquid fraction. Hence using the methods to compare the influence of different grain structures and solid fractions is recommended. High solid fraction (>90%) is particularly interesting, as the mushy zone at high solid fraction is supposed to be vulnerable and prone to form hot tearing.

Currently, the acquisition time of high speed X-ray tomography is still not short enough which restricts adopting this methodology at high strain rates. For instance, a displacement rate of 10 µm/s in the current semi-solid compression setup would cause enormous movement artefacts. It is hoped that the continuous advancement of X-ray detectors and acquisition techniques could further shorten the acquisition time, which will allow in situ 4D observation of semi-solid deformation at high strain rates.
6.2.2 Translating the experimental techniques to other semi-solid systems

In this study, a state-of-art methodology was developed to measure the rheological properties of multi-phase materials (solid, liquid and gas mixture), whilst at the same time tracking microstructural developments and mapping out the strain evolution. The approach is already being adopted within Prof. Lee’s group to study deformation responses of other multi-phase materials, ranging from magma to ice cream, demonstrating their applicability to a wild range of semi-solid systems.
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