NON-DENDRITIC STRUCTURAL EVOLUTION IN STIRRED Sn-15 WT.% Pb ALLOY MELTS

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Summary

Semisolid processing within the freezing range of metallic alloys has significant advantage over conventional die casting for near net-shape production of components. Rheological properties such as thixotropy and pseudoplasticity, essential for the processing in the semisolid route, originate from the non-dendritic structural evolution under melt stirring. Theories as varied as dendrite fragmentation by mechanical forces, continuous nucleation, dendrite remelting and cellular growth under melt shearing have been presented, but none of them provide a generalized explanation of various solidification structures formed under different melt shearing conditions. In this paper the structural evolution in Sn-15 wt.% Pb alloy is investigated utilizing different geometry of shearing devices and different shearing intensity. The experimental observations have been qualitatively corroborated by Monte Carlo simulation. It has been observed that the effect of melt shearing can be translated into the governing geometry of the diffusion field around the growing solid. Transitions from diffusive to turbulent flow through shear flow regimes then produce a subsequent morphological transition from dendritic to compact solidification structures via rosette morphology due to the corresponding change in the diffusion geometry in the melt.

Keywords: semisolid processing, solidification, microstructure, computer simulation

1 Introduction

Semisolid metal (SSM) processing between the liquidus and solidus, usually under mechanical or electromagnetic stirring, has received significant attention over the past 30 years due to its benefits over conventional die casting for near net-shape production of components [1,2,3]. The distinct rheological properties exhibited by semisolid slurries originate from the non-dendritic structural evolution during SSM processing under shear that also reduces porosity and segregation in the final product. The intensity of stirring and the time spent in the semisolid state determine the morphological evolution of the particles as equiaxed dendrites, rosettes or dense spheroids (often with entrapped liquid) under shear flow [1], and near mono-size fine spherical particles under high shear rate and intense turbulence in a twin-screw rheomoulding process [4].

In spite of the importance of morphological evolution during SSM processing, governing theories are very few and contradictory to each other. The most explicit theory so far is proposed by Doherty and coworkers who suggested dendrite fragmentation by the mechanical action of shear through a recrystallization mechanism...
followed by wetting of high-angle boundaries; particle crowding and the resultant diffusion field overlapping of individual particles were attributed to the non-dendritic structural evolution [5]. Recent 3-dimensional microstructural reconstruction in serial sectioned stir-cast structures seem to suggest that the dendritic fragmentation mechanism might not be as influential as believed and apparently fragmented particles in 2d microstructure are shown to belong to more complex 3d single crystals in reality [6]. Different other theories have been proposed but are somewhat restricted to individual growth patterns. Mullis proposed that dendrite tip bending due to thermosolutal convection alone can produce rosette morphology without invoking mechanical effects [7]. Hellawell, on the other hand, argued that dendrites are difficult to deform plastically by the mechanical action of shear and suggested that continuous nucleation in the absence of a distinct recalescence along with destabilization of diffusion geometry under shear promotes non-dendritic structure [8]. In an entirely different approach, Molenaar et al. proposed a cellular growth mechanism operative under melt shearing being responsible for rosette growth [9].

Most of these theories are restricted to simple shear flow and concentrate on specific microstructures rather than the multitude of microstructures observed under varied melt stirring conditions. The present work attempts a generalized explanation on the evolution of solidification structures as a growth phenomena depending on the nature of fluid flow prevailing under different stirring conditions.

2 Experimental

Stirring experiments were conducted on a model Sn-15 wt.% Pb alloy prepared from commercial purity (>99.8%) elemental metals. The alloy is melted and homogenized at 250 ºC inside a heated crucible and subsequently allowed to cool slowly to 195 ºC while being sheared using the stirrer at a fixed rotational speed. Temperature was monitored using two independent thermocouples inserted in the melt close to the stirrer. Different geometry of the stirrer such as plain cylindrical rod and a propeller with three blades were used to produce different stirring intensities. High intensity shearing was achieved using a co-rotating intermeshing twin-screw arrangement [4]. At predetermined temperatures, samples were collected from inside the stirring chamber, quenched in water, metallographically polished and examined under a Leica optical microscope. All the experiments were repeated number of times to ensure reproducibility of the results.

A Monte Carlo simulation was performed using a multi-particle diffusion limited aggregation approach to qualitatively corroborate the experimental results. A square lattice was randomly filled to represent a binary alloy. The atoms in the liquid were allowed to move randomly in diffusive motion and in a grossly directional manner under the influence of shear. Atomic movements in the liquid, attachment kinetics at the solid-liquid interface and the action of capillary force were incorporated in the model and solidification was seeded from an existing flat solid-liquid interface. The model concentrates on the growth features of the solid, which is the primary concern here and does not address nucleation in the melt. A detailed description of the model can be found in Ref. [10]
3 Results and discussion

Figure 1 presents the solidification microstructure of the Sn-15 wt.% Pb alloy quenched at 200 °C during continuous cooling from the liquid state under different stirring conditions. In the absence of melt stirring inside the same metallic crucible under the same heating and cooling environment as in the case of stirring experiments, large equiaxed dendritic growth of Sn-rich phase (white particles) is evident (Figure 1a). When the melt is sheared using a cylindrical stirrer operating at low shear rate (50 rpm) the structure is still predominantly dendritic albeit some coarsening is obvious as shown in Figure 1b. A serial sectioning of the sample confirmed that the dendrites are evenly distributed throughout the specimen and the apparently individual and clustered particles are connected in three dimensions to a larger dendritic stem. At higher intensities of stirring using the cylindrical rod, the microstructures observed are less dendritic but still complex and directional in nature. Microstructures of samples quenched during stirring using a propeller under low to intermediate stirring speeds (50-150 rpm) show an isotropic growth morphology of the solid as shown in Figure 1c. Individual rosettes can be observed evenly distributed throughout the microstructure.
with isotropic growth in all directions making them distinct from a dendritic structure, similar microstructure have been observed in all sections of the specimen in a serial sectioning experiment. When the melt is sheared in the twin-screw rheomoulding machine operating at a very high rotational speed of 1000, a microstructure consisting of very fine isolated and globular particles is observed as shown in Figure 1d.

From the experimental results it is evident that the nature of the stirrer governs the solidification microstructure. Different stirrer geometry was chosen to influence the nature of fluid flow created in the melt. A cylindrical rod was used to produce an overall low intensity simple-shear (laminar) type flow in the melt. A twin-screw machine, on the other hand, produces a complex flow behavior in the melt that can be characterized as largely turbulent in nature. A propeller creates a flow behavior in the melt somewhere intermediate between the two former stirrer geometries that is largely laminar with probably a chaotic flow pattern near the propeller blades where the intensity of shearing is very strong. To validate the hypothesis that the different structures evolved depending upon the nature of the fluid flow, Monte Carlo simulation results were produced and compared against the experimental observation.

It might be mentioned that in the stirred melt, the solid grows as isolated particles moving in the fluid and rotating under the action of the shear. Without the implementation of shearing, the particle morphology simulated resembles an equiaxed dendrite as shown in Figure 2a. The black regions in the figures represent the solid, while the solute and the solvent atoms in the liquid are shown in gray and white, respectively. The grayscale intensity in the melt represents the concentration of rejected solute (diffusion layer) around the solid-liquid interface. When a directional motion is implemented in the liquid (shown by the black arrowheads in the figures), a low intensity motion still produces a largely dendritic structure with well-developed primary and secondary arms (Figure 2b). On increasing the intensity of the shear flow (implemented by increased flow speed) coarsened solid

**Figure 2:** Simulated structures of isolated solid particles under (a) purely diffusive flow, (b) particle rotation under low intensity shear flow, (c) intermediate intensity shear flow and (d) turbulent flow.
A morphology resembling a rosette type structure evolves (Figure 2c). Under a turbulent flow pattern where the liquid atoms are allowed to impinge upon the solid from all directions, compact solid morphologies are observed as shown in Figure 2d.

The Monte Carlo simulation morphologies have resembled the solidification structures observed under the experimental conditions and suggest that the morphological evolution under melt stirring can be significantly influenced by the nature of the fluid flow. On the basis of the observations a growth influenced morphological evolution in sheared melts is suggested here.

Morphological development would depend on the geometry of the diffusion boundary layer around the growing particle, and therefore, the nature of fluid flow essentially governs the solidification morphology by influencing the characteristics of the diffusion boundary layer. Figure 3 presents a schematic illustration of the possible boundary layer geometries under different fluid flow conditions. In a pure diffusive flow (Figure 3a) solute transfer takes place by diffusion inside an infinite diffusion boundary layer in the absence of forced fluid flow. At low shear rate the flow can be visualized as essentially laminar with a finite boundary layer existing around the growing particle beyond which the melt becomes homogenized with respect to solute due to the imposed convection. Solute transport will still be dominated by diffusion inside the finite boundary layer and convective mixing will occur outside the solutal boundary layer assisting some coarsening. However, as the flow in the melt does not interact with the solid, the growth pattern will still be largely dendritic. At intermediate to high intensity of the laminar flow (and where a chaotic motion exists like near the blades of a propeller) the diffusion boundary layer will be further reduced and the growing solid particles rotating inside the flow will experience a periodic penetration of liquid inside the diffusion boundary promoting an alternate destabilizing and stabilizing effects on the solid-liquid interface thus producing rosette type morphology (Figure 3b).

Under turbulent motion in the liquid, the diffusion layer outside the solid-liquid interface will be completely destabilized and liquid penetration into the interdendritic interface will be completely destabilized and
region is likely at all times. This would prevent any possibility of constitutional undercooling and the solid-liquid interface will grow like a plane front producing compact globular particle morphology (Figure 3c).

4 Conclusions

Solidification microstructures in stirred Sn-15 wt.% Pb alloy seem to depend on the geometry of the stirring device and the stirring intensity used. A Monte Carlo simulation is performed to qualitatively corroborate the experimental investigation and based on the observation a growth induced morphological evolution is proposed.

It is proposed that dendritic growth is sustained in a low intensity shear flow, rosette morphology develops under particle rotation in an intermediate intensity shear flow and a compact solid morphology develops under high intensity turbulent flow. The effect of the nature of fluid flow on the solidification structure is explained on the basis of diffusion geometry maintained around the solid-liquid interface.

5 References