Controlled Nucleation Method: A New Process for Semisolid Metal Forming

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摘 要 Semisolid metal forming requires special feedstock material with a fine-grained and globular structure to achieve thixotropic properties. A number of methods have been developed to produce such feedstock materials. Controlled Nucleation Method (CNM) is a new and simple, cost effective method that has been developed by the University of Queensland. The CNM process does not use the conventional stirring process, instead, it uses solidification conditions to control nucleation, nuclei survival and grain growth, thereby produce fine and globular structures suitable for semisolid forming. No specialised equipment is required. The method can produce both semisolid slurry for rheocasting and semisolid billet for thixocasting. It can be applied to a wide range of alloys and can easily be incorporated into existing metal forming installation. Semisolid slurries/billets of hypoeutectic and hypereutectic aluminium-silicon casting alloys, aluminium wrought alloys and a magnesium alloys have been successfully produced.

关键词 Semisolid processing; solidification; semisolid feedstock; alloy microstructure

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1 Introduction

Semisolid metal forming (SSMF) is an emerging technology for near net-shape production of engineering components, in which metal alloy is processed at a temperature between its solidus and liquidus temperatures[1,2]. To be successful, SSMF requires special feedstock material with a structure of fine globular solid particles uniformly dispersed in a liquid matrix, to achieve thixotropic properties[3]. Such structures provide unique flow behaviour when sheared during the forming process[4], with minimal surface turbulence and splashing, and this smooth die filling assists in producing good microstructural integrity for the final products.

To develop this structure, loosely termed a thixotropic structure, external stirring is often applied during solidification of the melt, preventing growth with a dendritic morphology. Several methods have been reported to successfully produce the thixotropic structure in aluminium and magnesium alloys using stirring or forced convection. These include mechanical stirring[5], electromagnetic stirring[6], ultrasonic stirring[7] and, most recently, melt mixing[8]. The stirring methods usually involve extra equipment and complicated operation. This results in a significant cost increase for the feedstock, which is one of the main reasons that temper a wider application of SSMF. It has become more and more important to develop a simple, less expensive and easy implemented method to produce the semisolid thixotropic structure.

An alternative to the stirring process is solidification control[9~12]. During solidification, dendritic growth is affected by parameters such as nucleation, grain growth, solute redistribution, ripening and interdendritic fluid flow. The Controlled Nucleation Method (CNM) uses the control of certain solidification conditions, such as pouring temperature, cooling rate and grain refinement, to maximise grain nucleation and suppress dendritic growth, thereby producing a non-dendritic thixotropic structure for SSMF.

2 Process Description

This method was first developed in 1999 and initially named as Low Temperature Pouring[13~15]. Since then it was continually refined and further developed to become the Controlled Nucleation Method[4,16,17]. The main principle of CNM is to control solidification conditions to maximise grain density in the melt and promote grain growth in a non-or less-dendritic motion. The grain density again depends two factors: nucleation and nuclei survival. Usually the nucleation source is the heterogenous particles in metal. Constitutional undercooling ahead of the solid/liquid growth front make the particles heterogenous nucleation. Grain refiner is commonly added in many alloy systems to provide more heterogenous particles or/and make the heterogenous nucleation more favourable. It can be very effective for some alloys, but less successful in others, such as Mg(Al) alloys. Wall crystals provide another nucleation source that can be exploited. They are the crystals that are nucleated during pouring, at or near the relatively cold mould wall.

Formation of the wall crystals can be illustrated in Figure 1. There are two mechanisms. The wall crystals formed at the wall is supported by Ohno's Speration Theory[18]. It suggests that crystals first nucleate on the mould wall after pouring as the thermal undercooling on the cold wall is at a maximum (Figure 1 a). After a crystal has nucleated, growth occurs along the wall at first, where the temperature of the melt is lower. It then changes towards the melt as the diffusion of the solute rejected is more restricted along the wall. As the result, the crystal begins to take a necked shape. The wall crystals are subsequently detached or washed off by turbulence of the pouring and convection current in the melt. Formation of wall crystals near the wall is suggested in Chalmer's Free Chill Theory[19]. It indicates there is a thermally uncooled region adjacent to the cold mould wall (Figure 1 b), where the thermal undercooling is greater than the undercooling required for nucleation, $\Delta T_t > \Delta T_n$. Crystals nucleate in this region and then be transferred to the bulk of the melt.

(a) Ohno's Seperation Theory mechanism[18]

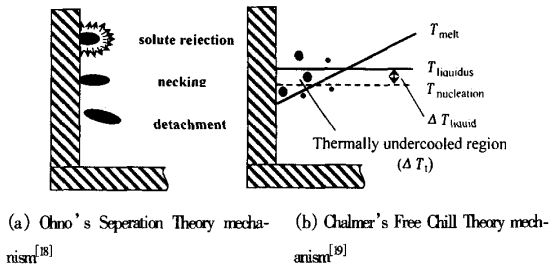(b) Chalmer's Free Chill Theory mechanism[19]

Figure 1 Schematic diagram of the mechanisms for wall crystal formation

Once the wall crystals are formed, they will be carried to the bulk of the melt either by residual momentum flows immediately after pouring or by thermosolutal convection, and serve as very effective nuclei. As the temperature of the bulk melt is higher than that at the wall region, crystal remelting occurs. The remelting rate is determined by the rate at which the local surrounding melt can supply heat. The crystals will decrease their sizes with time and some small crystals might disappear. The rate of nuclei survival becomes lower with an increase of melt superheat. High nuclei density combined with a slow cooling afterwards promote the ripening during the grain growth and lead to a globular structure of the primary solid.

The main features of the CNM are shown schematically in Figure 2. It can be broken down into four stages. First, molten metal with low superheat is poured into a steel mould. Second, wall crystals are formed and then carried into the bulk of the melt. If grain refiner is used, the heterogenous particles also serve as nuclei. It is possible to use extra mechanical or ultrasonic vibration to further assist the wall crystal detachment and transfer into the melt. Third, the metal can either cool slowly under a controlled cooling to the required solid fraction and serve as semisolid rheocasting slurry, or fully solidified as the semisolid thixocasting billet. The billet can latterly be reheated to the required fraction solid. Depending on the requirement for the formability of the semisolid slurry, isothermal holding can be applied to the reheating to further evolve the non-dendritic solid structure to a more globular structure for thixocasting. Finally, the semisolid slurry is transferred into a diecasting shot sleeve or a forging cavity to produce the semisolid forming components.
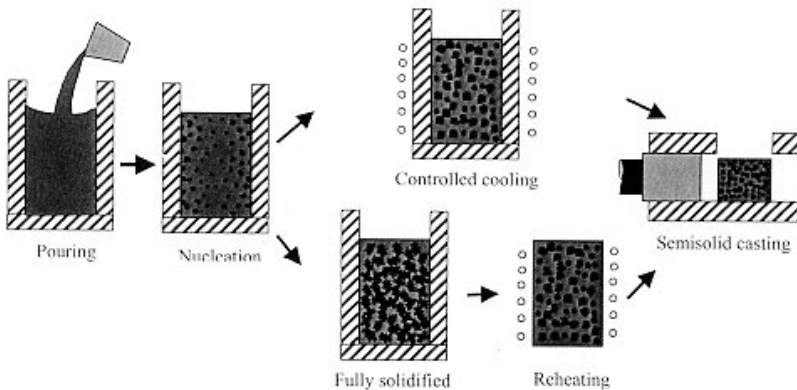

Figure 2 Schematic of the CNM process

The CNM process is simple and cost effective. It can be easily implemented in any foundry to produce their own semisolid slurry or billet. No specialised equipment is required. As it is an in-house operation, it overcomes the significant cost associated with the need to toll scrap and returns back to the feedstock supplier.

3 Contribution of the Wall Crystals

To determine the significance of the wall crystal on promoting the copious nucleation during solidification, a gauze

experiment was designed (Figure 3). Melt was cast into a cylindrical steel mould with a cylindric cavity 50 mm in diameter and 70 mm in depth. A stainless steel gauze cylindrical cup 30 mm in diameter was suspended in the centre of the cylindrical mould cavity with approximately 10 mm gap at the bottom. The gauze was made from stainless steel wires of 0.28 mm diameter with a hole aperture 0.56 mm square, serving as an effective mechanical barrier to the melt movement between inside and outside of the gauze.
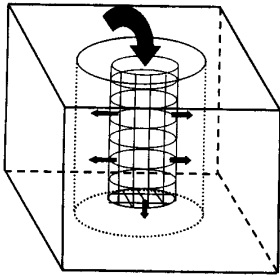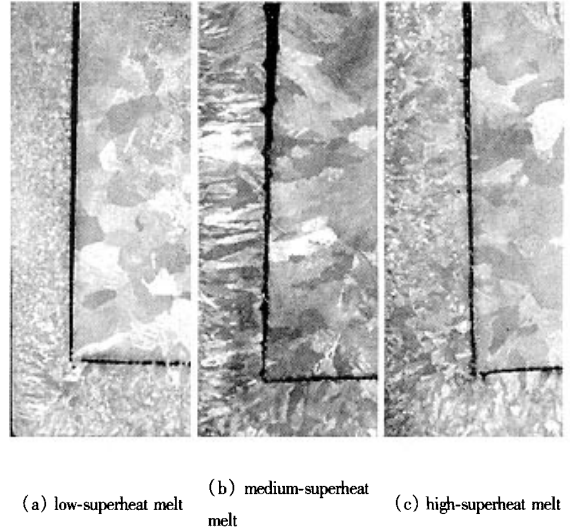

Figure 3   Gauze experiment

When the melt was poured into the mould, wall crystals were assumed to be formed at or near the mould wall. As the gauze prevented the crystals moving from the wall region to the centre of the melt, the wall crystals would be isolated in the region between the wall and the gauze. As the gauze heats up to the temperature of the melt very quickly during pouring, owing to its low thermal mass, it is reasonable to assume that of creation of wall crystals on the gauze would be much less likely than on the mould, and the number of crystals created by gauze would be quite small. Also with the slight insulating effect of the gauze and the relatively high temperature inside the gauze, these crystals tend to be remelted and make a limited contribution to grain formation. Therefore, the mechanism of wall crystals for microstructure formation would be not operative inside the gauze. The gauze experiment can to a large extent separate the contribution to grain formation from wall crystals and crystals formed by constitutional undercooling.

Figure 4 shows the macrostructures of the samples from the gauze experiment. A365 melt was poured inside the gauze with three different levels of melt superheat. The grain structures inside the gauze and outside the gauze were very different. The grains inside the gauze exhibited very coarse grains, while the grains outside the gauze were very fine, which indicated that wall crystals had a significant contribution to the microstructure formation under these conditions. The microstructure formation inside the gauze would be dependent on the heterogeneous nucleation on impurity particles, which had a much higher nucleation barrier than the wall crystals (in-

deed, the wall crystals were already nucleated crystals and the only barrier for the crystals becoming grains was the curvature undercooling that must be overcome for free growth). These heterogeneous nuclei gave rise to a much lower density of effective nucleation, thus a much coarser grain structure was formed.

(a) low-superheat melt   (b) medium-superheat melt   (c) high-superheat melt

Figure 4   Macrostructures of samples in gauze experiment. The wall is on the left side of each image and the centre-line of the sample is on the right

The grain structure outside the gauze varied with the melt superheat. It was very fine equiaxed at low-superheat melt, and changed to coarse equiaxed at medium level of superheat, then fully columnar grains for a high-superheat metal. This was because the wall crystals formed were still subjected to survival in the melt. The survival rate of the wall crystals would be much higher at a melt with low superheat. As the melt superheat increased, more wall crystals were remelted, resulting in a coarser structure. At the very high superheat melt, all of the wall crystals were remelted, a fully columnar structure was obtained between the wall and the gauze.

4   Experiments with CNM Process

The CNM process has been successfully applied to aluminium and magnesium cast alloys. The solidification condition used to control the nucleation and grain growth can be pouring temperature, cooling rate, mould temperature, mould thermal extraction, grain refiner addition.

5   Hypoeutectic aluminium-silicon alloy A356

A356 is a commercially significant alloy for SSMF. CNM

process has been applied to this alloy, and pouring temperature and grain refinement have been chosen to investigate the effect of the CNM process parameters on the microstructure of the semisolid materials. Figure 5 shows the semisolid microstructures of materials produced by CNM with different poring conditions. As the pouring temperature decreased from 725 ℃ to 650 ℃, the solid phase changed from a solid network to very irregular solid particles to uniform globular particles. For the 650 ℃-poured materials, the microstructure had an ideal globular structure with α-phase particles at a size of 100 μm uniformly distributed in the eutectic matrix. Nearly all of the eutectic was intergranular Figure 5 c). Figure 5 d is the combination of the low-temperature-pouring and grain refinement.

The material also developed a globular structure and actually it attained a globular state faster than the non-grain refined material. In A356, wall crystals made a significant contribution to the nucleation, theses wall crystals can only survived in the melt with low superheat, so low-temperature-pouring resulted in good globular structure. Grain refinement also made contribution to the nucleation and there was no nuclei remelting associated with this mechanism, however, the grain refinement itself was not sufficient to generate a thixotropic structure. The combination of low-temperature-pouring and grain refinement resulted in the materials more ready to develop into a globular structure and also widen the process window for the pouring temperature.

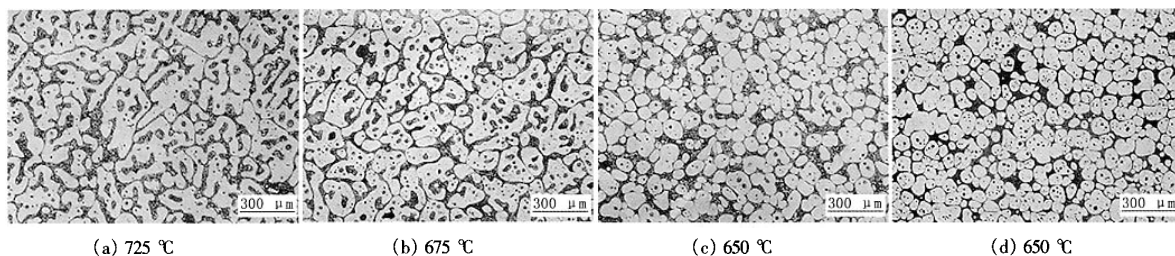| (a) 725 ℃ | (b) 675 ℃ | (c) 650 ℃ | (d) 650 ℃ |

Figure 5 Microstructures of the A356 materials produced by CNM with pouring temperature pouring combined with grain refinement

Semisolid casting was carried out using the semisolid structures produced, as shown in Figure 6. The high temperature pouring material only filled half of the die cavity (Figure 6 a). The die filling ability increased with the decrease of poring temperature. The 650 ℃-poured material almost completely filled the die except for one top corner (Figure 6 c). X-radiography revealed that the internal defects were significantly reduced compared to the 725 ℃- and 675 ℃-poured materials, however there was still some porosity in the first two steps. The grain-refined material resulted in a completely filled casting with all of the die details clearly visible (Figure 6 d). No defects were observed on the casting surface and the flow marks had become very faint. X-rays revealed a very sound casting free of internal porosity.

6　Hypereutectic aluminium-silicon alloy A390

Usually the cast microstructure of hypereutectic alloy A390 consists of large faceted primary Si phase and Al-Si eutectic phases. The CNM process can control heat extraction rate during solidification to prevent the "explosive" growth of silicon phase and produce the semisolid microstructure with the coexistence of primary silicon and primary aluminium. This coexistence is important for the formation of thixotropic structure because the amount of primary silicon by itself is insuffi-

cient for semisolid forming. Figure 7 shows the microstructure of A390 produced by CNM. The microstructure of the primary silicon structure was effectively controlled by AlP refinement and thermal refinement. AlP had a significant effect on size and morphology of the silicon phase. The size of silicon crystals in the AlP-refined materials was much smaller and more uniform than the non-AlP materials. Thermal refinement, which was resulted from the high cooling rate after the pouring, also had a strong effect on the primary silicon phase, comparing Figure7 a and Figure7 b. With the combination of AlP refinement and higher cooling rate, a very fine and globular structure was produced with silicon particles at about 20 μm and aluminium particles at about 60 μm (Figure7 d). Pouring temperature had no significant influence on the size and distribution of both primary aluminium phase and silicon phase.

7　Magnesium alloy AZ91

The CNM process has also been applied to magnesium alloys. Figure 8 shows the semisolid microstructures of magnesium alloy AZ91 with different pouring temperature and cooling rate. The sample poured at a temperature with higher superheat (125 ℃) had a very irregular solid phase (Figure 8 a), which was far from a globular structure and not suitable for semisolid processing. As the pouring temperature de-

creased, the solid phase gradually developed to a globular structure, Figure 8 c shows the microstructure poured at a lower superheat (10 ℃). The microstructure became very globular with a particle size of about 100 μm. Figure 8 d shows the

microstructure from the same superheat but at a higher cooling rate. The higher cooling rate resulted in a similar globular structure with slightly smaller solid particle size.
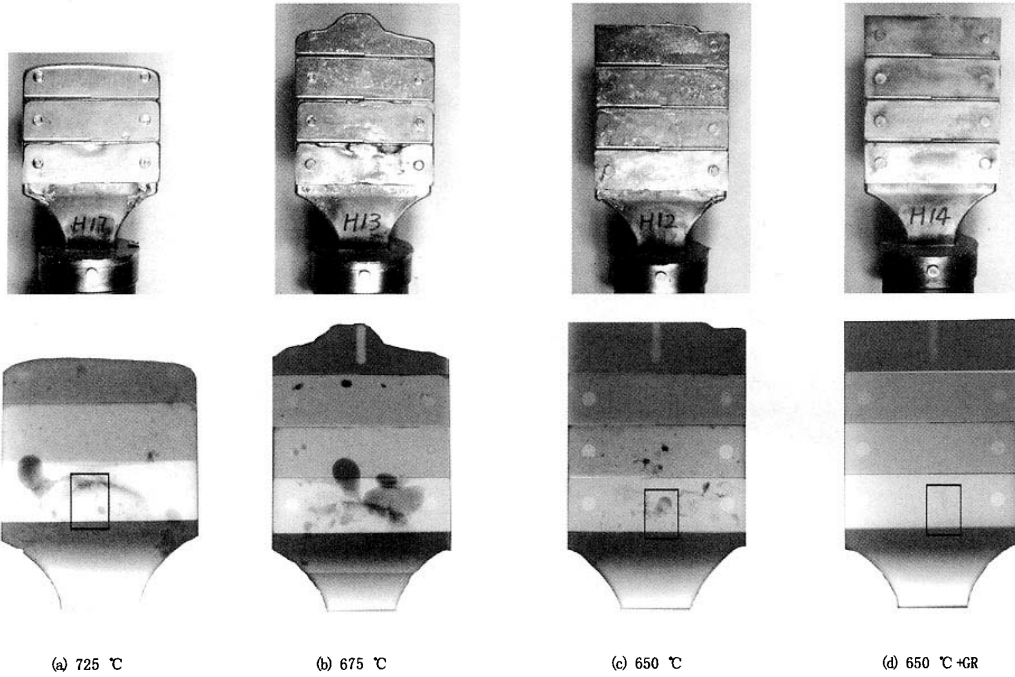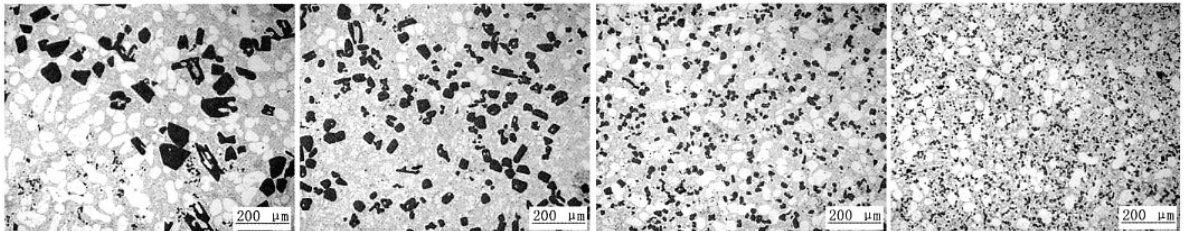

(a) 725 ℃   (b) 675 ℃   (c) 650 ℃   (d) 650 ℃ +GR

Figure 6   Semisolid castings and their X-radiographs; produced from CNM billets.
The billets were isothermally held at 580 ℃ for 5 minutes before casting

(a) no AlP-refinement and low cooling rate (b) no AlP-refinement and high cooling rate (c) AlP-refinement and low cooling rate (d) AlP-refinement and high cooling rate

Figure 7   Microstructures of A390 alloy produced by the CNM process
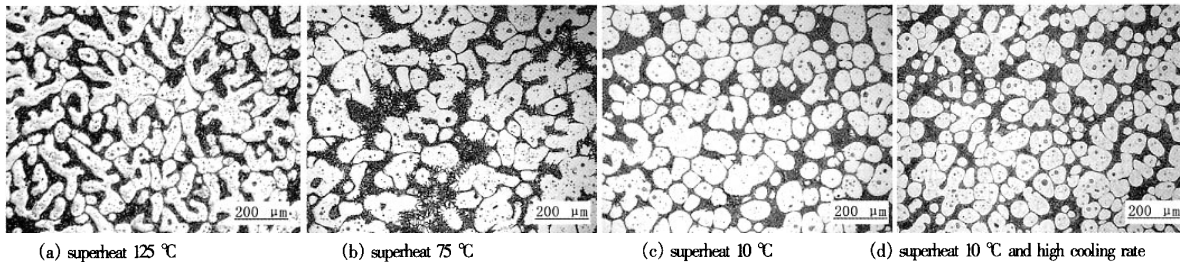
(a) superheat 125 ℃   (b) superheat 75 ℃   (c) superheat 10 ℃   (d) superheat 10 ℃ and high cooling rate

Figure 8   Microstructures of alloy AZ91 produced by CNM. The casting conditions are

8   Conclusions

A simple and easily implemented process for the preparation of slurry/billet for semisolid metal forming processes

has been developed. The CNM process uses the solidification conditions rather than stirring to control the nucleation and grain growth to produce globular structure. It can either produce semisolid slurry for rheocasting or semisolid billet for

thixocasting. The process can be applied to a wide range of aluminium and magnesium alloys. For different alloys, the process parameters which are chosen as the controlling factor to maximise the nucleation and nuclei survival can be different.

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