Centrifuge Melt Spinning—Merits and Limitations

Centrifuge melt spinning (CMS) is a recently proposed variant of rapid solidification processing. Mathematical modeling of the metal flow, heat transfer and solidification behavior in CMS shows that the new technique affords several distinct improvements to the production of rapidly quenched metallic ribbons by melt spinning, namely: enhancement of the heat transfer capability, better dimensional uniformity, and improvement in rapid solidification processing capability. The main limitation of CMS seems to be a propensity to increase microsegregation, as a result of the forced convection that leads to an increased fluid flow velocity during solidification. This limitation is compensated by the concurrent increase in solidification front propagation velocities.

INTRODUCTION

During the last two and a half decades, rapid solidification has become a major field of research activity in material processing. Although rapid solidification processing may involve many different techniques, the purpose is to impose high cooling rates during the solidification process.

The motivation for using high cooling rates lies in the resulting beneficial effects: more uniform and size-refined microstructures, and new classes of materials such as solid solutions with extended solid solubility, non-equilibrium crystalline phases and metallic glasses. The microstructural manifestations of rapid solidification stem from the growth rate (i.e., the solid-liquid interface velocity), which itself is connected to the extent of undercooling. The relative ability of each of the above-mentioned rapid solidification processes to achieve high cooling rates is related to their ability to enable high growth rates for the solid phase, by producing high undercooling at the advancing solid-liquid interface. The production of solid phases of uniform composition, with reduced micro- or macro-segregation of alloying elements, is indeed made possible in the rapid solidification regime.

Continuous rapid solidification casting methods generally involve a moving quench surface (most often rotating), on which the liquid alloy impinges. Acceleration is imparted to the liquid, resulting in efficient spreading of the stream, which then solidifies as a thin ribbon. Various modifications to this melt-spinning process have been implemented over the years, to improve the stability of the melt pool and to enhance the ribbon-substrate thermal contact.

Recently, a new variant, called centrifuge melt spinning (CMS), has been proposed. The novel feature of the new process is to mount the melt ejection system on a disc rotating coaxially with the chill surface, but in the opposite direction. The melt is ejected on the inner side of the chill surface.

THE CENTRIFUGE MELT SPINNING METHOD

Various melt spinning techniques are in use today. Generally, a fixed casting crucible is used, and the molten alloy is expelled by a gas overpressure. The melt impinges on a rotating surface and amorphous or microcrystalline ribbons or filaments are produced. Cooling rates commonly reach 10⁶ K/s. Generally, a fixed casting crucible is used, and the molten alloy is expelled by a gas overpressure. The melt impinges on a rotating surface and amorphous or microcrystalline ribbons or filaments are produced. Cooling rates commonly reach 10⁶ K/s.

However, there are some problems inherent to the technique that remain unsolved. When the quenching is performed on the outer side of the spinning roll, the short contact time may result in poor heat transfer between the melt and the substrate. Wetting of the casting rim by the liquid metal may be restricted due to the formation of an air boundary layer associated with the rotation of the rim. The rim-contact side of the solidified ribbons, therefore, shows a "wetting pattern," which provides information about the metal-rim interfacial characteristics. Both the wetting pattern and the thermal contact can be improved by increasing the ejection pressure. However, this yields, in the case of conventional melt spinning, an increase of the ribbon thickness and a lower cooling rate.

Other limitations are related to the use of high substrate velocities. The higher the substrate velocity, the thinner the ribbons. It has been shown that, at substrate velocities greater than 80 m/s, "catastrophic sticking" appears. The ribbon-substrate adhesion distance was found to vary, increasing in some cases with running time. Early parts of the cast ribbon leave the substrate at higher temperatures than the later parts in the same run, resulting in a longer contact.
distance. Welding of the surface substrate may even occur. The temperature of the casting surface increases as the melt puddle is ejected on it, to an extent at which enhanced melt wetting may occur. Magnetic and physical properties measured on a ribbon sample taken from a long and a short run under the same processing conditions differ substantially. This limits the use of a high substrate velocity in conventional melt spinning.

In some variants of melt spinning, the melt puddle is unconstrained. It can, therefore, distort and vibrate, as reported and calculated in Reference 18. Variations in the thickness and width of the ribbons, both along and across the casting direction, are common. Dimensional non-uniformity may also cause variability in the magnetic properties (amorphous) ribbons. Hence, vibrations and distortions of the melt puddle should be avoided as much as possible.

The new CMS device was first developed as a prototype that produces small quantities of melt-spun ribbons. Based on the experience gained on the prototype apparatus, a continuous casting device has been designed (Figure 1). Unlike conventional melt spinning, CMS uses a rotating disk holding the casting crucible. The copper rim which serves as the quenching substrate rotates coaxially with the disk, in the opposite direction. The rotational velocities of both disk and rim can be adjusted, as can the crucible orifice diameter and distance from the rim. Casting can be performed in a controlled atmosphere. The melt is extracted coaxially from the crucible by a centrifugal force, and impinges the inner surface of the copper rim. The volumetric flow rate of the melt is controlled by the crucible diameter and its spinning velocity.

As a first step in the goal of assessing the merits and limitations of CMS, a detailed parametrization study has been performed, using a binary Al-Ge alloy. The process variables were checked as to their influence on ribbon dimensions and other ribbon characteristics such as the wetting pattern (the roughness of the rim-contact side), the cooling rates as derived from the secondary dendrite arm spacings, and the occurrence of metastable alloy phases in the Al-Ge system. In a second step, a mathematical model for the process was derived, with special attention to the hydraulic features peculiar to CMS, in addition to the heat flow characteristics in the solidifying melt. As a result of the mathematical modeling, effective heat transfer coefficients have been evaluated and shown to be process dependent. The model accounts for the drawing mechanisms caused by the counter-rotation of the casting crucible and the quenching substrate, the additional velocity imparted to the melt due to convection, and the liquid viscosity changes with temperature during ongoing solidification.

### PROCESS FEATURES

The process parameters of CMS are: the crucible tangential velocity (Vc), which controls the ejection pressure (P_e); the ejection velocity (Ve); the volumetric flow rate (Q); and the Reynolds number (Re) for the flow; the copper substrate tangential velocity (Vs); the diameter of the crucible nozzle (d); the nozzle-substrate distance; and the melt superheat. The melt extraction velocity (Ve) is the sum of Vc and Ve. The range of values for the process parameters actually used, and the resulting Al-Ge ribbon characteristics, are given in Tables I and II.

The range for the apparent cooling rates developed on the rim-contact side of the ribbons has been calculated as a first estimation from secondary dendrite arm spacing (DAS) measurements. These were performed on fractured ribbon surfaces slightly etched, from regions in the ribbon where the growth of dendrites permitted the development of secondary arms, using the empirical power relationship for Al-Cu and Al-Si alloys:

\[(DAS) \cdot (CR)^{0.333} = 50 \mu m (K/s)^{0.333}\]

where CR is cooling rate. Several ribbons did exhibit a degenerated dendritic microstructure, which is generally admitted to be a result of higher cooling rates. Cooling rates higher than 10^6 K/s are uncommon in conventional melt spinning. From the CMS measurements, it would appear that cooling rates are enhanced by almost two orders of magnitude, a major merit of CMS.

Mathematical modeling of the metal flow, heat transfer and solidification behavior in CMS has stressed five facts.

### One

The heat transport in CMS is not of pure Newtonian nature, nor is it ideal. The criterion for determining which type of cooling prevails is the value of the Biot number (Bi): h_th/k_s. The letter h represents the heat transfer coefficient at the interface, th is the ribbon thickness, and k_s is the thermal conductivity of the ribbon material.

Cooling is essentially ideal when Bi > 30, and is Newtonian when Bi < 0.015. The average interfacial heat transfer coefficients, considered as stemming from a constant resistance heat transfer process, have been calculated by computing the ordinate intercepts of empirical curves of the ribbon thicknesses vs. squared root of contact time. As such, they range between 1.5 x 10^6 W/(m^2K) and 4.3 x 10^6 W/(m^2K), by a half to one order of magnitude higher as those assumed for copper substrate in conventional melt spinning.

### Table I. Range of Process Parameters for Al-Ge Ribbons Produced by CMS

<table>
<thead>
<tr>
<th>V (m/s)</th>
<th>P_e (KPa)</th>
<th>V_e (m/s)</th>
<th>Q (cm^3/s)</th>
<th>Re (x 10^5)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.25-25</td>
<td>1.8-269</td>
<td>0-78</td>
<td>1.3-5.0</td>
<td>5.3-23.8</td>
</tr>
</tbody>
</table>

### Table II. Characteristics of Al-Ge Ribbon Produced by CMS

<table>
<thead>
<tr>
<th>Width (mm)</th>
<th>Thickness ((\mu m))</th>
<th>DAS ((\mu m))</th>
<th>Cooling Rate (K/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.98-3.23</td>
<td>15-87</td>
<td>0.30-0.11</td>
<td>10^6-10^7</td>
</tr>
</tbody>
</table>

In this first order calculation, the peculiar flow nature of the melt in CMS was not taken into account. Taking \(h = 200 \text{ W/(m-K)}\) (for solid Al), and as ribbon thicknesses varied between 15 \(\mu m\) (obtained at high h values) and 80 \(\mu m\) (obtained at low h values), Biot numbers in CMS are in the range 0.1 < Bi < 2.0. It can be concluded that CMS works in the intermediate cooling regime. In fact, as will be shown later, the temperature profiles are generally quite different from those reported for the intermediate regime, because of the phenomenon of recalescence: rapid heating of the undercooled solidifying ribbon due to the desipation of the solidification latent heat.

### Two

Heat transport in CMS is directly affected by the increase of both the centrifugal forces that cause the ejection of the liquid melt onto the quenching substrate and that ensure prolonged contact of the solidifying ribbon on the heat extraction sink. Increasing the ejection pressures from 2 to 270 kPa causes the heat transfer coefficients to increase by a factor of 3.

### Three

Conversely to conventional melt spinning, two additional phenomena contribute to the heat transfer characteristics in CMS at high extraction velocities: forced convection and mechanical drawing of the melt. These phenomena are inherent to the fluid flow nature in CMS.

For constant crucible and quenching rim radial velocities, the volumetric flow of the liquid melt remains constant, as does the melt extraction velocity. The ribbon cross-section (which is the ratio of the two precedent variables), therefore, remains constant as well. As a result, the liquid metal stream ejected on the rim spreads only in the lateral direction, increasing the initial width of the ribbon (and reducing the ribbon thickness), until completion of solidification.

The value of the lateral spreading velocity is time dependent, due to the melt viscosity changes with decreasing temperature, and due to an additional centrifugal component added to the lateral velocity as a result of the quenching substrate spinning motion. Moreover,