# Analysis of process limits for continuous thixotropic slurry casting

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A criterion of stircastability is formulated, which relates the specific process limits for semicontinuous stircasting (rheocasting or thixotropic slurry casting) to alloy properties, and which can be used to decide which alloy compositions can be continuously stircast in a given apparatus. It provides a better understanding of the casting rate behaviour of alloys in continuous stircasting. For a specific type of apparatus, described in this paper, it predicts an upper and a lower limit of the nominal alloy composition, for continuous stircasting of thixotropic slurries. In practice, this means that eutectic or close to eutectic alloys cannot be stircast in this apparatus, and that stircastability decreases toward smaller freezing ranges.

# 1. Introduction

Stircasting (also termed rheocasting) is a casting process, in which semi-solid metal slurries are cast at temperatures in the liquidus-solidus region under mechanical stirring. This is illustrated in Fig. 1. Since about 1970, Flemings and Mehrabian and many other workers [1-5] performed extensive investigations on the behaviour and structure of semi-solid metal slurries. Two of the most important observations were: (i) when metal alloys are vigorously agitated during solidification, the solid which forms has a special nondendritic structure; (ii) semi-solid metal slurries behave as thixotropic fluids.

Recently, stircasting apparatus was developed for the semicontinuous production of thixotropic semi-solid metal slurries. The aim was to investigate casting rate behaviour of such slurries, and to see if the method is adequate for the continuous slurry production prior to other casting processes, e.g. diecasting. In the experiments which have been performed, a dependence of the casting rate on alloy composition was noticed, for several alloys [6-8]. In an attempt to explain this observation, it occurred that by analysis of the process limits one is able to define the composition limits for this particular method of continuous slurry casting. The authors believe that the analysis is general in principle, but that the composition limits in other methods may vary due to different specific process limits.

# 2. Mouldless stircasting of bars

In recent publications, basically two methods of stircasting have been introduced: (semi-)continuous stircasting [2-4] and so called "batch-stircasting" [5, 9–15]. It is worth noting that in "batch-stircasting", the semi-solid slurries are not actually cast, but just stirred during partial solidification, and then quenched at a given volume fraction solid [5, 12]. It is possible to produce "ingots" via batch-stircasting. This may be done simply by pouring an isothermally stirred slurry into a cold mould, as sketched in Fig. 2.

A method of continuous slurry casting introduced here, is the mouldless stircasting of bars. It is a somewhat modified version of continuous rheocasting, developed by Flemings and Mehrabian, and many others [2--4]. The casting unit is shown in Fig. 3a. The modification consists of a chill block, shown in Fig. 3b, and a displaceable stirrer. In this chill-block, a temperature gradient was created by means of an airflow. When the stirrer is moved upward, a narrow gap forms, through which the liquid emerges while it is brought into the semi-solid state. In this manner, it has been



Figure 1 Schematical representation of the stircasting process: (a) the annulus (chill-block), in which liquid metal, under mechanical stirring is cooled into the solid-liquid region; (b) conventional, and (c) typical nondendritic stircast structure.

possible to produce a cast bar without the use of a mould, as is illustrated in Fig. 3a. Products of Pb-30% Sn and Al-4% Cu are shown in Fig. 4.

The production of such bars requires a slurry flow which is constant within a certain tolerance. To ensure this it is necessary to control the (apparent) viscosity of the slurry. Since the apparent viscosity increases progressively as a function of the volume fraction solid, the temperature control in the chill-block is extremely important in order to maintain a constant slurry flow. Note that the apparent viscosity of a thixotropic slurry can be made to decrease at a constant fraction solid by increasing the stirring speed [5]; however use of this property of semisolid slurries would have required a more advanced process control which was not available to the present study.

The advantage of a solid bar is that it can be used both for die casting and directly for extrusion as well. However, in the process of mouldless stircasting under gravity, the casting rates are actually small. Therefore, this process is not adequate if large amounts of slurry are required.



Figure 2 Illustration of (a) and (b) "batch-stircasting" and (c) and (d) "thixocasting". (a) Isothermal holding of semisolid slurry, (b) ingot-making, (c) ingot sectioned into charges, which are reheated prior to diecasting, (d) die casting.

## 3. Process analysis

#### 3.1. General process limits

It appeared essentially that a constant slurry flow in mouldless stircasting is more easily obtained with alloys with long freezing ranges. The question arose which are the criteria for continuous stircasting and how these determine which alloy compositions can be used.

In any continuous slurry casting method, the prime aim would be to produce semi-solid slurries of reproducible properties. Any particular stircasting application may require specific slurry



Figure 3 Mouldless stircasting of bars: (a) casting unit and (b) chill-block.



characteristics. The question to be solved generally, is to design a process technique which meets the requirements in a particular application.

In principle, the morphology of the cast structure of any alloy composition is expected to change under the effect of stirring during solidification. For example in d.c.-casting, an extended equiaxed zone may be obtained by agitation in the mushy zone [16–18]. No specific properties of a semi-solid slurry are required here. In the process of thixocasting [4], i.e. the die casting of thixotropic metal slurries, it is required that the slurries are thixotropic. Joly and Mehrabian [5] found that Sn–15% Pb alloy shows no measurable thixotropy below a solid volume fraction of about 30%. It is assumed here that this is a lower limit



Figure 4 (a) Stircast bars of Pb-30% Sn; diameter 10 mm, length 500 mm. From left to right: (i) the period of the temperature wave in the chill-block, and the corresponding variation in viscosity, is reflected in the diameter of the bar; (ii) a bar, hollow over the total length; hollow bars were obtained with small vertical displacement of the stirrer during casting, i.e. with a small gap in the chillblock; (iii) bar diameter decreasing toward the top; (iv) liquid metal emerging from holding furnace ended this experiment; (v) deviation from the vertical axis normally leads to liquid emerging from the stircaster, as in (iv); (vi), (vii), (ix) under optimum conditions, the mouldless semicontinuous stircasting of bars can be a stable casting process, and the bar diameter can be held constant; (viii) thinner bars were obtained at a higher rate of descent, ridges in the bars are the result of a systematic variation in the rate of descent, which was caused by imperfections in gear. (b) Stircast bars of Al-4% Cu; diameter 25 mm, length 300 mm.

for thixotropy, which may be expected for all alloys discussed in this paper. In the case where thixotropic semi-solid slurries are to be transported, under gravity, or external pressure, there is also a maximum to the volume fraction solid  $f_s$ , since the (apparent) viscosity increases progressively as a function of  $f_s$  [5].

In short, in thixotropic slurry processing the general process limits are a lower and an upper limit to the volume fraction solid, with which a thixotropic slurry can be produced. As the specific process limits may vary with different methods, we will have to confine ourselves to one particular method, for which these are to some extent known, i.e. the method described in the previous section.

## 3.2. Specific process limits

The specific process limits are determined by the actual apparatus and the available facilities, and by the properties of the materials used. The process of continuous slurry casting works within a certain range of solid fractions which is determined by the degree of process control.

By the method described in Section 2, the slurries were produced under gravity. This is a specific limitation of vertical slurry casting, because the maximum allowable solid fraction with which the slurry may emerge is necessarily smaller than when an external pressure is applied. In this study, the maximum solid fraction with which slurries could be produced is not quite certain, but must be around 0.55. Another specific limitation of this particular method of slurry casting is a somewhat increased lower limit with which slurries emerge from the apparatus. This is because the cast bar will not have enough rigidity at solid fractions lower than about 0.35 [6]; it will collapse or bend away from the vertical axis, resulting in the emergence of liquid metal. Examples are shown in Figs. 4d and e. The fact that the casting rate is sensitive to the viscosity of the slurry is seen in Fig. 4a: here, the temperature wave in the chill-block has become visible as a periodic change in the diameter of the bar (about 20 to 30 mm). Note that the small ridges in each bar are due to a systematic variation in the rate of descent, which was caused by imperfections in the gear.

Until now, alloy properties have not been taken into consideration. To illustrate the influence of thermodynamic alloy properties, let us consider the  $T(f_s)$  curves of an arbitrary binary alloy, as shown schematically in Fig. 5. In this figure,  $T_e$  is the eutectic temperature,  $T_m$  is the melting point of the pure base metal, and  $f_s$  the volume fraction solid.  $C_0^i$  with i = 1, 2, ... are the nominal alloy compositions.  $T(f_s)$  curves can be calculated using the Scheil equation, written in the form [19]:

$$f_{\rm s} = 1 - \left(\frac{C_{\rm L}(T)}{C_0}\right)^{\frac{1}{k_0 - 1}} \tag{1}$$

The stircasting apparatus is operated in a region of

solid fractions between  $f_s^{\min}$  and  $f_s^{\max}$ , as indicated in the figure. At low nominal alloy content,  $C_0$ , a small temperature fluctuation corresponds to a large variation in the volume fraction solid, and to large changes in the apparent viscosity. Viscosity data for semi-solid aluminium alloys are not available, but as measured for Sn-15% Pb [5], the apparent viscosity increases from about 0.50 to 7 Pa sec (5 to 70 poise). Given the temperature fluctuations it will be difficult to maintain a constant slurry flow. With fluctuations to lower temperatures, there is a continuous risk that the semi-solid slurry freezes in the chill-block. During an accidental upward temperature fluctuation, there is the risk of emergence of liquid metal from the stircaster. As the absolute slope of the  $T(f_s)$ curve decreases, it is increasingly difficult to control the fraction solid. At high nominal alloy content, temperature fluctuations cause a much smaller variation in the solid fraction. Consequently, the viscosity and the flow rate are more easily kept constant.

A limiting  $|dT/df_s|_c$  value is directly related to the degree of process control. The latent heat extracted from a volume element of fluid, travelling through the gap can be written as:

$$q = \rho_{\rm s} H \frac{\partial f_{\rm s}}{\partial t} = \rho_{\rm s} H \dot{T} \frac{\partial f_{\rm s}}{\partial T} \qquad (2)$$

in which  $\rho_{\rm s}$  is the solid density, H is the heat of fusion,  $\dot{T}$  is the local cooling rate, and q is the heat flux. Although  $\dot{T}$  in this equation is actually undefined, because the temperature distribution in the chill-block is not steady, the formula illustrates that in a process controlled by temperature, an accidental delay in the cooling turn-off may easily cause too large a change in the fraction solid. This is not unlikely with an alloy at the critical  $|dT/df_s|_c$  value, particularly because temperature waves travel relatively slowly through the system. The limiting  $|dT/df_s|_c$  value determines the minimum nominal alloy composition which can be used. The maximum alloy composition is determined by the  $T(f_s)$  curve which is just in the work region of solid fractions for this particular method. In other words, knowing the specific process limits of the slurry casting process, it is possible to define regions of optimum stircastability in different alloy systems.

A criterion which defines the composition limits for the process of continuous slurry casting may now be formulated:



Figure 5 (a) Volume fraction solid as a function of temperature, for different nominal alloy compositions  $C_0^1 < C_0^2 <$  $C_0^3$ . With increasing nominal composition, a temperature fluctuation causes a decreasing variation in the fraction solid. At close to eutectic compositions, such as  $C_0^3$ , the minimum fraction solid required for thixotropy cannot be attained.

(i) Dependent on the type of stircasting apparatus and the specific process limits, a range of fractions solid can be defined, within which thixotropic semi-solid metal slurries can be produced at a constant average rate; the maximum stircastable alloy composition is determined by the  $T(f_s)$  curve which just covers this range,

(ii) Depending on the degree of process control, a minimum  $|dT/df_s|_c$  value can be determined, experimentally, which defines a minimum stircastable alloy composition.

### 4. Experimental results

Experiments have been performed using the alloys listed in Table I [6-8]. It is worth noting that average casting rates depend on the geometry of a particular chill-block, and are therefore not relevant to the present analysis.

In the Al-Cu system, it is observed that with decreasing alloy content, it is increasingly difficult to cast bars. More and more attention is required to control the temperature in the chill-block. The following result may illustrate this.

The left half of Fig. 6 schematically shows the stirrer/chill-block configuration used for the continuous slurry casting of Al-13% Cu, Al-10% Cu and Al-4% Cu. The chill-block and holding furnace are connected by means of flanges. Due to the temperature gradient in radial direction, these flanges act as cooling ribs. As a consequence, the aluminium alloy in the holding furnace will solidify against the flange, if no extra heat is supplied from the top or bottom of the flange.

The liquid stream into the chill-block is then prevented. With high copper content, however, i.e. as in Al-12% Cu and Al-10% Cu, the casting of bars succeeded very well, whereas with Al-4% Cu the temperature control was troublesome, and bars could not be obtained.

By changing the geometry of the stirrer it was possible to bring about a layer of  $Al_2O_3$  in the flange angle; this is shown in the right part of Fig. 6. Now, it appeared that the temperature could be controlled within a smaller interval (± 1.5 K) and that the slurry-casting of Al-4% Cu was without particular difficulty. However, with this new configuration, stircast bars of Al-2% Cu could not be obtained.

A second result which is relevant to the process analysis in the previous section, are the results of experiments with the ternary alloys

Т	A	В	L	E	I	Alloys	used
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Alloy	Stircastability
Al-2% Cu	not good
Al-4% Cu	good
Al-10% Cu	good
Al-13% Cu	good
Al-10% Mg	good
Al-4% Si	good
Al8% Si4% Cu	not good
Al-4% Si-4% Cu	good
Pb-30% Sn	good
Pb-40% Sn	not good



Figure 6 Chill-bldck modification. (a) Left, first version, not suitable for continuous casting of Al-4% Cu, due to radial heat flow through flanges, and (b) right, modified geometry with  $Al_2O_3$ -layer to reduce heat flow through flanges; suitable for continuous casting of Al-4% Cu.

Al-8% Si-4% Cu and Al-4% Si-4% Cu. It was not possible to create a constant slurry flow with Al-8% Si-4% Cu, which is easily explained by considering the equilibrium diagram, see [20]. At the eutectic temperature, the fraction solid is about 0.25. In the present method of mouldless stircasting this solid fraction is too small to prevent the emergence of liquid material from the apparatus; the viscosity of the slurry is simply too low. This fraction solid is also below the lower limit of thixotropy for Sn-15% Pb alloy [5], which gives some reason to suspect that Al-8% Si-4% Cu with a solid fraction as small as 0.25 cannot be produced as a thixotropic slurry.

A reduction of the silicon content to 4% increases the solid fraction at the eutectic temperature to about 60%. The casting of bars with Al-4% Si-4% Cu alloy was without particular difficulty.

Other alloys used were Al-10% Mg, Al-4% Si, and Pb-4% Sn. With the latter, stircast bars were not obtained, probably because the fraction solid at the casting temperature is too low, as with Al-8% Si-4% Cu.

### 5. Discussion

The results of the experiments with Al-4% Cu can be used to determine the limiting  $|dT/df_s|_c$  value mentioned in Section 3.2. With the improvements to the geometry of the stirrer and the chill-block, as described in Section 4, the critical slope of the  $T(f_s)$  curve was taken as the average absolute slope between  $f_s = 0.35$  and 0.55 of the  $T(f_s)$  curve of Al-4% Cu. The range  $0.35 < f_s < 0.55$  is chosen somewhat arbitrarily. The lower limit is based on the assumption that slurries must be thixotropic and must have some rigidity to produce a bar. The upper limit can be estimated from many microstructures. It follows that  $|dT/df_s|_c \approx 30 \text{ deg}$  $(\text{wt }\%)^{-1}$ .

It is now possible to define regions of optimum stircastability in different alloy systems. To this purpose, quantitative fractions of solid against temperature,  $T(f_s)$  curves, are required, which here, were calculated using second order polynomials for the solidus and the liquidus, determined by the least squares method, applied to solid and liquidus measurements [21-28].

Differentiating Scheil's equation, written as [19]

$$f_{\rm L}(T) = \left(\frac{C_{\rm L}(T)}{C_0}\right)^{1/(k_0 - 1)}$$
(3)

neglecting the term involving  $dk_0/dT$  (see Appendix), and substituting for  $C_L(T)$ :

$$C_{\mathbf{L}}(T) = a_{\mathbf{L}}T^2 + b_{\mathbf{L}}T + d_{\mathbf{L}}$$
(4)

in which  $a_{\mathbf{L}}$ ,  $b_{\mathbf{L}}$ , and  $d_{\mathbf{L}}$  are constants, we obtain

$$\frac{\mathrm{d}f_{\rm s}}{\mathrm{d}T} = \frac{2a_{\rm L}T + b_{\rm L}}{C_0(1 - k_0)} \left(1 - f_{\rm s}\right)^{2 - k_0} \tag{5}$$

As an example, the absolute reciprocal value  $|dT/df_s|$  as a function of  $f_s$  is shown in Fig. 7, for Pb-Sn. It is seen in this graph that the curve for Pb-12% Sn is just above the critical  $dT/df_s$  value of 30 deg(wt%)<sup>-1</sup> so that this will be the minimum tin content with which Pb-Sn slurries can be produced by the method described in Section 2.

In this manner, the regions of stircastable compositions were determined for the binary systems Al-Cu, Al-Mg, Al-Si, and Pb-Sn. These are shown in Fig. 8. Table I shows that the binary alloy compositions are all in these regions except for Al-2% Cu and Pb-40% Sn, for which stircast bars could not be obtained. Unfortunately, more experimental evidence supporting the critical  $|dT/df_s|_c$  value is lacking at the moment. The actual value, however, is not really important. More important is to recognize that process control by temperature in a nonsteady system of heat waves with varying amplitudes is troublesome. As already noticed in Section 3.2, the speed with which the waves travel through the system is relatively low. As a result, it takes a long time to react to changes in the fraction solid and viscosity. Therefore, the temperature does not seem to be an adequate control parameter for attaining a constant (apparent) viscosity of the exiting slurry.



Figure 7 Illustration with criterion of stircastability. Indicated are the work-region of fraction solid  $0.35 < f_s < 0.55$ , and the minimum  $|dT/df_s|_c$  value, which was experimentally determined at 30 deg (wt%)<sup>-1</sup>. The minimum stircastable alloy content, here Pb-16% Sn, is determined by the  $dT/df_s$  curve, which is above the critical  $|dT/df_s|_c$  value. The maximum stircastable alloy content, here Pb-36%% Sn, is determined by the  $T(f_s)$  curve, which must cover most of the work-region of the fraction solid.

#### 6. Summary and conclusions

It has been shown that apart from the general process limits for the continuous casting of thixotropic semi-solid metal slurries, a particular method imposes specific process limits which are determined by the actual apparatus and the available facilities.

The specific process limits define the composition limits in (binary) alloy systems with which alloys can be continuously stircast. For the particular method of mouldless semicontinuous stircasting introduced here, this implies that eutectic and close to eutectic alloys cannot be used and also that stircastability decreases toward smaller freezing ranges.

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**Appendix:** Derivation of Equation 4 Scheil's equation is written as

$$f_{\rm L}(T) = \left(\frac{C_{\rm L}(T)}{C_0}\right)^{\frac{1}{k_0 - 1}}$$
 (A1)

If  $k_0$  is a function of temperature, the following formula may be used to differentiate  $f_s$  with respect to T:

$$\frac{\mathrm{d}}{\mathrm{d}x}u^{v} = u^{v}\ln u \frac{\mathrm{d}v}{\mathrm{d}x} + vu^{v-1}\frac{\mathrm{d}u}{\mathrm{d}x} \quad (A2)$$

in which both u and v are functions of x. Substitution of

$$u = C_{\rm L}(T)/C_0$$

$$v = 1/[k_0(T) - 1]$$
(A3)

gives

$$\frac{\mathrm{d}f_{\mathrm{L}}}{\mathrm{d}T} = \left(\frac{C_{\mathrm{L}}}{C_{0}}\right)^{\frac{1}{k_{0}-1}} \left(\ln\frac{C_{\mathrm{L}}}{C_{0}}\right) \left[-\frac{\mathrm{d}k_{0}/\mathrm{d}T}{(k_{0}-1)^{2}}\right] + \frac{\mathrm{d}C_{\mathrm{L}}/\mathrm{d}T}{C_{0}(k_{0}-1)} f_{\mathrm{L}}^{(2-k_{0})}$$
(A4)

For Al-Cu, Al-Mg, and Al-Si alloys, the first term in this equation was found negligible, with respect to the second.



Figure 8 Regions of optimum stircastability, for the binary systems Al-Cu, Al-Mg, Al-Si, and Pb-Sn, for a work-region of fraction solid  $0.35 < f_s < 0.55$ , and a critical  $|dT/df_s|_c$  value of 30 deg (wt %)<sup>-1</sup>.

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