

The Controlled Diffusion Solidification Process: *Fundamentals and Principles*

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Introduction

Wrought aluminum-based alloys exhibit superior physical and mechanical properties compared to conventional shape casting alloys. However, wrought alloys cannot be cast into near net shapes because they develop hot tears and hot cracks during solidification. For this reason, these alloys are typically cast into ingots and are subsequently brought to final shape by mechanical processes such as rolling, extrusion, drawing and forging. These processes significantly increase the cost of the manufactured component. **Controlled Diffusion Solidification** (CDS) is a novel process that allows casting of wrought alloys directly into final shapes that are free of hot tears; thus CDS eliminates the added cost of machining. The process follows a different route from conventional casting methods. In CDS two liquid alloys of predetermined composition and temperature are mixed together so that upon solidification the resultant alloy has a globular rather than a dendritic microstructure as shown in Figure 1. The hot tearing tendency of wrought alloys originates from the inadequate permeability of their dendritic network, which obstructs the flow of interdendritic liquid and hinders compensation for shrinkage. The nondendritic microstructure made possible by CDS minimizes hot-tearing thus enabling wrought alloys to be cast directly.

The CDS has the potential to:

- Reduce the cost of multiple component assemblies
- Minimize several typical casting defects that are associated with solidification
- Reduce the time required to fabricate assemblies
- Reduce process variability by controlling the solidification
- Increase casting applications

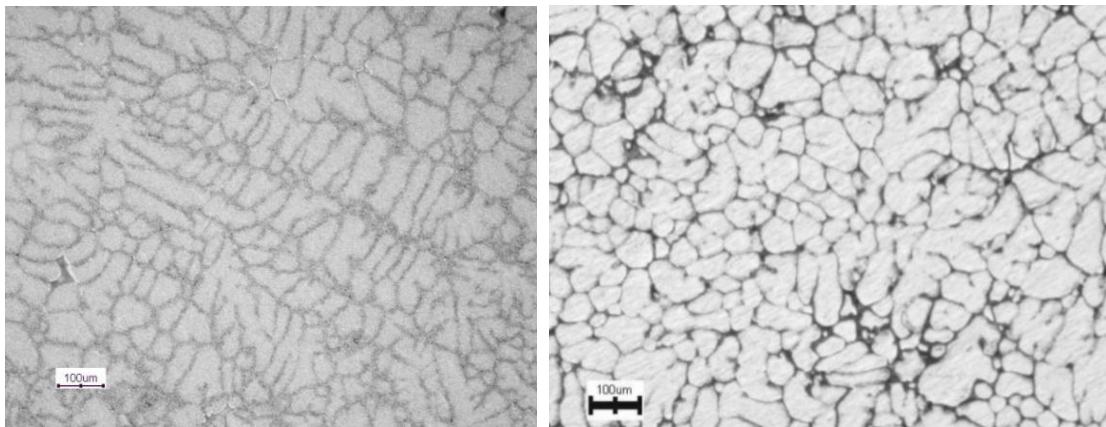


Fig. 1: a) 206 Conventional Casting- Dendritic

b) 206 via CDS - Globular Microstructure

Objectives

In order to take full advantage of the CDS process, it is of vital importance to understand its fundamental mechanisms and principles. Therefore the goal of this work was to develop the mechanisms underlying CDS, and to establish the range of flexibility of the process with respect to possible alloy compositions and processing parameters.

Specifically the objectives were to:

- Identify volumetric and temperature constraints for the two starting liquids.
- Formulate a theory of the nucleation and growth events that occur during CDS
- Construct a mathematical model that predicts the degree of undercooling for a binary alloy system as a function of mixing conditions
- Apply the globular stability model to predict instability of the interface at the latter stages of CDS
- Establish hot tearing tendency of alloys when cast via CDS.

Methodology

Since the CDS is a dynamic process where two phase non-isothermal flow and solidification occur simultaneously a hypothesis was initially formed. Specialized experiments were designed to confirm the hypothesis and novel techniques were developed to enable rapid quenching in order to investigate the intermediate mixing and solidification steps.

The CDS can be broken down into three phases. This made it easier to identify governing factors, and processing parameters. The three phases were:

1. Mixing
2. Nucleation
3. Growth

and can be visualized in Fig.2

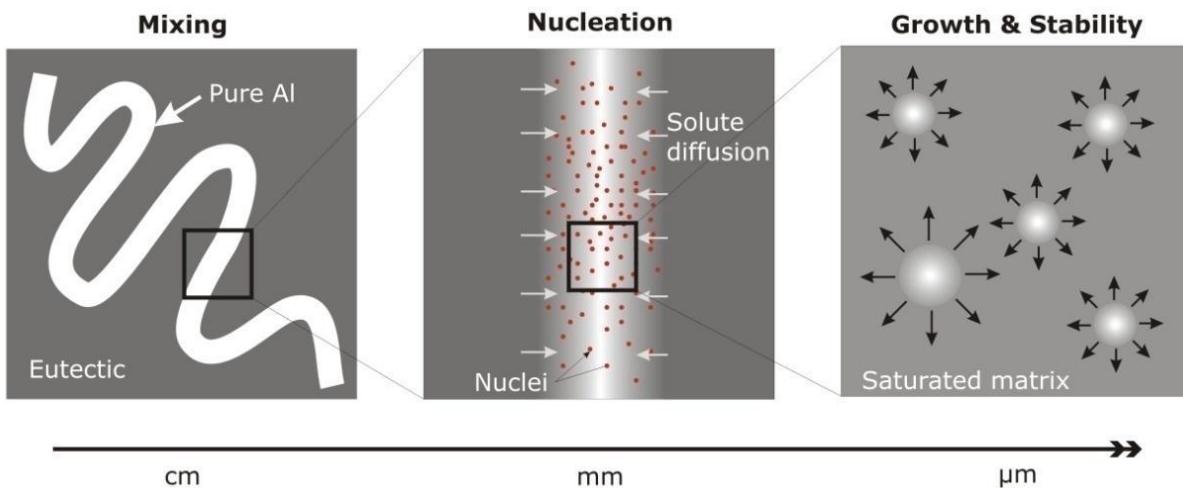


Fig. 2: (a) Pure Al is mixed with the eutectic liquid forming striations. (b) Undercooling in the Pure Al striations forces rapid nucleation while at the same time solute diffuses from the eutectic liquid in the Pure Al striations. (c) The Al nuclei grow in a solute rich matrix.

In summary, the first phase concerns fluid flow when the two liquids mix. The mixing procedure is critical because incomplete mixing can lead to segregation issues. The second phase studies the undercooling of the liquid Al, which results in rapid nucleation of solid Al particles. Rapid nucleation creates a high nuclei density of solid Al nuclei, which favors globular stability. The third phase examines the environment where the nuclei grow during CDS, which is different compared to that during conventional solidification. In CDS, the nucleated particles grow in a solute rich matrix, which slows down the growth and a planar solid-liquid front is favored. However, if the cooling rate is relatively high, the front can become unstable and break down into a cellular or dendritic one.

The detailed theoretical framework developed and the laborious experimental results quantify the effect of the process parameters and enable precise process control.

Results

The results of this research are presented below and are grouped by the three phases of the methodology.

A. Mixing of two liquids/melts

The first phase sheds light into manipulating the mixing parameters to guarantee a globular microstructure. After extensive experimental work, three mixing modes were identified upon mixing of two liquid melts. The mode can be selected by manipulating the temperature and the weight ratio of the two liquids. The mixing modes are presented in Fig.3. The Al-Cu binary system was selected for these experiments.

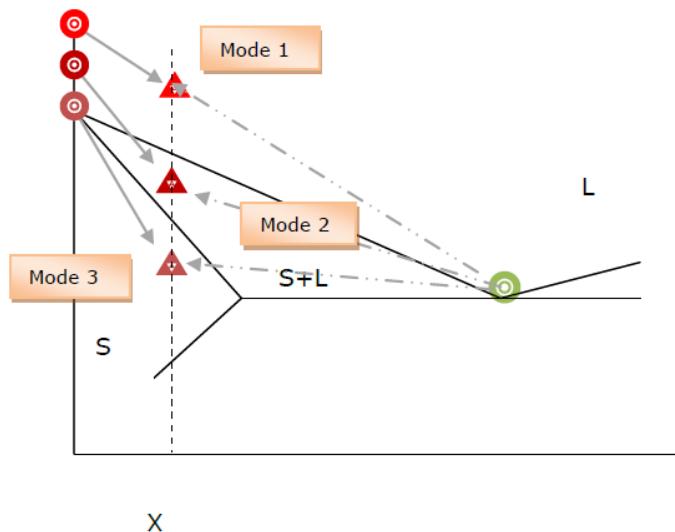


Fig. 3: Generic binary phase diagram depicting different mixing modes.

The mixing modes are presented below. The operative mixing mode in CDS is Reaction-Mixing and its boundary conditions dictate the range of castable alloys via CDS.

1. Ideal mixing

This mode of mixing occurs when the superheat of the pure substance is high enough that the resultant mixture finds itself in the liquid region of the phase diagram (Fig. 3). This situation is the same as alloying in the liquid state. The two liquids combine to form a homogeneous liquid of composition X that solidifies with the conventional solidification mechanism.

2. Reaction mixing

In this mode of mixing, partial solidification of the pure liquid occurs as it comes into contact with the lower temperature eutectic liquid. In the CDS process, the initial temperatures and compositions of the starting liquids are such that the resultant mixture is in the two-phase region of the phase diagram (Fig. 3).

3. Quenching

The third mode of mixing occurs when the superheat of the pure liquid is too low and/or the target composition is high in solute. This means that the weight ratio of the pure substance to the eutectic is low. If this is the case, then heat transfer will be the governing mechanism and the pure liquid becomes quenched before it is allowed to mix with the eutectic liquid. This extreme path will lead to considerable segregation (Fig. 3).

Typical microstructures of the three mixing modes are presented in Fig. 4. Observe the globular microstructure produced by CDS processing.

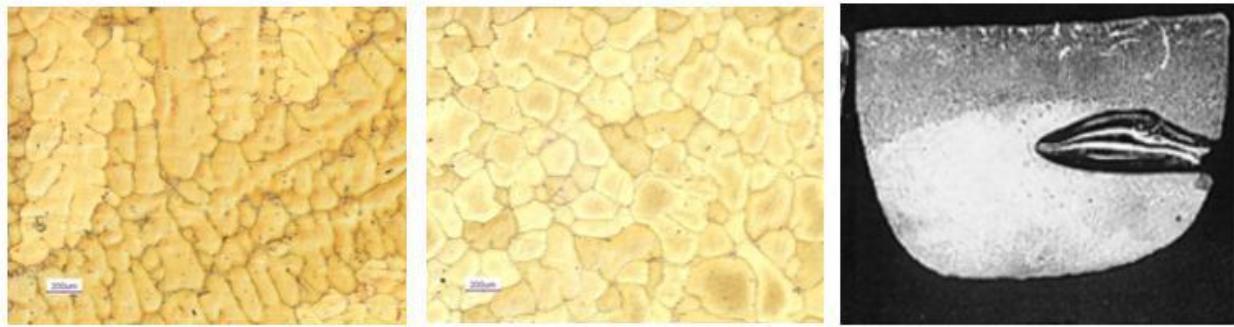


Fig.4: From left to right - a) Homogeneous liquid solidifying (dendritic microstructure) b) Reaction mixing, (globular) c) Quenching (severe segregation).

B. Nucleation

In order to understand the nucleation mechanism and quantify the undercooling during mixing a numerical model was formed to study the evolution of solute and temperature fields. The present model involved a single striation of Pure Al stretching in the infinite bulk of the second melt, which is an Al-33%Cu eutectic alloy.

Formulation that describes diffusion and reaction in stretching lamina in a Lagrangian frame of reference fixed on the lamina:

$$\frac{\partial C_a}{\partial t} + e_x x \frac{\partial C_a}{\partial x} = D_a \frac{\partial^2 C_a}{\partial x^2}$$

Results showed that the highest degree of undercooling occurs at the onset of the mixing process at the center of the Al striation. However there is also undercooling at the eutectic melt, at the position of the interface due to decreasing concentration, Fig.5. This suggests that an increase of the number of lamella in the mixture will increase the fraction of the undercooled regions.

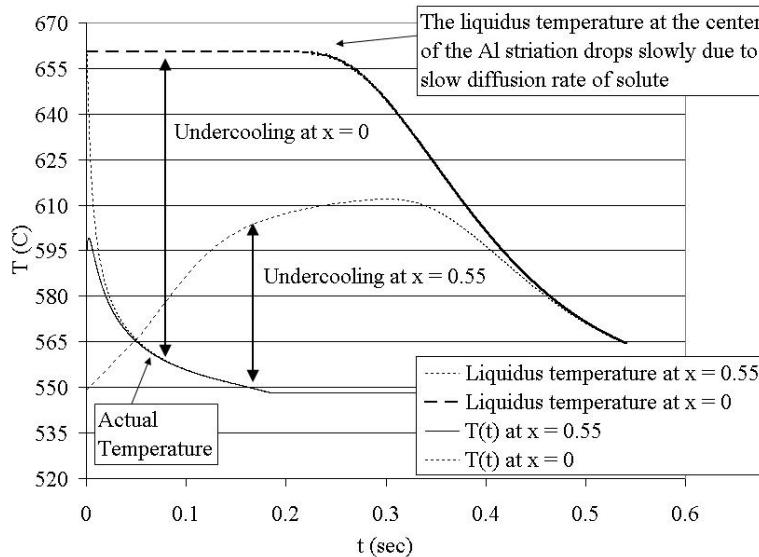


Fig. 5: Evolution of temperature and solute fields and degree of undercooling.

C. Growth-Stability

Globular stability theory was applied in determining the optimum process parameters for globular microstructure. A modified stability criterion was developed that can predict the microstructure in relationship to cooling rate and grain density, Fig. 6.

$$f_s = \left[1 - C \left(\frac{4\pi N}{3} \right)^{1/3} \left(\frac{(D_L)^3 \Gamma [m_L C_0 (k-1)]^2}{(\dot{T}_{max})^3} \right)^{1/7} \right]^3$$

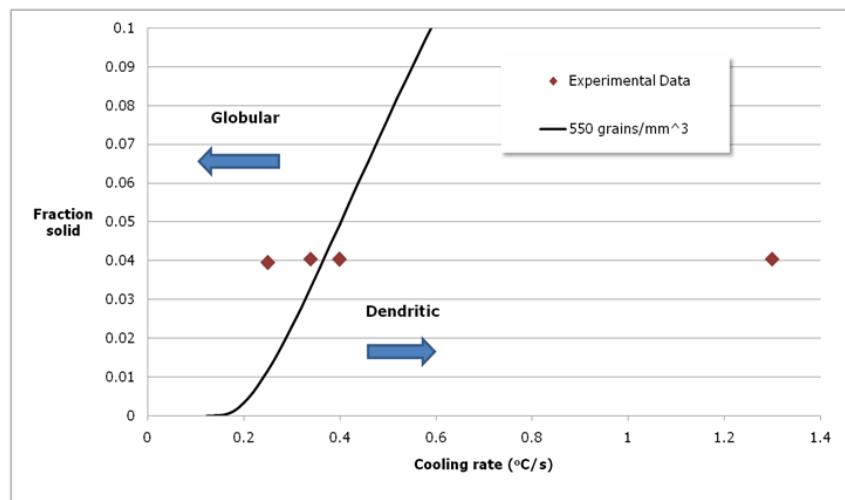


Fig. 6: The graph shows the stability region for an Al-4.5wt%Cu alloy cast via CDS under different cooling rates. The left of the curve denotes stability and the right indicates instability of the front. From the above graph

the combinations of fraction solid and maximum cooling rate can be extracted that ensure stable globular growth.

D. Reduction of hot-tearing tendency

In order to demonstrate the hot-tearing reduction of castings produced via CDS a ring mold was used. Among the alloys investigated were 206 and 2014. The results can be clearly seen in Fig. 7. Wrought alloy 2014 can be readily cast via CDS where it would be extremely difficult to cast via conventional methods.



Fig. 7. (a) 2014 Cast via CDS - Free of Hot tears. (b) 2014 conventionally cast - Moderate hot tear.

Outcome / Impact

The CDS process is a novel means of casting an alloy of a given composition by mixing two liquid entities of different compositions (and thus different solute amounts) as well as two different volumes of the two liquids. The process allows one to alleviate the casting issues that are encountered when casting alloys that have hot tearing tendencies – i.e., wrought Al alloys.

The following conclusions can be made from this work:

1. The governing mechanism of the Controlled Diffusion Solidification casting process was investigated and verified with experimental results. The nucleation mechanism was identified to occur in the undercooled Al-striations. The parameters for interface stability were also identified and investigated experimentally and theoretically.
2. The underlying mechanism is explained on the basis of mixing considerations, initial conditions and evolution of thermal and solute fields. Specifically, in order for the solid phase to nucleate and grow in a globular morphology, the two precursor alloys need to be mixed in suitable conditions that will follow the reaction-mixing mode. The reaction-mixing mode requires that the mixture's temperature is in the semi-solid region of the phase diagram. This forces rapid nucleation due to the high degree of undercooling in the Al-striations.
3. The underlying mechanism and the knowledge of the CDS process can be extended to other alloy systems. From a commercial perspective, foundries can take advantage of the hot-tearing free castings produced via CDS with shorter solidification times and cast wrought alloys without the hot-tearing issues.

4. CDS can be readily applied in sand and investment castings. The application in higher cooling rate casting processes may require the addition of grain refiners.
5. The operating window of the process was established to be 10°C, and the maximum cooling rate 0.4°C/sec. Higher cooling rates may be achieved by allowing a higher fraction solid before casting as shown in Figure 2. In fraction solids above 20% external force should be applied to fill the cavity.
6. Successful casting of alloys via the CDS was demonstrated as shown in Figure 3. Al alloys 206 and 2014 exhibited globular microstructure and the hot-tearing reduction was demonstrated in both alloys.

Publications

- K. Symeonidis, D. Apelian, M. M. Makhlof, "Controlled Diffusion Solidification: Application to Metal Casting", La Metallurgia Italiana, March 2009 (in press)
- K. Symeonidis, D. Apelian, M.M. Makhlof, "Study of Undercooling Mechanism in the Al-Cu Alloy System Cast via Controlled Diffusion Solidification (CDS) Process", NADCA Transactions; published by NADCA, Cast Expo, May 2008.
- K. Symeonidis, D. Apelian, M. M. Makhlof, "Controlled Diffusion Solidification: Application to Metal Casting", Proceedings of the 4th International High Tech Die Casting Conference (HTDC 2008 Brescia, Italy, April 2008) – Paper #39 (Invited Paper)
- K. Symeonidis, D. Apelian, M. M. Makhlof, "Controlled Diffusion Solidification", International Foundry Research/Giessereiforschung 60 (2008) No. 1 pp. 2 - 8
- K. Symeonidis, D. Apelian, M. M. Makhlof, "Controlled Diffusion Solidification: Application to Metal Casting", Metallurgical Science & Technology, Vol. 26-1, pp 30-36, 2008.
- K. Symeonidis, D. Apelian and M.M. Makhlof, " Controlled Diffusion Solidification", in the Proceedings of LMT – Light Metals Technology 2007, September 2007, Saint-Sauveur, Québec, CA, published by CANMET.