Temperature, density, and velocity distribution in the mixture prepared by controlled diffusion solidification process

Abbas A. Khalaf¹

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Abstract
Controlled diffusion solidification (CDS) is a casting process that depends on mixing two precursor alloys to make a required resultant alloy. The CDS process can form a non-dendritic microstructure happening from copious nucleation, resulting in minimizing the tendency toward hot tearing. Pure aluminum was mixed into Al-33wt%Cu through funnels that have 9 and 6 mm diameters to make Al-4.7wt%Cu. Ansys software was employed to simulate the temperature, density, and velocity redistribution in the mixture during the mixing step. The results show that the thermal curves give better indications to predict the periods of nucleation and the heat transferring to the environment. Furthermore, the copious nucleation that occurs in the CDS process can be improved by controlling the agitation during the mixing step by decreasing the velocity of the mixture near the crucible wall and increasing the velocity in the middle of the mixture. The velocity around 1.7 ms⁻¹ and 0.2 ms⁻¹ happening at the middle and near crucible wall, respectively, gives better non-dendritic microstructure.

Keywords Nucleation · Controlled diffusion solidification · Microstructure · Al alloys

Abbreviations
Alloy1 Precursor alloys
Alloy2 Precursor alloys
Alloy3 Resultant desired alloy
C₈ Copper concentration wt%
C₀ Alloy3 concentration wt%
Cₚ Specific heat of liquid (J kg⁻¹ K⁻¹)
K Liquid thermal conductivity (W m⁻¹ C⁻¹)
m₁ Mass of Alloy1
m₂ Mass of Alloy2
mr Mass ratio (m₁/m₂)
p Pressure (Mpa)
T Actual temperature (K)
T₁ Alloy1 temperature (K)
T₂ Alloy2 temperature (K)
Tₘ Minimum temperature (K)
Tₘ Melting temperature of pure metal (K)
T_Liq Liquidus temperature (K)
ΔΤ Undercooling below liquidus temperatures (K)
t Time (s)

Introduction
Aluminum-copper alloys play an important role in lightweight casting to improve fuel economy in the automotive industry [1, 2]. Aluminum wrought alloys could not be easily cast by conventional casting processes because they are prone to make hot tearing during solidification, where the inter-dendritic liquid at the end of the solidification process cannot effectively feed the shrinkage cavities created during solidification of the primary, secondary, and ternary branches of the dendrites because of creating a large and complex dendritic network [3, 4]. The CDS process is a casting process depending on mixing two precursor alloys that have different masses and temperatures to make copious nucleation that results in improving the mechanical properties by changing the microstructure from dendritic to small size non-dendrites that leads to minimizing the hot tearing tendencies [5–11].
The mixing step, nucleation, and growth are the steps of the mechanism of the CDS process presented by Apelian [12], Symeonidis [13], and Khalaf [6]. The results achieved from the successful CDS process are small-size globular morphologies forming from copious nucleation occurring during the CDS steps without needing to add a grain refiner [6, 8, 11, 14, 15].

Yang et al. [16] studied the microstructure of the 7xxx series aluminum alloys cast via the CDS process and conventional casting. Yang found that the intermetallic phases formed in the microstructure for the CDS process are significantly different from that forming in the conventional casting. Ghiaasiaan and Shankar [17] cast 7xxx series aluminum alloys via the CDS process; the mixture was directly poured into a metal mold designed for a tilt pour casting machine to make tensile test bars that were used to study the mechanical properties and the intermetallic phases forming in the eutectic phase. The results were compared with the conventional casting process. Ghiaasiaan expected that the solute redistribution during the solidification for the CDS process is significantly different from that in the conventional casting, leading to making the phase fraction of the eutectic phase greater in the CDS process.

Khalaf [18, 19] used Al-Cu and Al-Zn binary alloys system to study the microstructure formed by the CDS process experimentally and by simulation; the results exhibited that globular, rosette, and dendrites coexist in the entire microstructure, even though all the morphologies were exposed to the same thermal environment during growth; furthermore, anomalous morphologies form in the microstructure when using pure aluminum as a first precursor alloy that has a higher liqudus temperature. Recently, Khalaf [20] employed the concept of the two steps nucleation theory along with the mixing of two alloys to predict the transition temperature at which the metastable zone changes to the labile zone for undercooled Al-Si hypereutectic alloys. The mixing step was used to make the undercooling required to reach the transition temperature. Khalaf found the solute content, temperature distribution, and crucible temperature significantly change the size of the morphologies forming in the mixture. Khalaf [21] studied the difficulties associated with forming the non-dendritic microstructure created in Al-Si hypoeutectic alloys made by the CDS process; the results show that the mass ratio of the mixed alloys has a significant effect on forming the non-dendritic microstructure when mixing pure aluminum with Al-Si eutectic alloy; furthermore, the microstructure also can be changed when mixing pure aluminum with Al-Si hypereutectic alloys.

In the CDS process, nucleation occurs during the mixing step. The nucleation can take place without adding grain refiner, spontaneously in the mixture [20] or heterogeneously at the crucible wall [18]. The agitation of the mixture during the mixing strongly affects the nucleation rate and forms a non-dendritic microstructure [11, 22]. Many events are happening during the mixing step in the CDS process. This paper was dedicated to studying the critical events happening during the mixing step. These events can be detected by the thermal curve and simulation of the mixing step. The thermal curves were drawn from temperature data collected during the mixing and solidification steps in the CDS process. Simulations of the mixing step were carried out to support the analysis of the thermal results by studying the redistribution of the temperature, density, and velocity in the mixture. Experimental and simulation evidence along with microstructure study support the discussion in this publication.

2 Experiments

The CDS process depends on mixing two precursor alloys (Alloy1 and Alloy2) to make the resultant alloy (Alloy3). In this publication, Alloy1 and Alloy2 were pure aluminum and eutectic alloy (Al-33wt%Cu), respectively. The two precursor alloys were mixed with a mass ratio of 6 (mr = 6) to make Alloy3 (resultant alloy (Al-4.7wt%Cu)). Table 1 shows the designation of the experiments along with the independent parameters and the constants for all the experiments carried out in the present study. Figure 1a shows the location of the experiments involved in Table 1 on the Al-Cu phase diagram.

Three CDS experiments named Ex1, Ex2, and Ex3 were carried out by mixing Alloy1 into Alloy2 through a 9-mm diameter funnel. Alloy1 temperature was at 665, 670, and 675 °C, respectively. The temperature of Alloy2 was fixed at around 5 °C higher than the respective liquidus temperature. The mixture (Alloy3) solidifies in the same crucible of Alloy2. The funnel and crucible were made from baked clay for all experiments. Figure 1b shows a schematic diagram of the mixing step that was carried out through a funnel. In Fig. 1b, the locations of the funnel, crucible, Alloy1, and Alloy2 appear in the diagram. Three thermocouples were placed in the funnel and the crucible to collect the temperature data for the CDS experiments. In Fig. 1b, the thermocouple name T1 was placed in the funnel to collect Alloy1 temperature, and TL (lower thermocouple) and Tu (upper thermocouple) represent two thermocouples placed in the crucible to collect the mixture temperature during the mixing step.

The experiment Ex4 was carried out by re-melting 330 g of the CDS samples at 662 °C and pouring the molten alloy (Al-33wt%Cu) through a 9-mm funnel diameter into a hot empty crucible re-heated at 555 °C, where the empty crucible temperature was chosen to be equal to the temperature of
the crucible and Alloy2 for the CDS experiments. Figure 1c shows a schematic diagram representing the pouring step into a hot empty crucible. The experiment Ex5 was carried out by re-melting 330 g of the CDS samples at 659 °C and solidified at the same crucible on air. Ex5 represents conventional solidification.

Three CDS experiments named Ex6, Ex7, and Ex8 were carried out by mixing Alloy1 into Alloy 2 through a 6-mm diameter funnel with mr 6 and Alloy1 temperature at 667, 670, and 676 °C respectively. The mixture solidified in the same crucible as that of Alloy2.

Specimens were cut from the edge and middle of the samples for the experiments Ex1 to Ex8. The specimens were prepared for an optical microscope test, wherein they were mounted using bakelite mount, ground, and polished. Keller’s reagent (1 ml HF, 1.5 ml HCL, 2.5 ml HNO3, and remainder H2O) was used for etching the specimens. A stereo microscope type NIKON1 AZ 100 MI was used to

![Fig. 1 a Al-Cu phase diagram showing the location of alloy1, alloy2, and alloy3, b schematic diagram showing the mixing through a funnel for the CDS process, c schematic diagram for conventional casting poured into an empty hot crucible, and d schematic diagram showing the dimensions of the crucible and the specimens pointed by square for the optical microstructure images](image)

Table 1 Experimental designations with independent parameters and constants for the laboratory experiments

| Designation | Alloy 1 | | Alloy 2 | | mr | Alloy3 |
|--------------|---------|----------|---------|----------|---|---------|
|              | m1 (g)  | T₁ (°C)  | T₁Liq (°C) | m2 (g)  | T₂ (°C)  | T₂Liq (°C) | Co (wt %) |
| Ex1 (9 mm)   | 290     | 665      | 660      | 47      | 555      | 549      | 6        | 4.7 |
| Ex2 (9 mm)   | 290     | 670      | 660      | 47      | 555      | 549      | 6        | 4.7 |
| Ex3 (9 mm)   | 290     | 675      | 660      | 47      | 555      | 549      | 6        | 4.7 |
| Ex4          | 330 g of Ex1 to Ex3 samples re-melted at 662 °C and poured through 9-mm diameter funnel into an empty hot crucible preheated to 555 °C |
| Ex5          | 330 g of Ex1 to Ex4 samples re-melted at 659 °C and solidified conventionally at the same crucible |
| Ex6 (6 mm)   | 290     | 667      | 660      | 47      | 558      | 549      | 6        | 4.7 |
| Ex7 (6 mm)   | 290     | 670      | 660      | 47      | 558      | 549      | 6        | 4.7 |
| Ex8 (6 mm)   | 290     | 675      | 660      | 47      | 554      | 549      | 6        | 4.7 |

1 NIKON AZ 100 M is a trademark of Eberbach Corporation, Ann Arbor, MI.
capture microstructure images. Figure 1d shows the dimensions of the crucible and the location of the edge and middle specimens that were pointed by squares. The funnel and crucible have the same dimensions.

3 Heat balance during the mixing step

The CDS process depends on mixing two precursor alloys that have different masses and temperatures. The heat and the species transport between the two precursor alloys, while the heat transfers to the environment during the steps of the CDS process. The heat balance in the system is as follows.

3.1 Enthalpy of alloy 1

This kind of heat adds to the mixture when the alloy with higher temperature is mixed into the alloy with lower temperature. In this publication, Alloy 1 with the higher mass and temperature in the system was mixed into Alloy 2 with the lower mass and temperature. The instantaneous temperature of the mixture continues increasing with adding Alloy 1 through the funnel. The Alloy 1 temperature and volume rate of mixing play a capital role in changing the mixture temperature.

3.2 Enthalpy of mixing

The enthalpy of mixing adds to the mixture when the species of Alloy 1 and Alloy 2 diffuse from a higher concentration to a lower concentration [20].

3.3 Enthalpy of fusion

The nucleation occurs from the undercooled alloy that has a higher respective liquidus temperature in the CDS process. The latent heat of fusion issues to the mixture when the nucleation happens. The nucleation depend on the undercooling of Alloy 1 during the solidification. Two kinds of nucleation (heterogeneous and spontaneous nucleation) can occur during the solidification. Two kinds of nucleation (heterogeneous and spontaneous nucleation) can occur during the solidification process. The homogeneous nucleation occurs at the crucible wall when the mixture is free of grain refiner [6, 8, 18, 19]. The spontaneous nucleation happens from the undercooled alloy when supersaturation solution forms in the mixture [12, 20, 23]. Both kinds of nucleation depend on the undercooling of Alloy 1 during the mixing step.

3.4 Heat transfer from the mixture

The heat transports between the mixture and both the crucible and surrounding air. The crucible material and crucible temperature significantly affect the mixture temperature.

4 Simulation of mixing step

The mixing of the two precursor alloys in the CDS process has been simulated by Ansys Fluent® software. 2D axisymmetric, the volume of fluid (VOF), three phases, and k-ε with shear stress transport turbulent model has been employed to simulate the mixing step. In the model, pure aluminum (Alloy 1), Al-33wt%Cu (Alloy 2), and air were the three phases coexisting in VOF. Equations (1) to (3) represent the heat equation, Navier stocks equation, and continuity equation. These equations were numerically solved in the simulation. In Eq. (1), $C_p = 1179$ [24] and 953.9 J kg$^{-1}$ K$^{-1}$ [19] are the specific heat for Alloy 1 and Alloy 2, respectively [18–20]. $K = 95$ w.m$^{-1}$.K$^{-1}$ [25]. In Eq. (2), $\mu = 1.38$ mPa [6, 18, 19] and 5.6 mPa [21] are the viscosity for Alloy 1 and Alloy 2, respectively. $\sigma = 2375$ kg.m$^{-3}$ is the density for Alloy 1 (pure aluminum) at melting temperature ($T_m = 660.4$ °C) [6]. Equation (4) represents the density change with copper concentration and temperature for Al-Cu hypoeutectic alloys [6]. In Eq. (4), $\beta_T = -4.032 + (1.654E - 2)T - (2.53E - 5)T^2 + (1.23E - 8)T^3$ and $\beta_C = 22.19678 + (0.1629)C_{Cu} + (9.0973E - 4)C_{Cu}^2 + (2.004E - 5)C_{Cu}^3$ are variables depending on the temperature and copper concentration, respectively. The surface tension ($\sigma$) was employed in the simulation. Equation (5) [2] evaluates the surface tension ($\sigma$) calculated as a function of temperature starting from the liquidus temperature. In Eq. (5), $\sigma = 0.868$ Nm$^{-1}$ is for pure aluminum at melting temperature [12]. $\sigma_L = 0.951$ Nm$^{-1}$ and $(d\sigma/dT) = (-0.92 \times 10^{-4})$ Nm$^{-1}$ K$^{-1}$ were taken for Alloy 2 [2]. The Ansys default data was used to evaluate the required data for the air, crucible, and the funnel. The mesh size was 500 µm for air and 1 mm for the crucible and funnel. The heat transferring by radiation and convection to the environment has been employed in the simulation, where the bulk temperature and heat transfer coefficient for convection were 25 °C and 24 W.K$^{-1}$.m$^{-2}$, respectively. Figure 2a shows the initial condition of Alloy 1 and Alloy 2 placed into the funnel and the crucible along with the temperature of the environment prior to the mixing step:

$$\rho C_p \frac{dT}{dt} + \rho C_p u.\nabla T = \nabla.(K \nabla T)$$

$$\frac{d\mu}{dt} + \mu . \nabla u = \nabla. [-p + \mu(\nabla u)]$$

$$\rho \frac{dp}{dt} + p \nabla . u = 0$$
Results and discussion

Figure 2b–g show the optical microstructure images of the CDS experiments denoted as Ex1, Ex2, and Ex3 involved in Table 1. The images were taken at the middle and the edge of the specimens for the CDS process samples shown in Fig. 1d. Figure 2b and c show the optical microstructure images of the sample for Ex1 taken at the middle and edge, respectively. In Fig. 2b and c, a non-dendritic microstructure forms the entire microstructure, where globular morphologies with a few rosettes dominate the microstructure, and the arrows refer to some of the globular morphologies taken as an example. The average grain size was 230 µm. Figure 2d and e show the optical microstructure images of the samples for Ex2 taken at the middle and the edge, respectively. In Fig. 2d and e, globular rosettes with a few dendrites form the non-dendritic microstructure. Some of the rosette morphologies were pointed by arrows. The average grain size was 600 µm. Figure 2f and g show the optical microstructure images of the samples for Ex3 taken at the middle and the edge, respectively. In Fig. 2f and g, dendrites with a few rosettes form the microstructure, where some of the dendrites were pointed by arrows. The average grain size was 930 µm. The smaller average grain size creates in the Ex1 and Ex2 microstructure, establishing from copious nucleation taking place in the mixture [6, 8, 10, 11, 19, 20, 23]. In addition, the non-dendritic microstructure of the experiments Ex1 and Ex2 reveals that the parameters of Ex1 and Ex2 demonstrated in Table 1 are correctly chosen to make successful CDS experiments.

Figure 3a shows the thermal curves drawn from the temperature data recorded by the lower and upper (T_L and T_U) thermocouples for the experiment named Ex1. The thermal curves cover the period starting from the bigging of the mechanical mixing and continuing to the end of the solidification (eutectic solidification). In Fig. 3a, points A, M, and S represent the events appearing on the thermal curve that was drawn from the temperature data collected from the lower thermocouple. Where point A represents the starting of the mechanical mixing. Point M represents the maximum temperature reached in the mixture. The maximum temperature exists in the bottom crucible. Point S represents the starting of the final solidification (eutectic solidification) taking...
place in the mixture. Further in Fig. 3a, point K represents the temperature of the air in the crucible prior to the mixing step. The temperature of point K was collected from the temperature data recorded from the upper thermocouple (TU) that was inserted in the middle of the crucible over Alloy2. Figure 3b shows a magnified image of the area pointed by a square in Fig. 3a. In Fig. 3b, point Q represents the equalization of all temperature fields in the mixture. A suggestion is that the temperature equalizes in local areas after that the heat transfers from the bottom toward the top of the entire mixture [6]. Figure 3c–e show the transient thermal curve representing the temperature data collected from the lower thermocouple of the CDS experiment named Ex1, Ex2, and Ex3 for the interval starting from the mechanical mixing (point A) to the equalization temperature (point Q), respectively. In Fig. 3c–e, the points A, B, C, D, E, F, N, M, and Q represent events occurring during the mixing step happening at different times. The events take the same trend in the experiments Ex1 to Ex3 although the mixing happens at different temperatures of Alloy1.

Figure 3c shows the thermal curve of the CDS experiment named Ex1 for the interval starting from the mechanical mixing (point A) to the equalization temperature (point Q). In Fig. 3c, A, B, C, D, E, F, N, M, and Q are the events happening during the mixing step. The events were presented as follows:

- **Point A** represents the starting of the mechanical mixing. \( t_{\text{m}} = 0 \) is the time of the starting mechanical mixing. Figure 4a shows the simulation results carried by Ansys software of the density distribution at point A. The temperature and solute initial conditions of Alloy1, Alloy2, funnel, and crucible were taken for the experiment Ex1 involved in Table 1. In Fig. 4a, the enthalpy of Alloy1 starts to access the mixture.

- **Interval A-B** represents a sharp jump in temperature caused by the effect of adding the enthalpy of Alloy1. Figure 4b shows the simulation results for density distribution at point B, showing that only Alloy2 is in touch with the lower temperature crucible. This leads to inhibiting the heterogeneous nucleation from Alloy1 at the crucible wall. According to that, nucleation cannot occur at point B.

- **For interval B-C-D**, this interval appears on the thermal curves of the CDS experiments when Alloy1 is mixed into Alloy2 at superheat less than 10 °C. One
Fig. 4 Simulation results of density and temperature distribution for the points shown in Fig. 3c: a density distribution at point A, b density distribution at point B, c temperature distribution at points D and E, d density distribution at points E, e temperature distribution at points D, E, and F, f density distribution at point F, g temperature distribution at points F and M, and h density distribution at points M.
can suggest that the heat transferred between Alloy1 and Alloy2 leads to a drop in the temperature.

- For interval D-E, this interval represents a small deviation in the temperature gradient starting at point D. Figure 4c shows the temperature distribution at the points D and E taken at the line X–Y shown in Fig. 2a. The line was arbitrarily placed at 24 mm from the bottom of the crucible at the Alloy2 region. In Fig. 4c, the temperature of the crucible wall starts to significantly increase after point D as pointed by a square on the temperature distribution curves. Also in Fig. 4c, the temperature of the mixture (Alloy1 and Alloy2) is less than the respective liquidus temperature of Alloy1 ($T_{Liq}$), especially at the crucible wall. Figure 4d shows the simulation results for density distribution at point E, showing that Alloy1 is in contact with the lower temperature crucible. This leads to an increase in the probability of nucleating the primary aluminum in Alloy1 (pure aluminum) at the crucible wall. According to that, the copious nucleation can start after point E.

- Interval E–F represents a large deviation in the temperature gradient starting at point E. Point E represents the starting of the thermal recalescence in the thermal curve. The thermal recalescence happens from the latent heat issued from the nucleation that occurs during the mixing step [18, 19, 23]. Figure 4e shows the temperature distribution at the points D, E, and F taken at the line X–Y shown in Fig. 2a. In Fig. 4e, the hatched area gives an indication of the heat transfer from the mixture to the crucible, covering the period from the starting of the mechanical mixing till the time of point F. The hatched area is significantly bigger than that for the interval D-E that was pointed by the square. The suggestion is that the bigger amount of the heat transferring to the crucible makes the large deviation in the thermal curve although the heat continues adding to the mixture from the nucleation and the enthalpy of Alloy1. Figure 4e also shows that the temperature of the mixture at point F is less than the respective liquidus temperature of Alloy1 and it is higher than the temperature at point E. One can suggest that the probability of nucleation for the primary aluminum exists occurring with lower nucleation rate as the thermal curve moves toward the point F. Figure 4f shows the simulation results for density distribution at point F, showing that Alloy1 is in contact with the lower temperature crucible. This leads to continuing nucleating of the primary aluminum in Alloy1 (pure aluminum) at the crucible wall. Furthermore, the butter distribution of Alloy2 in the mixture happens at a time around and after point F.

- Interval F-M represents another deviation in the temperature gradient on the thermal curve of the experiment Ex1. Figure 4g shows the temperature distribution at the time of points F and M taken at the line X–Y shown in Fig. 2a. In Fig. 4g, the rate of the heat transfer to the crucible decreases after the time at point F. Furthermore, the temperature of the mixture almost equalizes at the bottom of the crucible, where the difference in the mixture temperature near the crucible wall and the middle of the mixture on the line X–Y is around 649.64–648.5 = 1.14 °C; furthermore, the actual temperature of point M shown in Fig. 4a is 650.3 °C collected by the lower thermocouple. The thermocouple reading was very close to simulation results (648.5 °C), where the difference exists because the latent heat of fusion issued from the nucleation of the primary aluminum was not employed in the simulation. Accordingly, the simulation results could be accepted. Point N appeared on the interval F-M represents the end of the mechanical mixing on which all the enthalpy of Alloy1 adds to the mixture. The temperature still increases after point N, and the temperature recalescence exists on the thermal curve continuing to the point M, suggesting that the nucleation continues taking place in the mixture after the ending of mechanical mixing, where the temperature of the mixture was less than the respective liquidus temperature of Alloy1. Figure 4g also shows that most of the crucible was at the initial temperature (554 °C). This leads to having a significant effect of the crucible temperature on the mixture temperature during the solidification of primary aluminum. Figure 4h shows the simulation results of the density distribution at point M, showing that Alloy1 with higher temperature dominates in the bottom of the mixture, leading to a gradient in the temperature happening between the bottom and the middle. The gradient in temperature obviously appears on the actual thermal curves shown in Fig. 3b, represented by the difference in the temperature between the points M and P that happen on the bottom and middle of the mixture, respectively. After point M, the heat transfers from the bottom toward the top of the mixture resulting in the equalization in temperature at point Q.

Figure 5a shows the thermal curves taken from the temperature data collected by the lower thermocouple for the experiments Ex1, Ex4, and Ex5 involved in Table 1. The thermal curves were superimposed at point A for comparison, where point A represents the starting of the mechanical mixing of Ex1. In Fig. 5a, the temperature difference between Ex4 and Ex1 represents approximately the effect of Alloy2 temperature on the mixture temperature, while the temperature difference between Ex5 and Ex4 represents approximately the effect of the crucible temperature on the mixture temperature. Furthermore in Fig. 5a, the effect of the crucible and Alloy2 temperatures on the mixture temperature increases with the elapsed time after settling down the mixture in the crucible. Figure 5b and c show
the optical microstructure images taken from the middle of the samples for the experiments named Ex4 and Ex5 involved in Table 1, respectively. Figure 5b shows the microstructure of the Ex4 that was carried out by pouring the resultant alloy (Alloy3 – Al-4.7 wt%) through a 9-mm diameter funnel into an empty crucible that was pre-heated at 555 °C. The Alloy3 was poured at 662 °C which is closer to the Alloy1 temperature of Ex1. Figure 5b reveals that the dendritic microstructure forms the entire solidified sample. The average diameter of the grain size was around 1600 µm. The average diameter is significantly bigger than that for the CDS experiment named Ex1 shown in Fig. 2b and c. Figure 5c shows the microstructure of the Ex5 that was carried out by melting the resultant alloy
(Alloy3 – Al-4.7 wt%) at 659 °C and solidified conventionally on air. In Fig. 5c, the typical dendritic microstructure of the conventional casting forms the entire solidified sample. The average size of the microstructure was around 3 mm [18]. Accordingly, in the CDS process, the mixing step, Alloy2 temperature, and crucible temperature play an important role in forming the non-dendritic microstructure with a small grain size without adding a grain refiner.

Figure 6a–f show the optical microstructure images of the CDS experiments denoted as Ex6, Ex7, and Ex8 involved in Table 1. The three CDS experiments were carried out by mixing Alloy1 into Alloy2 through a 6-mm diameter funnel. The images were taken from the middle and the edge specimens shown in Fig. 1d. Figure 6a and b show the optical microstructure images of the samples for Ex6 taken at the middle and the edge respectively. In Fig. 6a and b, globular morphologies with a few rosettes dominate the non-dendritic microstructure. The average grain size was around 200 µm. Figure 6c and d show the optical microstructure images of the samples for Ex7 taken from the middle and the edge specimens, respectively. In Fig. 6c and d, globular with more rosettes compared with the images of Ex6 forms the non-dendritic microstructure. The average grain size was around 250 µm. Figure 6e and f show the optical microstructure images of the samples for Ex8 taken from the middle and the edge specimens respectively. In Fig. 6e and f, globular rosettes with few dendrites form the non-dendritic microstructure. The average grain size was 820 µm[8]. The average grain size of the experiments Ex6 to Ex8 mixed through a 6-mm diameter funnel is significantly less than that for the experiments Ex1 to Ex3 mixed through a 9-mm diameter funnel, although the temperature of Alloy1, Alloy2, and the crucible was almost the same. In addition, the non-dendritic microstructure of the experiments Ex6 to Ex8 reveals that the parameters of Ex6 to Ex8 demonstrated in Table 1 are correctly chosen to make successful CDS experiments.

Figure 7a–c show the thermal curves taken from temperature data recorded by the lower thermocouple for the experiments named Ex6 to Ex8 designated in Table 1, respectively. In Fig. 7a–c, the events A, B, E, F, M, N, and Q obviously appear on the thermal curves, and take the same trend shown in Fig. 3c–e belong Ex1 to Ex3 that was mixed by 9-mm diameter funnel. However, the points that appeared in Fig. 7a–c take more time compared with that for Ex1 to Ex3 because the volume rate of entering Alloy1 into Alloy2 for the 6-mm diameter funnel is less than that for the 9-mm diameter funnel.

Figure 8a–f show the simulation results of the mixing step taking at the time representing the events taking place at the points A-B-E–F-M–N shown in Fig. 7a for the experiment named Ex6 carried out by the 6-mm diameter funnel at the almost same parameters for the experiment Ex1 that was carried out by 9-mm diameter. Figure 8a shows the simulation result of density distribution at starting of the mechanical mixing representing point A. Figure 8b shows the density distribution at point B, revealing that there is contact only between the Alloy1 stream and Alloy2. This leads that the recalescence B-C shown in Fig. 7a represents a heat transferring between Alloy1 and Alloy2 similar to that shown in Fig. 3c for the experiment Ex1. Figure 8c and d show the density distribution at points E and F, respectively, showing that the results take the same trend as the simulation results for the points E and F already discussed for the experiment Ex1 shown in Fig. 3c. Figure 8e shows the density distribution at maximum temperature (point M) existed in the thermal curve of Ex6. In Fig. 8e, Alloy1 is still adding at point M. However, for the experiment Ex1, the mixing of Alloy1 into Alloy2 is complete...
at point N that exists before point M and far from equalization temperature (point Q) as shown in Fig. 3c. Figure 8f shows the ending of the mechanical mixing for Ex6, happening at point N that exists after point M and near the equalization temperature (point Q) shown in Fig. 7a. The variance in the events happening at the points M and N for the experiments Ex6 and Ex1 happens because the lower volume rate associated with the 6-mm diameter funnel employed for Ex6 increases the filling time, leading to postponing the events resulting in improving the heat transferring from the mixture to the environment (crucible and air).

Figure 9a and b show the simulation results of the velocity distribution in the mixture taken on the line X–Y shown in Fig. 2a for the CDS experiments Ex1 and Ex6, respectively. Figure 9a shows the velocity distribution of Ex1 drawn for the simulation data that was taken at the time of points E, F, and N shown in Fig. 3c. In Fig. 9a, the velocity in the middle of the mixture is significantly higher than that near the crucible wall. Furthermore, the lower velocity exists between the crucible wall and the middle of the mixture. Furthermore, the mixing step starts with a higher velocity, and the velocity decreases as the time of mixing increases.

Fig. 9 Simulation results of the velocity distribution in the mixture at line X–Y shown in Fig. 2a for Ex1 and Ex6, a velocity distribution for Ex1, and b velocity distribution for Ex6

![Simulation results of the velocity distribution in the mixture at line X–Y shown in Fig. 2a for Ex1 and Ex6, a velocity distribution for Ex1, and b velocity distribution for Ex6](image)

Fig. 10 Comparison between the simulation results for the temperature distribution and velocity distribution at the points shown in the thermal curves for Ex1 and Ex6, a temperature at the time of point E, b temperature at the time of point F, c temperature at the time of point M, d velocity at the time of point E, e velocity at the time point F, and f velocity at the time of point N

![Comparison between the simulation results for the temperature distribution and velocity distribution at the points shown in the thermal curves for Ex1 and Ex6, a temperature at the time of point E, b temperature at the time of point F, c temperature at the time of point M, d velocity at the time of point E, e velocity at the time point F, and f velocity at the time of point N](image)
Figure 9b shows the velocity distribution of Ex6 drawn for the simulation data that was taken at the time of points E, F, M, and N shown in Fig. 7a. In Fig. 9b, the velocity distribution at the mixture takes almost the same trends happening at simulation results of Ex1 shown in Fig. 9a.

Figure 10a–c show a comparison between the simulation results of the temperature distribution for the experiments Ex1 and Ex6 mixed through 9-mm and 6-mm diameter funnels at the time of points E, F, and M shown in Figs. 3c and 7a, respectively. The temperature distribution data were taken at line X–Y shown in Fig. 2a. In Fig. 10a, the temperature distribution of the point E for Ex6 (6-mm diameter funnel) is significantly less than that for Ex1 (9-mm diameter funnel). This leads to an increase in the undercooling of Alloy1 resulting in improving the nucleation of primary aluminum for Ex6 from Alloy1 that has a temperature less than the respective liquidus temperature ($T_{\text{Liq}}$). Figure 10b and c show a comparison between the temperature distribution of Ex1 and Ex6 at points F and M, respectively. In Fig. 10b and c, the temperature distribution of Ex6 and Ex1 is almost the same at the points F and M. One can suggest that most of the copious nucleation taking place in the CDS process can occur at the interval E–F for the parameters under study.

Figure 10d–f show a comparison between the velocity distribution in the mixture between the experiments Ex1 and Ex6 that were mixed through 9-mm and 6-mm diameter funnels at the time of points E, F, and N shown in Figs. 3c and 7a, respectively. The velocity data were taken at the line X–Y shown in Fig. 2a. In Fig. 10d and e, the 6-mm diameter funnel makes more agitation in the middle of the mixture between the time of the interval E–F; however, the agitation decreases near the crucible wall compared with the 9-mm diameter funnel. According to these results, the nucleation improves at the crucible wall when decreases the agitation on the wall, and the higher agitation in the middle of the mixture gives better distribution of the primary aluminum nuclei that nucleate on the crucible wall. This gives acceptable interpolation to the smaller grain size forming with the 6-mm diameter funnel employed at the experiment Ex6. Figure 10f shows the velocity distribution at the end of the mixing step (point N). In Fig. 10f, the mixture velocity decreases to have less value with the 6-mm diameter funnel at the end of the mechanical mixing, where the lower volume rate of mixing Alloy1 into Alloy2 for the 6-mm diameter funnel significantly affects the agitation at the end of mixing step.

(Alloy3) by the CDS technique, where Alloy1 was mixed into Alloy2 through different size funnels having 9-mm and 6-mm diameter. Ansys software was employed to evaluate the density, temperature, and velocity redistribution in the mixture during the mixing step to support the experiments. The thermal curves, density distribution, temperature distribution, and velocity distribution have been employed to present a better environment to improve the copious nucleation taking place in the CDS process to form the non-dendritic microstructure, contrary to conventional castings in which dendritic microstructure has a large grain size forming the entire sample. The results are summarized below:

- The thermal curves can predict the starting of the copious nucleation events that appear as a thermal recalescence detected by the thermocouple. The thermal recalescence happens from the latent heat issued to the mixture.
- The heat transfer from the mixture to the baked clay crucible starts with a lower rate after the starting of the mechanical mixing and then drastically increases after 1 to 1.2 s depending on the amount of the mixture forming during the mixing for the experiments under study. This can predict by the change in the temperature gradient on the thermal curve.
- The velocity distribution in the mixture significantly affects the nucleation and the distribution of the nuclei in the CDS process. The maximum velocity appears in the middle of the mixture and the velocity decreases at the crucible wall while the minimum velocity exists in the rest of the mixture.
- The low value of the velocity at the crucible wall improves the nucleation rate from the highest liquidus temperature alloy when the alloy exists on the crucible wall; however, the higher velocity value in the middle of the mixture can enhance the distribution of the nuclei in the entire mixture.

The simulation results along with the experimental results elucidate the understanding of the events that appear in the thermal curves for the CDS process.

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Author contribution AAK designed the study, performed the research, analyzed the data, wrote the paper, and conducted experiments and data processing.

Availability of data and materials The experimental and simulation data are transparent.

Declarations

Ethics approval This article does not contain any studies with human participants performed by any of the authors.

6 Conclusions

The events happening during the mixing step of the CDS process have been presented. Pure aluminum (Alloy1) and Al-33wt%Cu (Alloy2) have been used to make Al-4.7wt%Cu...
Consent to participate This work was conducted with no human test subjects.

Consent for publication This work has consent for publication.

Conflict of interest The author declares no competing interests.

References

1. Fan Z, Fang X, Ji S (2005) Microstructure and mechanical properties of rheo-diecast (RDC) aluminium alloys. Mater Sci Eng A 412:298–306
2. Schmitz J, Brillo J, Egry I, Schmid-Fetzer R (2009) Surface tension of liquid Al-Cu binary alloys. Int J Mater Res 100:1529–1535. https://doi.org/10.3139/146.110221
3. Li S, Apelian D, Sadayappan K (2011) Hot Tearing in Cast Aluminum Alloys. Mater Sci Forum 690:355
4. Rappaz M, Dreze J-M, Gremaud M (1999) A new hot-tearing criterion. Metall Mater Trans A 30:449–455
5. Saha D, Shankar S, Apelian D, Makhlouf MM (2004) Casting of aluminum-based wrought alloys using controlled diffusion solidification. Metall Mater Trans A 35:2174–2180. https://doi.org/10.1007/s11661-004-0167-8
6. Khalaf AA (2016) Mechanism of controlled diffusion solidification: Mixing, nucleation and growth. Acta Mater 103:301–310
7. Khalaf AA, Ashrari P, Shankar S (2009) Controlled Diffusion Solidification (CDS): Conditions for Non-Dendritic Primary Aluminum Phase in Al-Cu Hypo-Eutectic Alloys. In: The Third International Symposium, TMS (The Minerals, Metals & Materials Society). pp 215–222
8. Khalaf AA (2010) Controlled diffusion solidification: process mechanism and parameter study. Ph.D Thesis, McMaster University
9. Khalaf AA, Ashrari P, Shankar S (2009) Formation of nondendritic primary aluminum phase in hypoeutectic alloys in controlled diffusion solidification (CDS): A hypothesis. Metall Mater Trans B 40:843–849
10. Khalaf AA, Shankar S (2011) Favorable environment for a non-dendritic morphology in controlled diffusion solidification. Metall Mater Trans A 42:2456–2465
11. Khalaf AA, Shankar S (2012) Effect of mixing rate on the morphology of primary Al phase in the controlled diffusion solidification (CDS) process. J Mater Sci 47:8153–8166
12. Apelian D, Makhlouf MM, Saha D (2006) CDS Method for Casting Aluminum-Based Wrought Alloy Compositions: Theoretical Framework. In: Materials Science Forum. pp 1771–1776
13. Symeonidis K (2009) The controlled diffusion solidification process: fundamentals and principles. Ph.D Thesis, Worcester Polytechnic Institute (WPI)
14. Ghiaasiaan R, Zeng X, Shankar S (2014) Controlled Diffusion Solidification (CDS) of Al-Zn-Mg-Cu (7050): Microstructure, heat treatment and mechanical properties. Mater Sci Eng A 594:260–277
15. Pourgharibshahi M, Divandari M, Saghaflan Larijani H, Ashtari P (2017) Controlled diffusion solidification processing: A review. J Mater Process Technol 250:203–219. https://doi.org/10.1016/j.jmatprotec.2017.07.018
16. Yang X, Li YD, Luo XM, Zhou HW, Cai QY, Li MI, Ma Y (2019) Microstructural evaluation and mechanical properties of 7075 aluminum alloy prepared by controlled diffusion solidification. China Foundry 16:238–247. https://doi.org/10.1007/s41230-019-9059-9
17. Ghiaasiaan R, Shankar S (2020) Microstructure, intermetallic phases, and fractography of the cast Al-5.8Zn-2.2Mg-2.5Cu alloy by controlled diffusion solidification. Metall Mater Trans A Phys Metall Mater Sci 51:4711–4726. https://doi.org/10.1007/s11661-020-05885-z
18. Khalaf AA (2021) Studying of non-dendritic microstructure forming in controlled diffusion solidification. Int J Met. https://doi.org/10.1007/s40962-021-00590-y
19. Khalaf AA, Shankar S (2020) Mechanism of anomalous grain formation during controlled diffusion solidification. JOM. https://doi.org/10.1007/s11837-020-04198-1
20. Khalaf AA (2022) Metastable transition temperature in undercooled hypereutectic Al-Si alloys. Scr Mater 210
21. Khalaf A (2022) Microstructure evolution of Al-Si hypoeutectic alloys prepared by controlled diffusion solidification. Int J Adv Manuf Technol 1–22. https://doi.org/10.1007/s00170-022-09075-6
22. Li Y, Zhang X, Ma Y et al (2015) Effect of mixing rate and temperature on primary Si phase of hypereutectic Al-20Si alloy during controlled diffusion solidification (CDS) process. China Foundry Res Dev 12:173–179
23. Khalaf AA, Takrouri KJ (2020) Spontaneous nucleation in hypoeutectic Al–Cu system by controlled diffusion solidification. SN Appl Sci 2:1337
24. Wu M, Ludwig A (2009) Modeling equiaxed solidification with melt convection and grain sedimentation-II. Model verification Acta Mater 57:5632–5644. https://doi.org/10.1016/j.actamat.2009.07.067
25. Kurz W, Fisher DJ (1984) Fundamentals of Solidification, 2nd edn. Trans Tech Publications, Aedermannsdorf

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