Modeling of microporosity formation during solidification of aluminum alloys

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Abstract. A two-dimensional (2D) multi-phase cellular automaton (MCA) model is adopted to simulate the dendrite and microporosity formation during solidification of aluminium alloys. The model involves three phases of liquid, gas, and solid. The effect of liquid-solid phase transformation on the nucleation and growth of porosity, the redistribution and diffusion of solute and hydrogen, and the effects of surface tension and environmental pressure are taken into account. The growth of both dendrite and porosity is simulated using a CA approach. The diffusion of solute and hydrogen is calculated using the finite difference (FD) method. The simulations can reveal the interactive and competitive growth of dendrites and micropores, and the microsegregation of solute and hydrogen. The porosity nuclei with large size are able to grow preferentially, while the growth of the small porosity nuclei is inhibited. Gas pores grow spherically when it is enveloped by liquid. After touching with dendrites, the shapes of pores become irregular. An increased initial hydrogen concentration reduces the incubation time of porosity nucleation, but increases the final percentage of porosity and the average porosity size at the eutectic temperature. With cooling rate decreasing, the competitive growth between gas pores becomes more evident, leading to non-uniform porosity sizes, and more irregular morphology of the porosities with larger size. The simulation results are compared reasonably well with the experimental data reported in literature.

1. Introduction
Castings of lightweight materials, such as aluminum and magnesium alloys, are widely used in the automotive and aerospace industries since they play a significant role in improving the performance and fuel efficiency [1]. The formation of microporosity in castings is, however, considered to be one of the primary defects affecting their mechanical properties, which is mainly caused by the insufficient liquid feeding and gas evolution during solidification [2].

Extensive work has been performed to simulate the microporosity formation during solidification of aluminum and other alloys over the last decades. Most models, including analytical solutions, criteria functions, Darcy’s law coupled with the conservation and continuity equations, and gas diffusion-controlled pore growth models, mainly focus on the prediction of the amount of porosity in castings, but without reproducing graphical morphology [3-10]. See et al. [11] compared deterministic (DE) model, continuum stochastic (CS) model and cellular automaton (CA) model for the prediction of microporosity in aluminum-silicon alloys. They considered that CA model provided an excellent tool for further exploration of the complex interaction between porosity formation and the developing microstructures. Lee et al. [12-15] developed the CA approach coupled with hydrogen diffusion controlled growth models to simulate the meso-scale morphologies of grain structure and porosity in...
two and three dimensions. In the models the internal pore pressure was compared with the surface pressure to determine the pore growth. Dong et al. [16], Conley et al. [17] and Bayani et al. [18] proposed CA based models to simulate the evolution of grains or dendrites and micro-shrinkages during solidification. Karagadde et al. [19] adopted an explicit enthalpy scheme coupled with a level-set (LS) method to capture the final pore morphology during the solidification of an aluminum alloy. The present authors [20, 21] proposed a 2D multi-phase cellular automaton-finite difference (MCA-FD) model, to simulate dendrite and hydrogen porosity formation during solidification of Al alloys. The pore growth was determined based on the difference of the local hydrogen saturation and hydrogen concentration. The model is able to reasonably describe a complete time-dependent interaction of dendrite solidification, the nucleation and growth of porosities, solute and hydrogen partitions at the interfaces, and the diffusion of both solute and hydrogen.

In this paper, the previously proposed MCA-FD model is adopted to simulate the nucleation and growth of microporosity, the interactions between dendrites and micropores, and the microsegregation evolution of hydrogen during solidification of an Al-7 wt.% Si alloy. The effects of the initial hydrogen concentration and cooling rate on the formation of microporosity are investigated.

2. Governing equations and numerical algorithm

2.1. Model description

The focus of the present work is to simulate the microporosity formation caused by the difference of hydrogen solubility between the liquid and solid phases. The effects of buoyancy, Marangoni flow, and external forces are not considered. Therefore, the formation of gas porosities and dendrites are driven by pure diffusion. In addition, the temperature in the computational domain is assumed to be uniform and cooled down with a constant cooling rate. The simulation is stopped when the temperature drops to the eutectic temperature.

2.2. Pore nucleation

A stochastic model is implemented to assign the locations of pore nuclei randomly. The distribution function \( dn / dS \) used for evaluating the number of gas pore nuclei is defined as

\[
\frac{dn}{dS} = \begin{cases} 
\frac{N_{\text{max}}}{(S_{\text{max}} - S_{\text{min}})} & \text{if } (C_s(H)/S_l(H) > S_n) \\
0 & \text{if } (C_s(H)/S_l(H) \leq S_n)
\end{cases}
\]

where \( N_{\text{max}} \) is the maximum pore nucleation density, \( S_{\text{max}} \) and \( S_{\text{min}} \) are the maximum and minimum pore nucleation saturation, respectively [10, 15]. \( S_n \) is the critical saturation criterion for porosity nucleation. \( C_s(H) \) and \( S_l(H) \) are the local hydrogen concentration (mol/m\(^3\)) and the local hydrogen saturation in liquid, respectively. When the local hydrogen concentration in liquid reaches a state of supersaturation, a pore nucleus with a certain radius might occur. According to Sievert’s law, the hydrogen saturation in liquid, \( S_l(H) \), of Al-Si alloys can be calculated by [10]

\[
S_l(H) = \sqrt{\frac{P_g}{P_{\text{ref}}}} \left( \frac{2760}{T} + 2.96 \cdot 10^{-5} W_S \right)
\]

where \( P_g \) is the internal pressure of a gas pore, \( P_{\text{ref}} \) is the reference pressure defined as the standard atmospheric pressure that is taken as 1 atm in this work, \( T \) is the temperature, and \( W_S \) is the mass fraction of solute Si. The internal pressure is affected by two factors: the atmospheric pressure and surface energy pressure, and the effects of the metallostatic pressure and shrinkage pressure are not considered. Therefore, \( P_g \) in the two dimensions is given by: \( P_g = P_{\text{ref}} + \lambda_{\text{LG}}/r_p \), where \( \lambda_{\text{LG}} \) is the surface tension between liquid and gas, and \( r_p \) is the radius of the gas pore. Based on the above equations, the radii of gas pore nuclei formed under different hydrogen saturations can be calculated.
### 2.3. Microporosity and dendrite growth

The CA technique is adopted to simulate the growth of both microporosity and dendrite. The two-dimensional computational domain is divided into a lattice of square cells. Each cell is characterized by different variables, including hydrogen concentration, solute concentration, crystallographic orientation of a dendrite, temperature, phase fractions (solid, liquid, and gas), and cell states including liquid cell \((f_s+f_g=0)\), solid cell \((f_s=1)\), the gas/liquid (G/L) interface cell \((0<f_g<1, \text{ and } f_s=0)\), the solid/liquid (S/L) interface cell \((0<f_s<1, \text{ and } f_g=0)\), and the gas/liquid/solid (G/L/S) interface cell \((0<f_s+f_g<1)\), where \(f_g\) and \(f_s\) are the gas fraction and solid fraction of each cell, respectively. The cells that belong to a same pore share the characteristics of this pore, such as the hydrogen concentration, the internal pressure and the volume of the pore.

Assuming that a gas pore denoted as \(A\) obeys the ideal gas law, the gas volume increment of the G/L interface cell \((i,j)\) that belongs to the pore \(A\) can be calculated by

\[
\Delta V^{(i,j)}_A = R_H \frac{G^{(i,j)}_g RT}{P_g} \sum G^{(i,j)}_s \Delta \left( \frac{f_s}{f_g} \right)
\]

where \(R_H\) is the quantity of hydrogen absorbed by all G/L interface cells of the pore \(A\), \(R\) is the gas constant, \(G^{(i,j)}_g\) is the geometrical factor of the G/L interface cell, \((i,j)\), and \(\sum G^{(i,j)}_s\) is the sum of all the geometrical factors of all G/L interface cells in pore \(A\). The geometrical factor \(G_s\) is related to the state of neighboring cells and is defined by

\[
G_s = \min \left[ 1, \frac{1}{2} \left( \sum_{m=1}^4 S'_m + \frac{1}{\sqrt{2}} \sum_{m=1}^4 S''_m \right) \right]
\]

where \(S'\) and \(S''\) indicate the state of the nearest neighbor cells and the second-nearest neighbor cells, respectively, and \(f_g\) is the gas fraction of neighbor cells. The geometrical factor defined by equation (4) is used to account for the effect of neighbor cell state on pore growth. Equation (4) also reflects the effect of the second nearest neighbor cells is weaker than those of the nearest cells.

Therefore, the gas fraction of the G/L interface cell in pore \(A\) at time \(t_n\) can be defined as

\[
f^{(i,j)}_{g,A} = \sum_{m=1}^N \frac{\Delta V^{(i,j)}_A}{V_c}
\]

where \(N\) is the iteration number, and \(V_c\) is the volume of cell. After a time step interval, the gas fraction of a G/L interface cell of pore \(A\) is updated using equation (5). When the gas fraction of the G/L interface cell becomes 1 through \(N\) time iterations, the cell state is changed from the G/L interface to gas pore. Then, the newly formed gas cell captures its liquid neighboring cells to be the new G/L interface cells. The gas fractions of these new interface cells are also calculated using equation (5) in the next time step, and thus the pore growth continues.

The dendrite growth is also simulated by the CA approach. The governing equations and numerical algorithm for the simulation of dendrite growth can be found elsewhere [20].

### 2.4. Solute and hydrogen diffusion

During the process of dendrite growth and gas pore formation, a solute gradient exists ahead of the S/L interface, while a hydrogen gradient exists ahead of both the S/L and G/L interfaces due to the solute and hydrogen partitions at the interfaces. The solute and hydrogen gradients lead to the solute and hydrogen diffusion in the domain. The partitions and diffusions of solute and hydrogen are calculated by

\[
\frac{\partial C_i(X)}{\partial t} = D(X) \cdot \nabla^2 C_i(X) + R(X)
\]
where $C(X)$ is the concentration, $D(X)$ is the diffusion coefficient, $X$ represents solute silicon or hydrogen, the subscript $i$ represents solid or liquid, respectively. $R(Si)$ indicates the amount of solute rejected at the S/L interface, and $R(H)$ includes hydrogen partitions at the S/L interface and the G/L interface. The solute silicon and hydrogen partitions between liquid and solid at the S/L interface are considered according to

$$C_i^s(X) = k_s C_i^l(X)$$  \hspace{1cm} (7)

where $C_i^s(X)$ and $C_i^l(X)$ are the interface silicon or hydrogen concentrations in solid and liquid phases, respectively, and $k_s$ is the partition coefficient. Therefore, silicon and hydrogen partitions at the S/L interface in one time step can be obtained by

$$R(X)_{s/l} = \begin{cases} \Delta f_s \left( C_i^s(Si)(1-k_s) \right); (X = Si) \\ \Delta f_s \left( C_i^s(H)(1-k_H) \right); (X = H) \end{cases}$$  \hspace{1cm} (8)

On the other hand, the amount of hydrogen absorbed by the pore, $R(H)_{G/L}$, is evaluated by

$$R(H)_{G/L} = \sum_j (1 - f_s^{i(j)} - f_s^{i(j)})(C_i(H) - S_i(H))V_i$$  \hspace{1cm} (9)

where $j$ represents all G/L interface cells of a same pore, and $f_s^{i(j)}$, $f_s^{i(j)}$, $C_i(H)$ and $S_i(H)$ are the gas fraction, solid fraction, hydrogen concentration, and hydrogen saturation in liquid, respectively, at the G/L interface cell.

Equation (6) is solved using an explicit finite difference schedule. It is considered that the diffusion coefficient of hydrogen in liquid is about two orders of magnitude larger than that of solute Si, two different time steps are thus used in the simulations to increase the computational efficiency. The time step for the calculations of the solute Si diffusion, the growth of both dendrites and gas pores, is determined by

$$\Delta t = \min \left\{ \frac{\Delta x^2}{4.5D(Si)} , \frac{1}{4.5\Delta f_{s,max}} \right\}$$  \hspace{1cm} (10)

where $D(Si)$ is the diffusion coefficient of solute Si in liquid, and $\Delta f_{s,max}$ is the maximum increment of solid fraction in one time step. On the other hand, the time step for hydrogen diffusion, $\Delta t_{H}$ is determined by $\Delta t_{H} = \Delta x^2 / 4.5D(H)$, where $D(H)$ is the diffusion coefficient of hydrogen in liquid. In the simulations, if the ratio of two time steps is $N_i = \Delta t / \Delta t_{H}$, the calculation of hydrogen diffusion is run for $N_i$ times using the time step $\Delta t_{H}$. Then, the diffusion of solute silicon, and the growth of dendrites and gas pores are calculated using $\Delta t$ for one time step.

2.5. Coupling growth of microporosity and dendrite

In the present work, the CA approach [20] is used to simulate the dendrites growth. The solute Si and hydrogen rejected from the newly formed solid cells are calculated by equation (8) and then $R(X)_{s/l}$ is added into the remaining liquid in the same S/L interface cell and its surrounding neighbouring cells to keep the conservation of solute Si and hydrogen in the domain. Then, the diffusions of solute and hydrogen are calculated by equation (6). With dendrites growing, the local hydrogen concentration increases. Equation (1) is used to estimate the possibility of gas pore nucleation. If a gas pore is nucleated, the increasing gas fraction is calculated by equations (2)–(5) and (9), and the radius of the gas pore is updated. This series of calculations are repeated until the end of simulation. The physical parameters used in this work can be found elsewhere [20].
3. Results and discussion

3.1. Evolution of microporosities and dendrites

Figure 1 shows the evolution of porosities and dendrites of an Al-7 wt.% Si alloy with a cooling rate of 8° C/s and an initial H concentration of 0.6 mol/m$^3$. The calculation domain consists of a 300×300 mesh with a uniform mesh size of 3 μm. The pore nucleation parameters $N_{max}$, $S_{max}$, $S_{min}$, and $S_n$ are taken as $1 \times 10^{11}$ m$^{-3}$, 1.6, 2.3, and 1.2, respectively. These parameters are taken as constant in all simulations in the present work. Locations for each pore nucleation and solid seeds with various crystallographic orientations are assigned randomly in the domain. As shown in figure 1, with the temperature decreasing, dendrites grow with developing the main arms along their crystallographic orientations, and side branching of the secondary arms. Meanwhile, the H concentration increases in liquid and it is higher in the area close to dendrites (figure 1 (a)). According to equation (2), the solubility of hydrogen decreases with decreasing temperature. When the local H concentration in liquid is higher than the supersaturation for porosity nucleation, several gas pore nuclei with different radii appear (figure 1 (b)). Owing to the effect of surface tension, the gas pores grow spherically in liquid until they have impinged upon dendrites (figure 1(c)). After the gas pores encounter the dendrites, the shapes of gas pores become irregular (figure 1(d)). Since the pore nucleation and growth consume H atoms, the H concentration in liquid, shown in figure 1(d), is lower than those in figures 1 (a)-(c), and even lower than the initial concentration of 0.6 mol/m$^3$, except the bottom right of the domain, due to the fact that no gas pore appears in the area. It also can be seen that some pores with relatively smaller size barely grow, even though the local H concentration around them is actually higher than that around the pores grow obviously. This is because the smaller the pores, the higher the internal pressure. Thus, the smaller pores have higher H solubility than that of the larger pores (equation (2)), leading to a smaller gas volume increment (equations (3) and (9)). Therefore, the pores with larger size are able to grow preferentially, while the growth of the small pores is limited.

![Figure 1](image)

**Figure 1.** Evolution of microporosity, dendrite and H concentration field of an Al-7wt.%Si with a cooling rate of 8°C/s and an initial H concentration of 0.6 mol/m$^3$ at different temperatures: (a) 608.5°C, $F_s=4.0\%$, $F_g=0$; (b) 601.1°C, $F_s=20.0\%$, $F_g=0.16\%$; (c) 590.8°C, $F_s=43.5\%$, $F_g=0.41\%$; (d) 572.8°C, $F_s=49.3\%$, $F_g=1.44\%$. (Numbers in the figures show the local H concentration in liquid phase. $F_s$ and $F_g$ are the total solid fraction and percentage of porosity in the domain, respectively).

3.2. Effect of initial hydrogen concentration

Figure 2 shows the simulated H concentration fields and morphologies of dendrites and microporosities at the eutectic temperature with different initial H concentrations of 0.5 mol/m$^3$ and 0.7 mol/m$^3$. The other conditions are identical with those of figure 1. It can be seen that porosity size becomes larger with the higher initial H concentration. It is understandable that with a higher initial H concentration, the amount of hydrogen atoms rejected from the growing dendrites increases (equation (7)), leading to the increase of the local H concentration. More hydrogen atoms in liquid are available for pore growth (equations (8) and (9)). Therefore, the higher initial H concentration will produce the pores with larger size. Since the large pores absorb more hydrogen atoms, the H concentration in liquid at the eutectic temperature for the case of a higher initial H concentration is even lower than that of the lower initial H concentration as shown in figure 2.
Figure 3 represents the percentage of porosity varying with solidification time with a cooling rate of 8°C/s and different initial H concentrations. Other simulation conditions are identical with those of figure 1. It is observed that the porosity percentage increases slowly at the early stage, then increases rapidly, and slows down again at the late stage. It is obvious that there is an incubation period for pore nucleation, the higher the initial H concentration, the shorter the incubation time. It is understandable that the higher initial H concentration results in a higher local H concentration in liquid, which shortens the time required to reach the supersaturation for pore nucleation. It also can be seen from figure 3 that the final percentage of porosity of the case with higher initial concentration is larger than that of the lower one. Figure 4 shows the average pore radius at the eutectic temperature as a function of initial H concentration with a cooling rate of 8°C/s. It can be seen the average pore radius at the eutectic temperature increases with increasing initial H concentration when the initial H concentration is higher than 0.4 mol/m³. However, when the initial H concentration is lower than 0.4 mol/m³, the average pore radius is relatively small. It seems that there is a critical level of the initial H concentration, below which gas pores could barely grow.

![Figure 3. Percentage of porosity as a function of time with a cooling rate of 8°C/s and various initial H concentrations.](image1)

![Figure 4. Average pore radius at the eutectic temperature as a function of initial H concentration with a cooling rate of 8°C/s.](image2)

### 3.3. Effect of cooling rate

Figure 5 shows the simulated H concentration field and morphologies of dendrites and microporosities at the eutectic temperature with different cooling rates of 2°C/s, 8°C/s and 15°C/s. Other simulation conditions are identical with those of figure 1. It is observed that with a higher cooling rate, the
dendrites become finer, and the sizes of finally formed pores are smaller and more uniform. These results are coincident with experimental observations [22, 23]. Dendrites grow rapidly with finer and longer arms at a higher cooling rate. Thus, the complex dendrite network could be formed sooner, which blocks hydrogen diffusion in liquid. Therefore, less hydrogen atoms are available at the G/L interface, leading to the growth of porosities to be inhibited. Moreover, a higher cooling rate also reduces the time for hydrogen diffusion and pore growth. Since less hydrogen atoms are consumed by gas pores at a higher cooling rate, the H concentration in liquid at the eutectic temperature increases with increasing cooling rate as shown in figure 5.

Figure 5. Hydrogen concentration fields and morphologies of dendrite and microporosity with an initial H concentration of 0.6 mol/m$^3$ and different cooling rates: (a) 2°C/s, (b) 8°C/s, (c) 15°C/s.

Figure 6 shows the percentage of porosity as a function of temperature with different cooling rates. The initial H concentration is 0.6 mol/m$^3$. Other simulation conditions are identical with those of figure 1. As shown, with increasing cooling rate, gas pores nucleate and start to grow at lower temperatures. The final percentage of porosity tends to decrease as the cooling rate increases, which can explain the reason why the H concentration in liquid at the eutectic temperature shown in figure 5 (c) is higher than that in figure 5 (a) and (b). The tendency of the porosity percentage varying with cooling rate shown in figure 6 is consistent with the experimental data obtained by Fang and Granger [24], and Lee and Hunt [25], as well as the simulation results obtained by Felicelli et al. [6], and Carlson et al. [7]. Figure 7 represents the maximum pore radius as a function of cooling rate with an initial H concentration of 0.6 mol/m$^3$. It is found that with decreasing cooling rate, the final maximum porosity radius decreases obviously. This implies that at a slow cooling rate, the pore nuclei with relative larger size tends to grow preferentially, while the growth of small pore nuclei is limited, leading to the formation of non-uniform porosities as shown in figure 5 (a).

Figure 6. Percentage of porosity as a function of cooling rate with an initial H concentration of 0.6 mol/m$^3$.

Figure 7. Maximum pore radius as a function of cooling rate with an initial H concentration of 0.6 mol/m$^3$. 
4. Conclusions
A previously proposed 2D multi-phase cellular automaton-finite difference (MCA-FD) model is adopted to simulate the evolution of porosities and dendrites during solidification of an Al-7wt.%Si alloy. The nucleation and growth of porosities, dendrite solidification, solute and hydrogen partitions at the interface, and the diffusion of both solute and hydrogen are involved in the model. The interactive and competitive growth of dendrites and porosities, and the microsegregation of solute and hydrogen are observed. It is found that the pores with relatively larger size grow preferentially, while the growth of small porosity nuclei is limited. The gas pores grow spherically until they have impinged upon the solid dendrites.

The effects of the initial H concentration and cooling rate on microporosity formation are investigated. The results show that as the initial H concentration increases, the incubation time of porosity nucleation tends to decrease, while the final percentage of porosity and the average porosity size at the eutectic temperature increases. With cooling rate decreasing, the percentage of porosity increases, and the competitive growth between porosities becomes more obvious. Thus, a low cooling rate leads to non-uniform porosity sizes and increased maximum porosity size. The simulation results agree reasonably well with the experimental data.

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References
[1] Ambroziak A and Korzeniowski M 2010 Arch. Civ. Mech. Eng. 15
[2] Kim J R and Abbaschian R 2011 J. Mater. Sci. 46 6213
[3] Li K D and Chang E 2004 Acta Mater. 52 219
[4] Ferreira I L, Lins J F C, Moutinho D J, Gomes L G and Garcia A 2010 J. Alloy Compd. 503 31
[5] Voller V R 1998 Can. Metall. Quart. 37 169
[6] Felicelli S D, Wang L, Pita C M and Obaldia E E D 2009 Metall. Mater. Trans. B 40B 169
[7] Carlson K D, Lin Z, and Beckermann C 2007 Metall. Mater. Trans. B 38B 541
[8] Pequet C, Gremaud M and Rappaz M 2002 Metall. Mater. Trans. A 33A 2095
[9] Spittle J A, Almehedani M and Brown S G R 1994 Cast Met. 7 (1) 51
[10] Atwood R C, Sridhar S, Zhang W and Lee P D 2000 Acta Mater. 48 (2) 405
[11] See D, Atwood R C and Lee P D 2001 J. Mater. Sci. 36 3423
[12] Lee P D and Hunt J D 2001 Acta Mater. 49 1383
[13] Lee P D, Atwood R C, Dashwood R J and Nagaumi H 2002 Mater. Sci. Eng. A 328 213
[14] Atwood R C and Lee P D 2003 Acta Mater. 51 5447
[15] Lee P D, Chirazi A, Atwood R C and Wang W 2004 Mater. Sci. Eng. 365 57
[16] Dong S Y, Xiong S M and Liu B C 2004 Mater. Sci. Technol. 20 23
[17] Conley J G, Huang J, Asada J and Akiba K 2000 Mater. Sci. Eng. A 285 49
[18] Bayani H, Mirbagheri S M H, Barzegari M and Firoozi S 2014 J. Mater. Res. Technol. 3 55
[19] Karagadde S, Sundarraj S and Dutta P 2012 Comput. Mater. Sci. 65 383
[20] Zhu M F, Li Z Y, An D, Zhang Q Y and Dai T 2014 ISIJ Int. 54 384
[21] Li Z Y, Dai T and Zhu M F 2013 Acta Metall. Sin. 49 1032
[22] Lashkari O, Yao L, Cockcroft S and Maijer D 2009 Metall. Mater. Trans. A 40A 991
[23] Zhao L 2012 PhD Thesis (Nanjing: Southeast University)
[24] Fang Q T and Granger D A 1989 AFS Trans. 97 989
[25] Lee P D and Hunt J D 1997 Acta Mater. 45 4155