Coupling in situ synchrotron X-ray tomographic microscopy and numerical simulation to quantify the influence of intermetallic formation on permeability in aluminium–silicon–copper alloys

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Abstract

The influence of the $\beta$-Al$_5$FeSi intermetallic phase on permeability evolution during solidification in an Al–Si–Cu alloy with a columnar dendritic microstructure has been numerically studied at solid fractions between 0.10 and 0.85. The fluid flow simulations were performed on a semisolid microstructure extracted directly from a single solidifying specimen, enabling the first study of permeability variation on an individual microstructure morphology that is evolving in solid fraction. The 3-D geometries were imaged at the TOMCAT beamline using 4-D (3-D + time) in situ synchrotron-based X-ray tomographic microscopy. The results illustrate the major effect of intermetallic particles on flow blockage and permeability. Intermetallics that grow normal to the flow direction were found to have a greater impact on the flow field in comparison to intermetallics in the parallel flow direction. An analytical expression, based on the anisotropic Blake–Kozeny model, was developed with a particle blockage term that takes into account the effects of intermetallic particles on permeability. In the regime of primary-phase solidification, a good fit between the analytical expression and the simulation results is found.

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1. Introduction

Defect formation during industrial metal alloy casting processes is closely linked with fluid flow in the mushy zone since the resistance to flow through the solid network causes a pressure drop in the liquid that can contribute to porosity formation [1], macrosegregation [2] and hot tearing [3,4]. Understanding the science behind these defect formation mechanisms will provide the information to create better-designed alloys. One parameter used to characterize the fluid flow in the mushy zone is permeability, i.e. a tensor measure of the ease of fluid flow through the solid network [5]. Permeability is a critical parameter in macroscopic casting models for predicting defects and is related to the geometry of the liquid channels and the grain morphology [6,7].

Iron is a common impurity element in aluminium alloys that is difficult to remove during processing. In Al–Si and Al–Si–Cu foundry alloys, the iron impurities may form a $\beta$-Al$_5$FeSi intermetallic phase that can be detrimental to the mechanical properties of these alloys [8,9]. The presence
of the $\beta$-Al$_3$FeSi phase may also increase the porosity content [10,11], and it has been suggested that this might be due to the intermetallic phase; causing flow blockage in the interdendritic channels [12], acting as pore nucleation sites [13] and/or aiding pore growth [14]. However, the effect of intermetallic formation on permeability remains an open question.

In semisolid aluminium alloys, flow of liquid metal through the evolving interdendritic channels is driven by an approximate 7% volumetric shrinkage [15] associated with the liquid-to-solid phase transformation. The permeability of such alloys has traditionally been measured using an apparatus that applies a pressure gradient to a partially solid sample while measuring the discharge velocity of a working fluid. Studies using this type of device (e.g. [16–23]) have focused on measuring the near-eutectic permeability of equiaxed dendritic structures while attempting to reduce errors associated with the tendency of the microstructure to change during testing due to coarsening and other diffusion processes [24].

Alternatively, it is possible to develop predictive numerical models assessing permeability as detailed in Refs. [25,26]. Numerical models solve the Stokes equations for a domain representing the interdendritic liquid. The obvious advantage of using a simulated microstructure is that the evolution of permeability with solid fraction can be easily studied for different grain sizes and morphologies. However, the main challenge with this method is the geometry of the domain itself. Since permeability is a characteristic of porous media that is based on the geometry of the flow channels, i.e. channel width, surface area and tortuosity, an accurate computational domain is critical for numerical determination of this parameter [27].

Recently, attention has been focused on using 3-D synchrotron X-ray tomographic microscopy (also known as synchrotron X-ray tomography) to acquire representative semisolid geometries for assessment of permeability during equiaxed dendritic [25] and columnar [28] solidification, and at both near-eutectic [26,27] and low-eutectic [29] alloy compositions as a function of solid fraction. In these recent studies, the tomographic imaging was performed on quenched samples followed by image analysis to distinguish the eutectic phase (interdendritic liquid) from the dendrite skeleton. The results have shown a generally good agreement between the calculated permeability and experimental reference data [25,28].

Numerically, permeability is defined by Darcy’s law as [30]:

$$v = \frac{K}{\mu_L} \nabla p_L - \rho_L g$$

(1)

where $K$ is the specific permeability of the porous medium, $\mu_L$ is the viscosity, $v$ is superficial flow velocity of the liquid, $\rho_L$ is the liquid mass density, $g$ is the acceleration due to gravity and $\nabla p_L$ is the pressure gradient in the liquid. For equiaxed dendritic structures, the isotropic Carman–Kozeny equation [31] is most often adopted to approximate the permeability in semisolids. However, for columnar dendritic structures, the permeability is anisotropic due to the directional nature of the dendrite arms [17], and thus the Carman–Kozeny equation is not appropriate.

To investigate anisotropy in permeability, Poirier [6] used a multilinear regression analysis of experimental data to propose an empirical relationship based on the Blake–Kozeny equation. Schneider et al. [32] and Heinrich and Poirier [33] continued work in this area and proposed multiple expressions for different ranges of liquid fraction. In an alternate approach, Santos and Melo [34] used the Hagen–Poiseuille flow equation and derived a physically based permeability expression for flow both parallel to and normal to the primary dendrite arms that included terms for tortuosities of the interdendritic liquid channels in directions parallel to and normal to the primary dendrites. In both cases, the permeability predictions were found to be in good agreement with experimental data.

In this study, the influence of intermetallic formation on permeability in columnar dendritic structures has been investigated in an Al-A319 alloy that forms the $\beta$-Al$_3$FeSi phase during solidification. Specifically, numerical fluid flow simulations were performed after capturing geometries of the semisolid microstructures using 4-D (3-D + time) in situ synchrotron X-ray tomographic microscopy. Simulations both with and without intermetallics were conducted. Unlike previous numerical studies of permeability that utilized quenched specimens in combination with post-mortem tomographic imaging [25–29], this work has extracted the geometry directly from a microstructure captured during solidification. This is the first use of in situ 4-D tomographic microscopy for such studies, and the results provide a novel, detailed permeability assessment of an individual microstructure morphology that is changing both in solid fraction and in intermetallic phase evolution rather than assessing a post-mortem set of snapshots typically given by quenched experiments.

2. Methods

2.1. In situ observation via synchrotron X-ray tomographic microscopy

The in situ solidification experiment was performed on the TOMCAT beamline of the Swiss Light Source (Paul Scherrer Institut, Switzerland) [35] using a bespoke induction furnace [36]. The experiment was conducted on a commercial Al-A319 alloy with composition Al–7.52Si–3.53Cu–0.59Fe (wt.%) and a secondary dendrite arm spacing of ~30 μm. The specimen geometry was a cylinder 2.5 mm in diameter and 4 mm in length. To contain the molten metal, the specimen was placed in a high-purity boron nitride (BN) holder, and temperatures were recorded using a K-type thermocouple placed underneath the holder. Heat extraction to induce solidification was achieved via ambient furnace cooling. There was a small thermal gradient present in the furnace, which resulted in
the solidification microstructure being dendritically elongated in the heat extraction direction.

The experimental procedure consisted of first gradually heating the specimen to 650 °C, then applying a 5 min hold to ensure thermal equilibrium, and finally cooling to below the solidus temperature (505 °C) of the specimen at a rate of 0.05 °C s⁻¹. During the cooling stage, 1001 radiographs were captured over 180° of rotation in 2 s, and this image acquisition sequence was repeated every 60 s. From these images and the corresponding angular information, tomographic 3-D volumes of 1104 × 1104 × 1468 voxels with a voxel size of 2.75 μm per side were generated using standard reconstruction algorithms [37,38]. Fig. 1 shows a 3-D rendering of the full dendritic structure that was captured at 582 ± 1 °C, corresponding to a solid volume fraction of 0.27. The calculation of solid fraction was done by applying a marching cubes algorithm [39] on the segmented solid phase. The process of image segmentation is detailed in the following section. In total, 13 3-D volumes were acquired during the solidification sequence over a solid fraction range between 0 and 0.85. Although the time between sequential 3-D volumes was ~1 min due to camera memory limitations, the fast tomographic imaging (2 s) and slow cooling rate ensured that the microstructural evolution during acquisition of the radiographs for a single 3-D volume was small, minimizing movement artifacts.

2.2. Image segmentation

After image acquisition, significant post-processing was necessary to segment the three phases: primary solid, intermetallics and liquid. First, all of the datasets were rotated to align the primary dendrites with the x-axis in order to reduce errors in the flow studies. Second, a subvolume of 400³ voxels was extracted from each 3-D volume to reduce the computational cost. This volume meets the mesh size and physical length scale requirements for a representative volume element previously identified by Bernard et al. [25] and Fuloria et al. [28]. Third, a 3-D anisotropic diffusion filter [40] was applied to each dataset to reduce noise. Last, 3-D region growing segmentation [41] and manual segmentation processes were applied to separate the Al-rich dendrites along with Al–Si eutectic (if present) and the intermetallics from the solidifying liquid. An example of this process is shown in Fig. 2: Fig. 2a corresponds to the original data rotated and a smaller volume selected (i.e. the highlighted box in Fig. 1); Fig. 2b and c correspond to the segmented Al-rich and intermetallic phases, respectively; Fig. 2d and e correspond to the full segmented datasets both without and with intermetallics, respectively, for use as input geometries for the numerical simulations.

2.3. Flow simulation

In order to determine the evolution of permeability in the semisolid Al-A319, the flow problem based on the Stokes equations was simulated in the 13 tomographic datasets using the commercial computational flow
dynamics (CFD) software Avizo XLab Hydro (VSG, France). This software, developed based on research by Bernard et al. [25], utilizes a finite volume method applied directly to a tomographic dataset for flow predictions. The Stokes equation describing interdendritic flow at the microscopic scale can be expressed as:

\[-\nabla p + \mu \nabla^2 v = 0\]  \hspace{1cm} (2)

\[\nabla \cdot v = 0\]  \hspace{1cm} (3)

assuming gravity is ignored. 50 Voxel layers of divergence and convergence channels were added to both the inlet and outlet faces in order to create a fluid stabilization zone where the pressure is quasi-static and the liquid can freely spread on the inlet face [42].

The flow studies were performed in each of the three directions along the x, y and z axes of the 3-D volume, both with and without intermetallics. As the columnar dendrites were aligned with the x-axis, inlet flow along the x-axis gives the permeability for liquid flow parallel to the primary arms, while permeability in the normal direction was taken as the average value calculated with inlet flows along the y- and z-axes. The boundary conditions for the flow simulations were as follows: a low Reynolds number velocity inlet (Re < 0.1), free pressure outlet (0 Pa), no-slip conditions on the remaining four outer walls, and no-slip conditions on the interior walls representing solid–liquid interfaces. The liquid viscosity was given a value of 0.001 Pa s.

3. Results and discussion

Cross-sectional slices taken from the 3-D tomographic datasets showing the semisolid microstructure of Al–A319 are shown in Fig. 3 for 582 ± 1, 575 ± 1, 568 ± 1 and 555 ± 1 °C. It can be seen that the contrast between each phase is excellent. Primary Al-rich dendrites and Al–Si eutectic appear dark, the Fe-rich intermetallic phase appears very bright, and the solidifying liquid is grey. This high contrast allowed for good segmentation of the structure for the flow simulations. As solidification proceeds, the individual dendrites grow (Fig. 3a), intermetallics form (Fig. 3b) and finally the Al–Si eutectic phase forms in the interdendritic regions, which completes the solidification process (Fig. 3d). Concurrently, the primary and secondary dendrite arms appear to be coarsening (Fig. 3b and c). Supplementary Fig. S1 illustrates the evolution of primary Al-rich dendrites and Al–Si eutectic formation via 2-D projection mapping.

The experimentally observed evolution of the solid fraction of all phases ($f_S$) and intermetallic fraction alone ($f_I$) both as a function of temperature, along with the corresponding lever rule, and Scheil limits are shown in Fig. 4. Each square ($f_S$) and circle ($f_I$) represents the value obtained from each of the 13 tomographic datasets acquired during solidification. The solid fractions for the four slices given in Fig. 3 are indicated by the dashed lines labelled I–IV, which provide clarity and enable direct comparison between the two figures. The values derived from the 3-D images match closely to the lever-rule approximation for this slowly solidifying case. There is also a good match in the sequence of phase formation, i.e. the tomographic images adequately capture the first appearance of the intermetallic precipitation at 578 ± 1 °C (compared to the nucleation temperature of 580 °C from thermodynamic calculations) as well as the first appearance of the eutectic transformation at 561 ± 1 °C (compared to the nucleation temperature of 565 °C from the prediction). The high degree of similarity in the solidification sequence between the 3-D images and theory provides good confidence in the segmentation methodology and thus the input geometries for the resulting flow simulations; they can be assumed to provide an excellent quantitative characterization of the actual solidification microstructure.

3.1. Flow simulations

The pressure contours and flow velocities resulting from the flow simulations in the x-direction, parallel to the primary dendrites, are shown in Figs. 5 and 6 for various temperatures in the semisolid regime. These images show the liquid domain (i.e. the inverse of the segmented regions shown in Fig. 2) for $f_S$ ranging from 0.30 to 0.56 in the cases without (left column) and with (right column)
intermetallics. It can be seen in Fig. 5 that the pressure required for flow increases with increasing solid fraction, as expected ($f_S = 0.30, 0.36$ and $0.41$, Fig. 5a, c, e, respectively). The addition of intermetallics ($f_S = 0.30, 0.36$ and $0.41$, Fig. 5b, d, f, respectively) causes a significant change in the pressure contours. Comparing Fig. 5c with d, and also Fig. 5e with f, it is clear that there is a strong increase in the pressure required for fluid flow when intermetallics are present. Fig. 5g and h provide a measure of the interaction between the liquid, primary dendrites and intermetallics at $f_S = 0.33$, and show the fluid velocity through the channels in 3-D (complete velocity fields are given in Supplementary Movie M1). The intermetallics are large and plate-like, and it is immediately clear that they will block many of the channels. The blockages will be accommodated by either restricting flow to the few channels that remain unblocked by intermetallics, or by forcing flow back and forth between channels in order to go around the blockages. This second case would be particularly relevant as intermetallic growth progresses and every channel contains intermetallics.

Cross-sectional images showing the fluid velocity magnitudes in the solidifying microstructures are given in Fig. 6. Fluid flow occurs from left to right, and the qualitative effect of the intermetallic is now plainly evident. Beginning with Fig. 6a and b at $f_S = 0.30$, it can be seen that the addition of one intermetallic particle partially blocks the top channel (Fig. 6b, upper left-side plate in white). With increasing solid fraction (i.e. $f_S = 0.36$ (Fig. 6c and d), $f_S = 0.41$ (Fig. 6e and f) and $f_S = 0.56$ (Fig. 6g and h)), the flow fields become more complex. For example, at $f_S = 0.41$ (Fig. 6e and f), the flow has transitioned from a relatively constant velocity without intermetallics (Fig. 6e) to a velocity field that is channel-dependent, with blockages, and sudden jumps in velocity with the presence of the $\beta$-Al$_5$FeSi (Fig. 6f). Note that while it may appear from these flow patterns that mass is not conserved, this is not the case as these images are 2-D cross-sections of 3-D simulations. Liquid that is blocked in plane will circulate out of plane.

From a qualitative perspective, the results shown in Figs. 5 and 6 demonstrate that Fe-containing intermetallics strongly block the interdendritic channels at high solid fractions. Iron impurities are known to increase porosity content and hot tearing. These simulations verify that
channel blockage, as previously theorized [43–45], is indeed one of the causes of increased pore and hot-tearing defect formations. Note that these results do not disprove the other postulated mechanisms; rather, they clearly highlight and quantify the importance of flow effects.

3.2. Permeability assessment

The permeability values extracted from the flow simulations are shown in Fig. 7, for flow (a) parallel and (b) normal to the primary columnar dendrites both without and with intermetallics. The evolution in solid fraction as measured from the tomographic datasets is also provided, including the points for intermetallic and eutectic formation. Beginning with Fig. 7a, it can be seen that the permeability decreases monotonically with temperature as expected because of the thickening of the primary dendrites as solidification progresses. The effect of intermetallics on the calculated permeability values normal to the primary dendrites, shown in Fig. 7b, shows similar

Fig. 6. 2-D cross-sectional views of the calculated 3-D velocity magnitudes showing the fluid flow within the interdendritic region during solidification both without (a, c, e, g) and with (b, d, f, h) intermetallics at solid fractions of (a, b) 0.30 (578 ± 1 °C), (c, d) 0.36 (572 ± 1 °C), (e, f) 0.41 (565 ± 1 °C) and (g, h) 0.56 (561 ± 1 °C). The primary Al-rich dendrites and eutectic are coloured grey while the intermetallics are white.

Fig. 7. The evolution in permeability for liquid flow (a) parallel and (b) normal to the primary columnar dendrites both without and with intermetallics. Dashed-lines indicate the temperatures where the nucleation of intermetallic and eutectic Al phases were observed in the 3-D renderings.
trends to those discussed for the parallel flow in Fig. 7a. However, the additional loss in permeability related to the intermetallic particles is considerably lessened. As shown in Fig. 2, the primary channels in the x-direction are large and continuous, while the channels in the y- and z-directions are already discontinuous, impeding flow. Hence, the addition of intermetallic particles does not increase the channel tortuosity in the normal direction to any great extent. Previous research has shown that the permeability parallel to the columnar dendrites decreases with increasing secondary dendrite arm spacing, \( \lambda_2 \) \cite{[18,34]}. Based on the tomographic datasets presented in this work, the loss of permeability due to increasing \( \lambda_2 \) is most likely linked to channel blocking. When this quantity is large, the actual number of channels will decrease, leading to an increased likelihood of blocking by intermetallic particles or perhaps by stray grain formation, grain motion, etc.

The results shown in Fig. 7 are unique in that they quantify, for the first time, the variation in permeability with solid fraction on the same specimen. These results were made possible due to the in situ acquisition of the tomographic datasets. Other researchers \cite{[25–29]} have performed similar simulations on a series of specimens quenched at various intervals within the semisolid regime to achieve a primary phase fraction matching a specific solid fraction. This previous work provided good insight into permeability development but each specimen had a different microstructure and hence provided considerable scatter in the numerical results. Also with the exception of Ref. \cite{[28]}, this previous work was performed on grain-refined alloys with equiaxed globular grains. Permeability in globular structures will be considerably different in comparison to dendritic structures since there is no well-defined primary channel to drive fluid flow, and further, blocking by intermetallics may be intergranular and hence more irregular. These results, therefore, stand alone in characterizing permeability with regards to columnar dendritic microstructures.

Fig. 8 shows a simulation of interdendritic liquid flow around an actual intermetallic particle. In Fig. 8a, the flow is vertical and down, and hence parallel to the platelet’s orientation, while in Fig. 8b the flow is horizontal and to the right, and hence normal to the platelet. It can be observed that the disturbance around the normal platelet is considerable and thus will significantly affect permeability while the corresponding disturbance around the parallel platelet is minimal. Based on this simple flow-field visualization, it is clear that any permeability assessment involving a channel blocking mechanism must include knowledge of the shape and orientation of the blocking particles. In our solidification sequence, the orientation of the intermetallics in relation to the primary dendrites fits a normal distribution representation with \( \mu = 90^\circ \) and \( \sigma = 35.5 \). As shown in the companion study \cite{[36]}, intermetallics have a preferential growth direction that is parallel to the secondary dendrite arms and hence perpendicular to the primary trunk, which, based on Fig. 8, would increase their blocking efficiency.

3.3. Determination of an analytical permeability expression

The flow simulations presented above enable the influence of intermetallics to be introduced into analytical expressions for semisolid permeability. In this work, the calculated values of permeability have been fitted to an empirical relationship based on the Blake–Kozeny equation following the procedure of Poirier \cite{[6]}. An additional term, \( 1 - \beta f_I \), where \( \beta \) represents the impact factor of the intermetallics on flow blockage, has been added to account for the effect of \( f_I \) as shown below:

\[
K_p = (1 - \beta f_I)K_{p}^{orig}
\]  
\[
K_n = (1 - \beta f_I)K_{n}^{orig}
\]  
\[
K_{p}^{orig} = C_1 f_I^2 \lambda_1^2 \frac{1}{1 - f_L}
\]  
\[
K_{n}^{orig} = C_2 \left( \frac{\lambda_1}{\lambda_2} \right)^p f_I^2 \frac{\lambda_1^2 \lambda_2^2}{1 - f_L}
\]  

where \( K_p \) and \( K_n \) are the permeabilities in the directions parallel and normal to the primary dendrites, \( K_{p}^{orig} \) and \( K_{n}^{orig} \) are the original expressions of Poirier \cite{[6]}, \( \lambda_1 \) and \( \lambda_2 \) are the primary and secondary dendrite arm spacings, \( f_L \) is the liquid fraction, and \( C_1, C_2, p, n \) are fitting constants calculated from the regression analysis. For the case without intermetallics, \( f_I = 0 \), reducing Eqs. (4) and (5) to the general anisotropic Blake–Kozeny permeability expressions. The value of \( \beta \), along with the fitting constants \( C_1, C_2, p, n \), is summarized in Table 1. These values were obtained via a non-linear least-squares regression analysis of the simulated permeability results taking into account only the values that did not contain eutectic Al–Si. The constants obtained by Poirier are also provided in Table 1 for comparison purposes. Note that while the values of \( f_I \) were obtained directly from the tomographic datasets, data from thermodynamic calculations could be utilized for implementation of Eqs. (4) and (5) into casting models for predicting defects.
The comparison between the simulated permeability results and the analytical expressions based on Eqs. (4), (5) and Table 1 is shown in Fig. 9. It can be seen that the fits in both the parallel (Fig. 9a) and the normal (Fig. 9b) directions are excellent at \( f_L > 0.4 \), i.e. in the regime of primary phase solidification. Thus, the permeability of columnar dendritic microstructures with intermetallics is shown to be predictable by adding an impact factor to the Blake–Kozeny expressions. The value of \( b \) is observed to be larger when comparing the case of flow in the parallel direction \((b = 15)\) to the case of flow in the normal direction \((b = 10)\). As shown earlier in Fig. 8, greater impact on the flow fields can be found when plate-like intermetallic particles grow normal to the flow direction. It would then be expected that blocking particles with less complexity (e.g. globular shape) would have smaller \( b \) factors.

For values of \( f_L < 0.4 \), the new analytical expression for permeability deviates from the simulation results. At this point, the eutectic reaction has commenced, and this will strongly affect channel blockage as the eutectic Al–Si may form alongside primary Al-rich dendrites [46] and/or may cause complete closure of small side-branch channels (e.g. Fig. 4e and g) and/or may cause the formation of preferential flow paths. The deviation may also be due to the relative effects of intermetallic formation and eutectic formation on channel tortuosity, which could be significantly increased in these cases.

The analytical permeability expressions were originally derived based on dendritic microstructures, and while the presence of eutectic will increase the solid fraction, it will not necessarily increase tortuosity (i.e. Eqs. (6) and (7) may not apply in the case of columnar dendritic structures with a large eutectic fraction). This corresponds to the finding of Khajeh and Maijer [27] that, for large values of \( \theta \), the total permeability is controlled only by the permeability of the dendritic network. In contrast, the channel-blocking mechanism resulting from intermetallic formation will definitely increase tortuosity in both parallel and normal flows. Further study of the impact of eutectic formation on fluid flow is thus required in order to include this aspect of the problem into an analytical expression for permeability containing both intermetallics and a significant eutectic constituent.

### 3.4. Comparison with experimental data and analytical approximations

Murakami et al. [17,18] using borneol–paraffin and Poirier [6] using Pb–Sn have measured the permeability for different values of \( \lambda_1, \lambda_2 \) and \( f_L \) for flow both parallel to and normal to the primary dendrite direction. These experimental results, together with the analytic Hagen–Poiseuille (Santos and Melo [34]) and Blake–Kozeny (both this work and Poirier [6]) expressions, are compared in Fig. 10 against the permeability values extracted from the present flow simulations. In order to put all of the results on the same figure, a dimensionless form of permeability is adopted. This figure shows that the flow simulations match reasonably well with the experimental data for both paral-
lel and normal flow regimes. Regarding the analytical expressions, while the expressions also match reasonably well with the flow simulations without intermetallic particles, the best fit is obtained with the anisotropic Blake–Kozeny model using the constants derived in this work because this expression illustrates good agreement for both parallel and normal flow regimes. Although the differences between the permeability values using Poirier’s constants and the constants derived in this work are small, especially in the case of normal flow, a significantly better fit is found at higher liquid fractions. This is because Poirier only had experimental data up to \( f_L < 0.6 \) to perform the regression analysis, whereas tomographic datasets with \( 0.6 < f_L < 0.9 \) were available in this work.

4. Conclusions

A novel method for coupling in situ 4-D synchrotron X-ray tomographic microscopy with numerical simulations was developed to quantify the permeability of solidifying microstructures. This new technique captured, both in situ and in 4-D, multiple solid fractions \((0.1 < f_S < 0.85)\) of a single Al–Si–Cu specimen during solidification. With such information, the development of multiple phases (primary, intermetallics and eutectic) and accurate measurements of the permeability and fluid flow of the evolving sample were possible for the first time. The results qualitatively and quantitatively demonstrate the blocking effect of intermetallics on fluid flow and the ensuing loss in permeability, particularly in the direction parallel to the primary dendrites. The results also show that the \( \beta \)-Al\(_5\)FeSi intermetallic precipitates reduce the permeability of the semisolid metal, potentially explaining the increased susceptibility of such materials for casting defects such as hot tearing and microporosity.

This first direct quantification of permeability was then captured in an analytical expression based on the anisotropic Blake–Kozeny relation which can easily incorporate macroscopic flow simulations of defect formations during solidification processes. This new expression includes an additional term, \( 1 - \beta f_L \), to characterize the effect of the intermetallic phase. A good fit for parallel and normal flows to both the simulation results and experimental data from the literature is found when using this new expression. This factor takes into account the dependency of morphology and orientation of blocking particles. Thus, it can be adjusted and used for predicting the influence of blocking particles in other systems, e.g. in carbides in Ni-base superalloys [47], in non-metallic inclusions in steel, and in fibre assembly [48].

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Appendix A. Supplementary material

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References

[1] Lee PD, Chirazi A, See D. J Light Metals 2001;1:15–30.
[2] Beckermann C. Macrosegregation. In: Buschow KHJ et al., editors. Encyclopedia of materials: science and technology. New York: Elsevier Science; 2001. p. 4733.
