Evolution of the Specific Solid-Liquid Interface Area in Directional Solidification

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EXTENDED ABSTRACT

A key aspect in predicting the microstructure in castings is the detailed knowledge of how geometrical features evolve over time during solidification. Often, local features, such as the secondary dendrite arm spacing, are used for the geometrical characterization of the microstructure. However, they represent incomplete descriptions of the solid structure and their measurement can become difficult during the late stages of solidification, when the structure undergoes fundamental transformations. Alternatively, integral measures, such as the specific area of the solid-liquid interface, can be introduced that more generally characterize the overall morphology. One definition of the specific interface area is the amount of interface area A per volume of the enclosed solid phase V_s,

$$S_s = A / V_s \tag{1}$$

which may also be considered a characteristic inverse length scale of the microstructure. Another definition is the ratio of the interface area A to the sample volume V containing both solid and liquid phases

$$S_v = A / V = f_s S_s \tag{2}$$

where $f_s = V_s / V$ is the solid volume fraction. S_v is also referred to as the interfacial area density. Both quantities are key ingredients in volume-averaged (macroscopic) models of alloy solidification and are needed, for example, in modeling of microsegregation (back-diffusion) or melt flow through the mush.

Under isothermal conditions, the evolution of the inverse specific interface area is usually described by the following relation for surface energy driven coarsening

$$S_s^{-1} = (S_{s0}^{-n} + K t)^{1/n} \tag{3}$$

where t, n, S_{s0}, and K are time, coarsening exponent, specific interface area at $t = 0$, and coarsening rate constant, respectively. For volume diffusion-limited coarsening at a constant solid fraction an exponent of $n = 3$ has been firmly established by both experiments and theory. A model has been developed for the case of concurrent growth and coarsening, in which an exponent of $n = 3$ was obtained even in the presence of solidification [1].

In contrast to pure coarsening, solidification implies that the solid fraction increases over time. Eventually, the specific interface area becomes strongly affected by coalescence and the theory of Ref. [1] is no longer valid. For processes that involve only growth, but no surface energy driven coarsening, the interfacial area density S_v is often correlated to f_s by

$$S_v = C f_s^p (1-f_s)^q \tag{4}$$

where C, p, and q are constants. According to Eq. (4), S_v experiences a steep increase during growth, goes through a maximum, and then decreases due to impingement and coalescence of interfaces. Different values for the exponents p and q have been suggested in the literature. Speich and Fisher [2] found that data from a broad range of recrystallization experiments could be described by $p = q = 1$.

In summary, Eqs. (3) and (4) are useful relations for the specific interface area, but are limited to seemingly opposing cases. While Eq. (3) was developed for the isothermal case, where the interface area evolves over time due to coarsening, Eq. (4) is meant to describe situations where the solid fraction varies with time due to growth but the interface area does not change when the solid fraction is held constant. Hence, the question remains how these two models can be combined for situations that involve both growth and coarsening, such as dendritic solidification of alloys.

In this work we have studied the kinetics of the solid-liquid interface of a columnar dendrite of an Al-6wt.%Cu alloy by performing a 3D phase-field simulation. The computational setup corresponds to a Bridgman experiment where solidification occurs in a fixed temperature gradient and at a constant cooling rate. All details of the model and the calculations can be found in Ref. [3]. Computed solid-liquid interface shapes at various times are shown in Fig. 1. Simultaneous growth and coarsening of the dendritic structure can be clearly observed. The interface area and solid volume are integrated over a representative volume element, and their time evolution is plotted in Fig. 2.

Figure 3 shows the computed solid fraction as a function of the scaled temperature. As expected, the solid fraction is equal to zero until the scaled temperature reaches the dendrite tip temperature; afterwards, the solid fraction increases sharply with increasing scaled temperature. This solid fraction variation can be compared to the classical lever rule and Scheil equation predictions, which assume that the dendrite tips are located at the equilibrium liquidus isotherm. Other than for the dendrite tip undercooling effect, the lever rule and the Scheil equation closely bound the solid fraction variation from the phase-field simulation.

The computed interfacial area density S_v is plotted in Fig. 4 against the solid fraction. The figure shows that S_v varies in accordance with Eq. (4). By fitting the present data to Eq. (4), it is found that the exponents are equal to $p = 0.99$ and $q = 0.92$, which is close to $p = q = 1$ suggested in Ref. [2]. Clearly, exponents of $p = q = 1/2$ do not fit the simulation results. It is interesting to note that Eq. (4), which was originally developed for pure growth, fits the present data for the interfacial area density, even though dendritic solidification is characterized by concurrent growth and coarsening.

The calculated temporal evolution of the inverse specific interface area S_s^{-1} is shown in Fig. 5. The data for S_s are fit to Eq. (3) in order to determine the exponent n. Figure 5 shows that an exponent of $n = 3$ is obtained for short times ($t < 2$ s), while an exponent of $n = 0.86$ fits the simulation data at longer times ($t > 2$ s). The exponent of $n = 3$ during the initial growth stage is in agreement with the finding in Ref. [1] for concurrent growth and coarsening of spheres in the limit of low solid fractions. The solid fraction at $t = 2$ s is equal to 0.5, indicating that coalescence of solid can be neglected up to this fraction. The exponent of $n = 0.86$ observed at higher solid fractions ($t > 2$ s) can be explained by inserting the Scheil equation into Eq. (4), assuming that $p = q = 1$ and using $S_s^{-1} = f_s / S_v$. This analysis [3] shows that $n = 1 - k$, where a partition coefficient of $k = 0.14$ is used in the present simulation.

Considerable additional research is necessary to obtain a generally valid relation for the evolution of the specific interface area in alloy solidification. Simulations are underway that investigate the effect of different cooling rates and other alloy characteristics on the interface evolution.

Key Words: Interfacial area density, directional solidification, coarsening, phase-field simulations

REFERENCES

1. L. Ratke, C. Beckermann, Acta Mater **49**, 2001, 4041–4054.

2. G. Speich, R. Fisher R, Recrystallization, Grain Growth and Textures (ASM, Materials Park, OH), 1966.

3. H. Neumann-Heyme, K. Eckert, C. Beckermann, IOP Conf. Series: Materials Science and Engineering **84**, 2015, 012072.

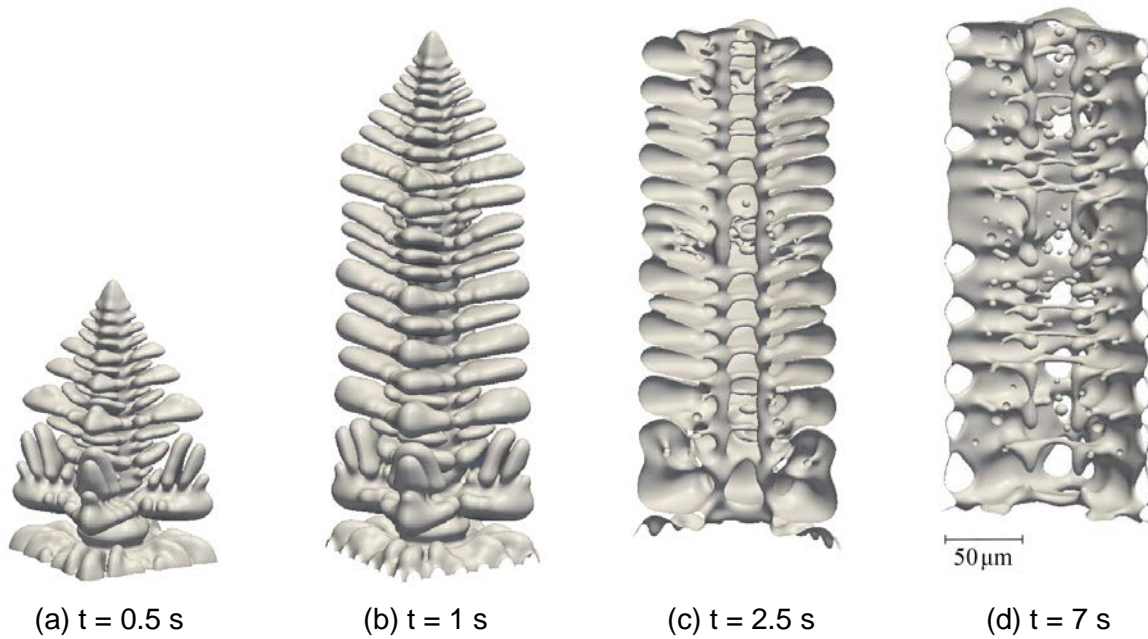| (a) t = 0.5 s | (b) t = 1 s | (c) t = 2.5 s | (d) t = 7 s |

Fig. 1: Evolution of the dendrite geometry: (a)-(b) full view of the growing dendrite, (c)-(d) cutaway view of half of the dendrite during the coarsening stage.
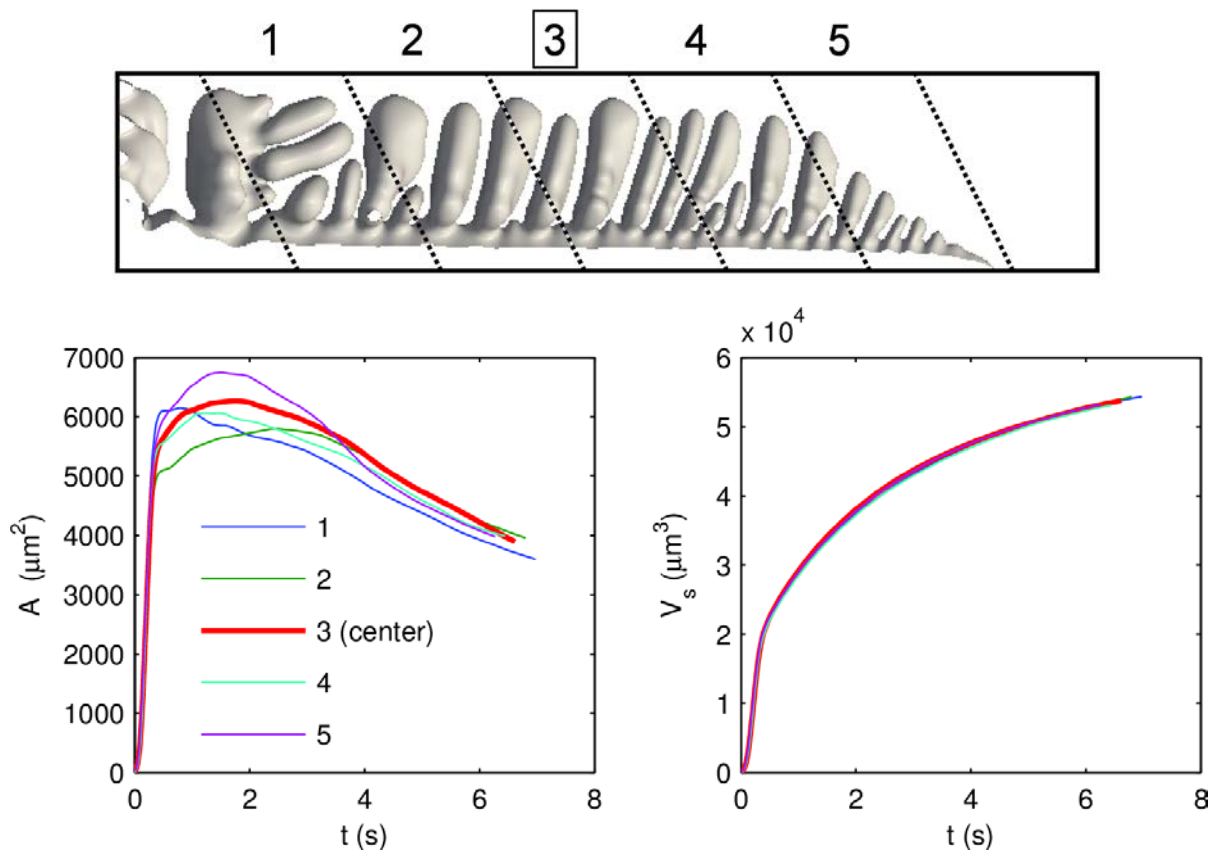

Figure 2: Averaging volumes at different positions along the growth direction (top), and evolution of the averaged interface area and solid volume (bottom).
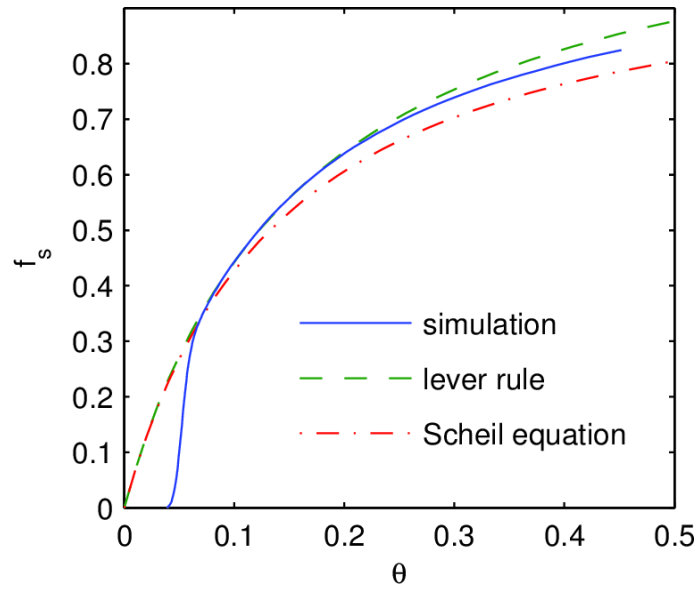
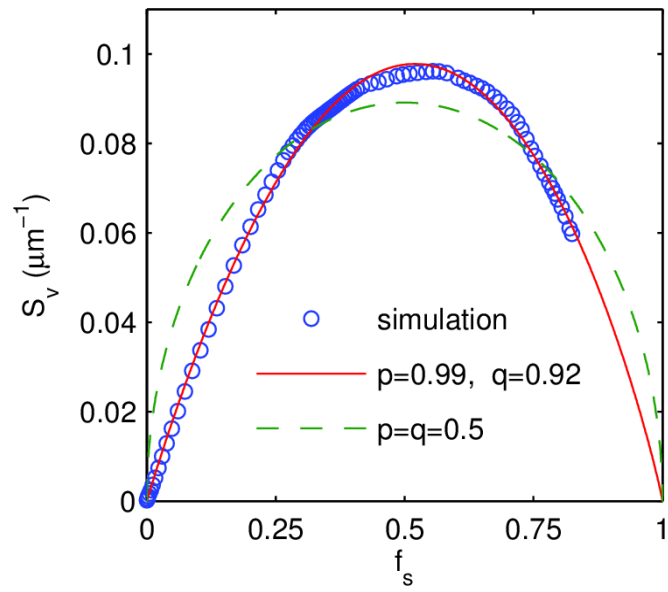Figure 3: Solid fraction as a function of the scaled temperature.

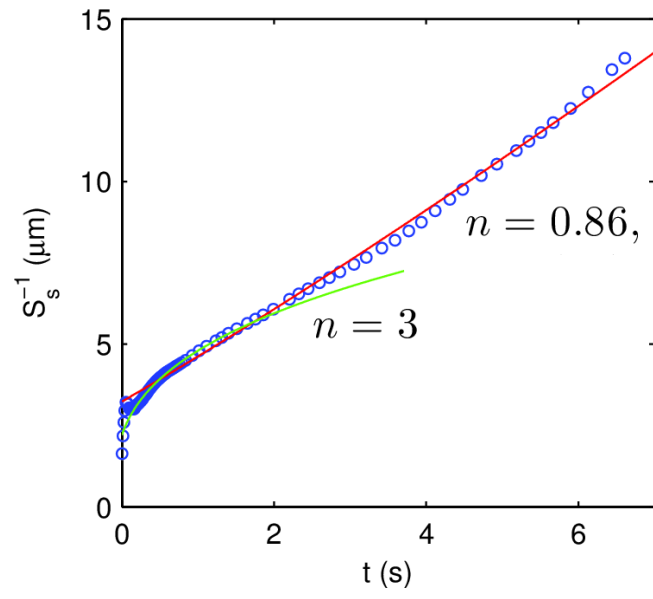Figure 4: Variation of the interfacial area density with solid fraction.
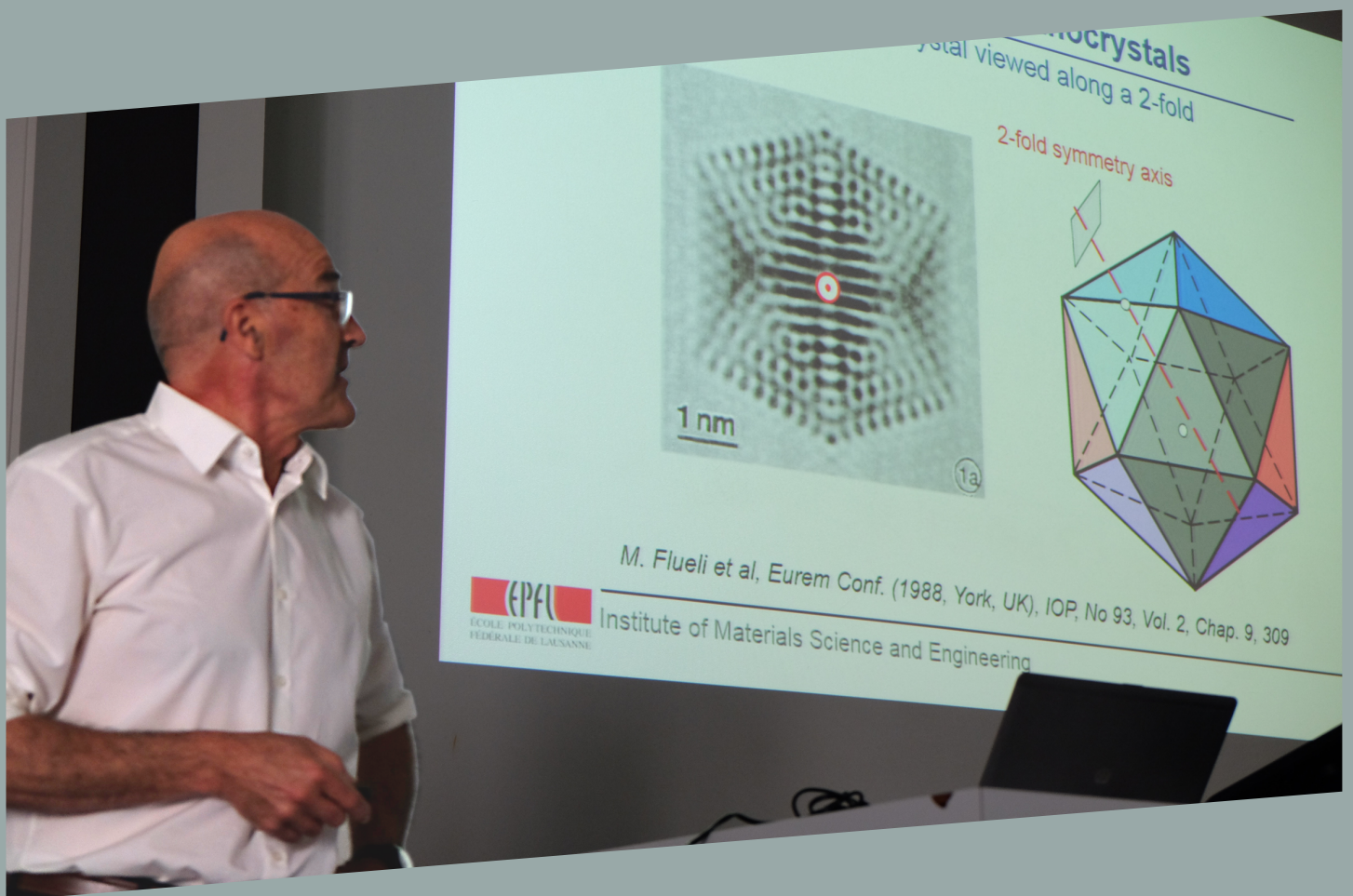
Figure 5: Evolution of the characteristic length scale.

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