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Microstructural evolution during temperature gradient zone melting: Cellular automaton simulation and experiment

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ABSTRACT

The microstructural evolution in mushy zones of alloys due to temperature gradient zone melting (TGZM) is studied by simulations using a two-dimensional quantitative cellular automaton (CA) model and *in situ* observations of directional solidification with a transparent organic SCN-ACE alloy. The present model is an extension of a previous CA model by involving the mechanisms of both solidification and melting. The present CA model is adopted to simulate the temporal evolution of the position and velocity of a liquid pool migrating in the solid matrix of a SCN–0.3 wt% ACE alloy under conditions that the pulling velocity is either lower or higher than the critical pulling velocity. The CA simulated position and velocity curves agree well with analytical solutions. Simulations are also performed for the microstructural evolution of columnar dendrites in a SCN–2.0 wt% ACE alloy held in a stationary temperature gradient using the present CA model and a previous CA model that does not include the melting mechanism under otherwise identical conditions for comparison. The results show how melting is essential to dendrite arm migration in a temperature gradient. The time-averaged velocities of arm migration obtained from the present CA simulations increase with increasing temperature gradient and with decreasing the length between the initial arm position and the liquidus. This agrees reasonably well with experimental measurements and analytical predictions. The mechanisms of dendrite arm migration are investigated in detail by comparing the local equilibrium and actual liquid compositions at solid/liquid interfaces. The simulations render visualizing the complex interactions among local temperature, solute distribution/diffusion, and solidification/melting during the TGZM process.

1. Introduction

Microstructural evolution of alloys during directional solidification is of persistent interest in both scientific research and industrial practice. Owing to the presence of temperature gradients during directional solidification, local remelting and solidification in the mushy zone occurs, involving either liquid inclusions migrating in the solid phase, or a sequence of dendrite arms moving in the liquid phase, in both cases towards higher temperatures. This remelting/solidification phenomenon is termed temperature gradient zone melting (TGZM).

The mechanism of TGZM for the migration of secondary dendrite arms and interdendritic liquid pools during directional solidification with an arbitrary pulling velocity, V_p, is schematically illustrated in Fig. 1. In the presence of a temperature gradient, the adjustment of the temperature dependent local equilibrium compositions at neighboring solid/liquid (S/L) interfaces establishes a composition gradient across the interdendritic liquid pool between the two adjacent arms. As seen from Fig. 1, the solute concentration at the cold side of the interdendritic liquid pool is higher than that of the hot side. The composition gradient drives solute atoms to diffuse from the colder side with higher composition to the hotter side with lower composition. Local solute enrichment at the hotter S/L interface will lead to its remelting, while solute depletion at the colder S/L interface will cause solidification. As a result, this remelting/solidification process results in the migration of the interdendritic liquid pools towards the high temperature direction [1]. Apparently, the TGZM effect of interdendritic pools is equivalent to the process that the front edges of secondary arms freeze, while the back edges melt, leading to the migration of the secondary arms towards the high temperature direction. The TGZM effect is known to alter the solidifying microstructure and microsegregation remarkably [1]. It has also been recognized that

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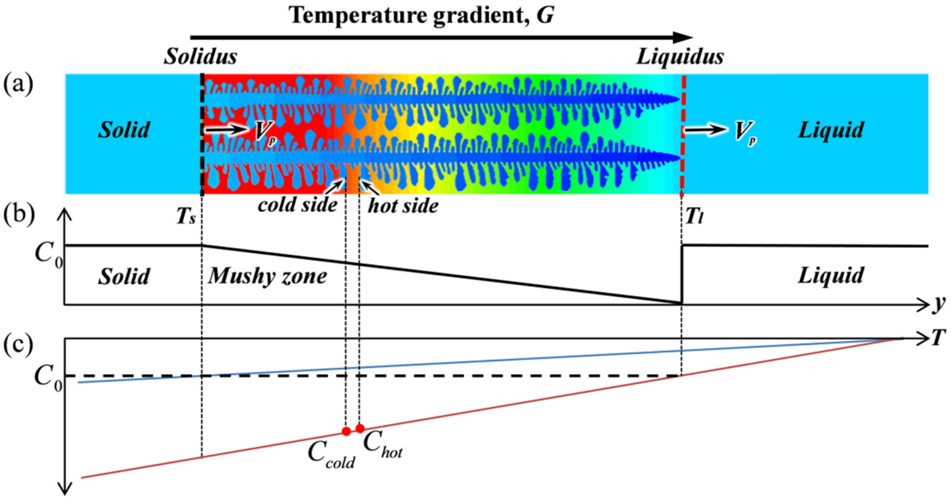

Fig. 1. Schematic illustration of secondary dendrite arms and interdendritic liquid pools migrating in a mushy zone due to TGZM with an arbitrary pulling velocity, V_p: (a) schematic sample; (b) concentration distribution in a sample with complete mixing in the liquid (e.g. due to stirring) and (c) correlation with the liquidus and solidus lines in the phase diagram.

the TGZM effect has a wide variety of applications in both scientific research and industrial practice, such as etching silicon wafers [2], fabricating thermoelectric materials [3], growing bulk silicon crystals [4], measuring diffusivities [5], and alloying and purifying materials on a small scale [6]. Imposing a temperature gradient on solid/liquid microstructures of alloys can also result in morphological instability at migrating S/L interfaces during TGZM, which can be utilized in temperature gradient transient liquid phase diffusion bonding [7].

Assuming local equilibrium at the S/L interface, and neglecting thermal transport and solute diffusion in the solid, analytical models have been developed to describe the migration behavior of liquid droplets/pools due to TGZM [8,9]. Recently, on the basis of the Allen and Hunt (AH) model [8], some of the present authors proposed an analytical model that allows an explicit solution of the time-dependent position and migration velocity of a liquid droplet/dendrite arm in a mushy zone for both static and dynamic conditions [9]. Criteria are derived for critical pulling velocity and critical droplet position, both yielding constant relative positions of the migrating droplet with respect to the moving liquidus and solidus.

Numerous experimental studies have been carried out to investigate the phenomena and mechanisms of TGZM by *in situ* observations using synchrotron-based X-ray radiography [10,11] or by *post-mortem* analyses of samples quenched from mushy zones [12–15]. Li et al. [10] and Nguyen-Thi et al. [11] observed the phenomena of secondary dendrite arm migration due to TGZM using *in situ* synchrotron X-ray radiography. They measured the remelting and solidification rates of the back and front edges of the secondary dendrite arms and the average migration velocities; the measured data were compared to the analytical predictions obtained from the AH model. Detailed studies on microstructural evolution and concentration distribution changes in mushy zones in steep temperature gradients were performed by the group of one of the present authors through *post-mortem* analyses of quenched samples [12,13]. In the studies, the solidified samples with equiaxed grains were partially melted in a temperature gradient and held for different times. It is demonstrated that as compared to the initial microstructure of equiaxed grains (before applying the temperature gradient), changes of grain size and grain aspect ratio as well as concentration distributions in the mushy zone occur, which is caused by simultaneous remelting/solidification. The TGZM mechanism was also extended to peritectic solidification to explain the experimental observations. Liu et al. [14] and Peng et al. [15] performed experimental studies on the microstructural evolution during directional solidification or thermal stabilization of Al-Ni and Sn-Ni peritectic alloys. The behavior of the secondary arm migration for the primary and peritectic phases, as well as the morphological evolution of the interface between different mushy zones were observed. These experimental studies have provided a lot of important information about the mechanisms that occurred during TGZM. Nevertheless, at present accurate measurements of the variations of local temperature and local composition at the S/L interfaces are still challenging, even though some image processing techniques have been developed for liquid composition measurement through analyzing the brightness distributions (grey levels) on the experimental images taken by *in situ* synchrotron X-ray radiography [16]. Thus, quantitative analysis of TGZM phenomena is not yet exhaustive in experimental studies.

In the recent decades, in parallel to the development of experimental techniques and analytical models, numerical modeling has become a powerful and indispensable tool in studies of microstructural evolution for various types of phase transformations. Phase-field (PF) models inherently comprise the mechanisms of both solidification and melting and thus are capable of appropriately describing the migration of liquid droplets and secondary dendrite arms due to TGZM. PF simulations have recently been performed to investigate liquid droplet migration and mushy zone solidification in steep temperature gradients [9,17]. The PF method is, however, known to demand a massive computational resources. The diffuse interface feature of the PF approach poses the challenge to quantitatively describe microstructural evolution during TGZM on experimentally relevant length and time scales. In addition, solidification and melting mechanisms are usually inseparable in PF simulations, and thus the role of melting in TGZM cannot be separated from that of solidification.

Cellular automaton (CA) models can reproduce most of the microstructural features observed experimentally during solidification with an acceptable computational efficiency. It has thus drawn considerable interest in academia and led to the development of various models based on the CA technique to simulate a wide variety of solidification microstructures [18–27]. However, all existing microscale CA models only consider solidification, but