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液态金属快速凝固过程的 PFM 模型^①

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[摘 要] 近年来发展起来的 PFM 模型根据“溶质捕捉”和“溶质牵引”两种机制对金属及合金快速凝固过程进行数学描述及模拟, 较好地反映了金属凝固的物理本质。介绍了 PFM 模型发展过程、研究现状及理论模型, 并对发展前景进行了展望。

[关键词] 相区场模型; 溶质捕捉; 合金凝固; 枝晶生长

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凝固过程的物理本质及数学模拟一直是凝聚态物理和材料科学领域的研究重点之一。在平衡凝固和近平衡凝固条件下, 经典热力学和扩散动力学可以较好地解决问题^[1, 2]。但在快速凝固过程中, 金属液的冷却速度很快, 不仅固相和液相内部的溶质来不及充分扩散, 凝固界面上的溶质迁移也偏离平衡。合理地反映界面迁移过程中的物质、能量传输过程是数学模型必须考虑的问题。

对凝固前沿固液界面的处理大体可以归为两类^[3~9]: 不连续界面模型 (Sharp interface model)^[10] 和扩散界面模型 (Diffusive interface model)。不连续界面模型针对凝固界面两侧的微小区域进行研究, 认为该区域内由独立的固相和液相组成, 两者之间的界面为简单的几何界面。该类模型的界面生长速度和液相浓度之间的关系, 较好地反映了溶质传输过程。但该类模型存在两个明显的缺点: 1) 忽略了界面前沿实际存在的固液共存区, 使结果和实际情况不能相符合; 2) 将界面温度和液相浓度用不同的控制方程来表示, 而不是有机的结合起来。

扩散界面模型认为液相和固相之间存在具有一定厚度的界面, 该界面区域在浓度、密度和温度等性质上和毗邻的液相和固相区都有着一定的差别, 凝固组织前沿的生长和该界面向液相内部的推移是同时进行的。同时, 固相和液相之间的传输过程也必须经过这个区域。通过 Ginzburg-Landau 相变理论^[11, 12] 发展起来的相区场模型 (Phase-field model) 是其中重要的代表^[13]。

根据对界面微区自由能密度表达方式的不同,

相区场模型又可以分为 3 类: 由 Wheeler, Bottinger 和 Mcfadden 建立的 WBM 模型^[13~16], Steinbach 等人建立的适用于稀溶液的模型^[17, 18], 以及 Losert 等人建立的模型^[19, 20]。其中以 WBM 模型的应用最为广泛^[13, 14, 21, 22], 另外两种则由于有过多的假设条件而缺乏普遍性^[23]。本文将 WBM 模型为例说明相区场模型中的核心问题。

1 数学模型

1.1 PFM 模型

基于 Helmholtz 自由能公式, Caginalp 和 Xie 给出的 WBM^[13, 24~28] 模型具有如下形式:

$$F = \int_{\Omega} [f(\phi, c, T) + \frac{\epsilon}{2} |\nabla \phi|^2] dv \quad (1)$$

式中 Ω 为研究对象所处的微区, 梯度项反映了界面迁移过程中能量的变化, 常数 ϵ 为梯度能量系数。二元合金自由能密度函数 $f(\phi, c, T)$ 由相区场函数 ϕ 、浓度场函数 c 以及温度场函数 T 共同决定:

$$f(\phi, c, T) = p(\phi)f_s(c, T) + [1 - p(\phi)]f_L(c, T) + \frac{W(c)}{4}g(\phi) \quad (2)$$

相区场函数 $\phi(x, t)$ 用以表征凝固界面中每一点相的特征^[21]。其中的双阱函数 $g(\phi) = \phi^2(1 - \phi)^2$, 在 $\phi = 0$ 和 $\phi = 1$ 处有极值, 分别对应于纯液相和固相。 $p(\phi)$ 满足 $p(0) = 0$ 和 $p(1) = 1$, 用来表

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明凝固界面的微区中由于温度和浓度不同而引起的差异, 具有不同的多项式表达方式^[29, 30]。\$W(c)\$是和表面能以及界面厚度有关的能垒大小。

为使 Helmholtz 自由能函数随时间变化单调递减, 可令 \$\partial \phi / \partial t = -M_1 \partial F / \partial \phi\$, \$\partial c / \partial t = \nabla \cdot M_2 \cdot [c(1-c)] \nabla (\partial f / \partial c)\$。式中常数 \$M_1\$ 为界面动力学系数, 迁移系数 \$M_2\$ 为和溶质扩散系数有关的常数。

通过上述方程, 可以写出如下的界面生长速度 \$v\$ 的控制方程表达式, 这里的 \$z\$ 满足 \$-l/2 < z < l/2\$。

$$\frac{-v}{M_1} \frac{d\phi}{dz} = \epsilon^2 \frac{d^2\phi}{dz^2} - \frac{\partial f(\phi, c, T)}{\partial \phi} \quad (3)$$

$$\frac{-v}{V_m} \frac{dc}{dz} = \frac{dI}{dz} \quad (4)$$

1.1.1 溶质分配驱动力

对式(4)进行积分可得 \$v(c - c_s) V_m = J\$, 扩散通量 \$J = -c(1-c) M_2 V_m^{-1} d\mu/dz\$, 界面层化学位具有如下的形式。积分后便可以得到溶质分配驱动力的表达式:

$$\frac{d\mu}{dz} = \frac{-v_m J}{c(1-c) M_2} = \frac{-v(c - c_s)}{c(1-c) M_2} \quad (5)$$

$$\mu(c_L) - \mu(c_s) = -V \int_{-l/2}^{l/2} \frac{c - c_s}{c(1-c) M_2} dz \quad (6)$$

1.1.2 相变驱动力

对式(3)两边同乘 \$d\phi/dz\$ 并进行积分, 得到

$$\frac{-v}{M_1} \int_{-l/2}^{l/2} \phi_z^2 dz = \frac{\epsilon}{2} [\phi_z^2]_{-l/2}^{l/2} - \int_{-l/2}^{l/2} f_{\phi} \phi_z dz \quad (7)$$

分步积分得到

$$\begin{aligned} \int_{-l/2}^{l/2} f_{\phi} \phi_z dz &= [f(\phi, c)]_{-l/2}^{l/2} - \int_{-l/2}^{l/2} f_c \phi_z dz \\ &= [f(\phi, c)]_{-l/2}^{l/2} - \int_{-l/2}^{l/2} f_c (c - c_s) dz \\ &= [f - (c - c_s)f_c]_{-l/2}^{l/2} + \int_{-l/2}^{l/2} (c - c_s) \mu_z dz \end{aligned}$$

由 \$\mu = \partial f / \partial c\$ 结合(6)式上式可以重写为

$$\begin{aligned} \int_{-l/2}^{l/2} f_{\phi} \phi_z dz &= [f - (c - c_s)f_c]_{-l/2}^{l/2} + \\ &\quad \frac{V_m}{v} \int_{-l/2}^{l/2} J \mu_z dz \quad (8) \end{aligned}$$

令 \$a = \int_{-l/2}^{l/2} \phi_z^2 dz\$, 同时 \$\frac{\epsilon}{2} [\phi_z^2]_{-l/2}^{l/2} \approx 0\$, 则可以得到相变时自由能变化的表达式, 亦及相变的驱动力:

$$\Delta F_S = f(c_s) - [f(c_L) + (c_s - c_L)f_c(c_L)] \quad (9)$$

1.2 溶质捕捉模型^[21, 31~33]

二元合金的溶质模型是通过对生长前沿浓度变化的分析而建立起来的。为此, 需要对扩散和凝固所需要的能量进行分析。设固相和液相内的自由能密度分别为 \$f_s(c_s, T)\$ 和 \$f_L(c_L, T)\$, 其中的 \$T\$ 代表温度, \$c_s\$ 和 \$c_L\$ 分别代表固相和液相中溶质的摩尔分数。在液相和固相中 A 组元和 B 组元的化学位可以表示如下:

$$\mu_A^L = f_L(c_L, T) - c_L \frac{\partial f_L}{\partial c_L}(c_L, T) \quad (10)$$

$$\mu_B^L = f_L(c_L, T) + (1 - c_L) \frac{\partial f_L}{\partial c_L}(c_L, T) \quad (11)$$

$$\mu_A^S = f_s(c_s, T) - c_s \frac{\partial f_s}{\partial c_s}(c_s, T) \quad (12)$$

$$\mu_B^S = f_s(c_s, T) - (1 - c_s) \frac{\partial f_s}{\partial c_s}(c_s, T) \quad (13)$$

热力学平衡时界面处的化学位相等, \$\mu_A^S = \mu_A^L\$ 且 \$\mu_B^S = \mu_B^L\$。可以通过互扩散化学势来表示平衡条件: \$\mu^L = \mu_B^L - \mu_A^L = \partial f_L / \partial c_L\$, \$\mu^S = \mu_B^S - \mu_A^S = \partial f_s / \partial c_s\$。凝固时的自由能变化可以表示为

$$\begin{aligned} \Delta F_S &= f_s(c_s, T) - [f_L(c_L, T) + \\ &\quad (c_s - c_L) \frac{\partial f_L}{\partial c_L}(c_L, T)] \quad (14) \end{aligned}$$

该式表明了液相向固相传质的能量变化情况。平衡时, \$\mu^L - \mu^S = 0\$, \$\Delta F_S = 0\$。在稀溶液条件下, 通过 \$\mu^L - \mu^S = 0\$ 可以推导出 \$c_s = k_E c_L\$, 并得到平衡分配系数 \$k_E\$ 的定义式。通过 \$\Delta F_S = 0\$ 推导出液相温度的表达式 \$T = T_M + m_L c_L\$。在非平衡的条件下, \$\mu^L - \mu^S \neq 0\$, \$\Delta F_S \neq 0\$, 两式分别可以理解为溶质分配、相变的驱动力。

1.2.1 分配系数

假设凝固界面厚度的尺度为原子尺寸, 同时为简化表达, 添加热力学因子 \$c_L(1 - c_L)\$, 则扩散通量 \$J\$ 可以表示为如下形式:

$$J = \frac{-M_0}{V_m} c_L(1 - c_L) \frac{\mu^L - \mu^S}{l} = \frac{v}{V_m} (c_L - c_s) \quad (15)$$

式中 \$V_m\$ 为摩尔体积, \$M_0\$ 为与浓度相关的迁移系数, \$v\$ 为界面生长速率。则界面生长速度和溶质分配驱动力有如下关系:

$$\mu^L - \mu^S = -v \frac{(c_L - c_s)l}{M_0 c_L(1 - c_L)} \quad (16)$$

在稀溶液中, 分配系数可以表达为 \$\ln(k/k_E) = v(1 - k)/v_D\$。

1.2.2 界面温度

通过凝固自由能变化 \$\Delta F_S\$ 可以获得界面温度

的表达式。自由能变化和界面迁移速率具有如下的关系^[21],

$$v = v_c [1 - \exp(\frac{V_m \Delta F_S}{RT})] \approx -v_c \frac{V_m \Delta F_S}{RT} \quad (17)$$

该模型假设凝固过程中不存在能量损失, 其中 v_c 为可能的最大生长速度。在溶质捕获模型中, 由于是开放系统, 存在着能量的耗散 ΔF_D , 则用于凝固过程的自由能为 $\Delta F_C = \Delta F_S - \Delta F_D$, 式(17)相应地变为

$$v = v_c [1 - \exp(\frac{V_m \Delta F_C}{RT})] \approx -v_c \frac{V_m (\Delta F_S - \Delta F_D)}{RT} \quad (18)$$

Aziz 和 Kaplan 专门提出耗散自由能的表达式, 并由此推出稀溶液条件下界面温度的表达式^[34, 35]。其中的参数 α 可以为溶质从液相向固相浓度转变的度量, $\alpha = 1$ 时表示存在溶质牵引效应, $\alpha = 0$ 则不存在溶质牵引。 m_L 为平衡液相线斜率, 动力学系数 $u = v_c L / (RT_M^2)$ 。

$$\Delta F_D = (c_L - c_S) \{ [\mu_A^S(c_S, T) - \mu_A^L(c_L, T)] - [\mu_B^S(c_S, T) - \mu_B^L(c_L, T)] \} \quad (19)$$

$$T = T_M + \frac{m_L c_\infty}{k} \cdot \left\{ \frac{1 - k + [k + (1 - k) \alpha / \ln(k/k_E)]}{1 - k_E} \right\} - \frac{u}{u} \quad (20)$$

2 应用及展望

针对纯金属材料的平衡及非平衡过程应用 PFM 模型已经有多年的历史, 随着计算机计算能力的不断提高, 已经能够模拟诸如树枝晶生长等复杂的动态过程^[36~42]。

针对二元合金凝固过程的研究是 WBM1 模型建立以后的事情^[13, 21, 37], Xie 和 Caginalp 将 PFM 模型应用于二元合金的非平衡凝固过程。此后, 针对二元合金非平衡凝固组织生长的研究越来越多^[14, 37~46], 并和实验结果取得了较好的一致^[21, 47]。

随着研究的不断深入, 针对更深层次现象的研究也越来越多。Beckermann 等人研究了凝固过程的对流条件影响以及微观偏聚现象^[48~51], Conti 等人针对 PFM 模型中的热现象进行了研究^[19, 52]。合理借鉴物理学和其它领域的一些理论, 可以帮助完善 PFM 模型。如 Cahn-Hilliard 方程对形核(调幅分解前期)的处理非常成功^[53~59], 而 PFM 模型对形

核的描述并不充分。PFM 模型还不能解决由于晶格类型不同而导致的枝晶生长各向异性问题, 而固态相变弹性场理论对各向异性弹性场的处理则较为成熟^[60, 61]。此外, 枝晶生长作为一种典型的自组织现象, 自组织生长的 DLA 动力学模型^[62~65], 也有一定的参考价值。

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Phase-field model of rapid solidification process of liquid alloy

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[Abstract] In recent years, phase-field model(PFM) of alloy solidification based on solute trapping and solute drag was introduced as a powerful tool to simulate the process of solidification. An overview introduction about its development was given and an emphasis on its theoretical frame was put.

[Key words] phase-field model; solute trapping; alloy solidification; dendritic growth

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