Research Article

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Cellular Automaton Modeling of Phase Transformation of U-Nb Alloys during Solidification and Consequent Cooling Process

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Abstract: The microstructure evolution of U-Nb alloys during solidification and consequent cooling process was simulated using a cellular automaton (CA) model. By using this model, γ phase precipitation and monotectoid decomposition were simulated, and dendrite morphology of γ phase, Nb microsegregation and kinetics of monotectoid decomposition were obtained. To validate the model, an ingot of U-5.5Nb (wt.%) was produced and temperature measuring experiment was carried out. As-cast microstructure at different position taken from the ingot was investigated by using optical microscope and SEM. The effect of cooling rate on γ phase precipitation and monotectoid decomposition of U-Nb alloys was also studied. The simulated results were compared with the experimental results and the capability of the model for quantitatively predicting the microstructure evolution of U-Nb alloys during solidification and consequent cooling process was assessed.

Keywords: U-Nb alloys; dendrite growth; monotectoid decomposition; cellular automaton; numerical simulation

1 Introduction

Uranium is widely used in the nuclear industry, especially in nuclear power plants, because of its high density and unique nuclear properties [1–5]. However, the poor corrosion resistance limits its potential use. In industrial applications, niobium is always added to uranium for increased oxidation resistance and enhanced ductility [4]. Due to the structure heredity, as-cast microstructure is very impor-

tant to its mechanical properties and the design of heat treatment process. The as-cast microstructure including the grain size and fraction of each phase as well as the composition distribution has a great influence on the heat treatment and final properties. However, less attention has been paid to the study of solidification process of U-Nb alloys compared with the research on heat treatment. Therefore, it is necessary to investigate microstructure evolution of U-Nb alloys during solidification process.

To date, there have only been a few published studies on phase transformation of U-Nb alloys during solidification and cooling process. Ma *et al.* [6] investigated the solidification of U-2Nb alloy by means of microscope and SEM, and the behavior of the segregation and the precipitated phase were determined. Chen [7] produced U-5Nb alloy in CaO crucible with a protective coating by vacuum-induced melting. The solidifying microstructure was obtained by quenching method, and microstructural morphology and the type of inclusion were investigated. Jackson *et al.* [8] investigated the solidification microstructure and solute microsegregation of U-6Nb alloy. The results showed that the most difference of Nb concentration at the grain boundary and the grain interior was about 2.5Nb, and it is hard to eliminate them in subsequent heat treatment process. Chen *et al.* [5] investigated the effects of the carbon addition on the morphology, composition, size distribution and formation mechanism of inclusions in U-5.5Nb alloy. On the whole, little attention has been paid to the solidification process of U-Nb alloys and it is lack of in-depth understanding on solidification microstructure.

U-Nb alloys are expensive and also radioactive, which limits the experimental research. In recent years, a variety of computer models, such as cellular automaton (CA) [9–16], Monte Carlo [17, 18] and phase field [19–21], have been developed to simulate the microstructure evolution of solidification process. However, due to the lack of experimental data, there was little study about the microstructure simulation of U-Nb alloys during solidification process. Li *et al.* [10] developed a two-dimensional CA model to simulate the dendrite morphology and microsegregation of U-

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6Nb alloy, and the dendrite growth and Nb microsegregation behavior was simulated. Su *et al.* [16] used a CA model to investigate the solidification microstructure and composition distribution of U-5.5Nb under different thermal conditions of cooling rate. Studies mentioned above focused on the nucleation and growth behaviors of primary phase of U-Nb alloys, while the monotectoid decomposition during the consequent cooling process was usually neglected. The results already known show slower cooling rates allow for Nb diffusion of increasing scale, and a lamellar structure associated with the monotectoid reaction occurs when the cooling rate is less than 0.2 K/s [3]. So, it is necessary to develop a model to simulate the through-process microstructure evolution of U-Nb alloys during solidification and consequent cooling process.

In this paper, a CA model has been developed to simulate the microstructure evolution of U-Nb alloys during solidification and consequent cooling process. By using the developed model, γ phase precipitation, Nb microsegregation behavior and monotectoid decomposition of U-Nb alloys were simulated, and the characteristic of solidified microstructure was predicted. Experiments were carried out, and the capability of the model for describing the microstructure evolution was discussed.

2 Model description

The phase diagram of U-Nb alloys is illustrated in Figure 1, and analysis of the solidification path was made with U-6Nb (U-14 at.%Nb) as an example. When $T < T_L$ (T_L is liquidus temperature), the molten metal transforms into γ phase until this stage is complete. When the temperature reaches T_m (T_m is monotectoid temperature), the monotectoid decomposition, $\gamma_1 \rightarrow \alpha + \gamma_2$, occurs. The lamellar structure associated with the monotectoid microstructure nucleating at inclusions and prior- γ grain boundaries. In the present model, the dendrite growth process of γ phase, Nb microsegregation behavior and consequent monotectoid decomposition can be simulated, and the as-cast microstructure can be obtained.

The CA method was used in the simulation. A two-dimensional computational domain was discretized into square cells with the cell size of Δx . Each cell has the following variables: (1) grain identifying variable; (2) cell status, which is one of L, γ , M, and L/ γ interface, where L is molten metal, γ phase precipitation from molten metal, M is monotectoid microstructure; (3) concentration (C_L and C_S); and (4) solid fraction (f_S). The state of the cells can be identified as solid ($f_S = 1$), liquid ($f_S = 0$) and L/ γ in-

Figure 1: The phase diagram of the U-Nb system reviewed by Koike *et al.* [1].

terface ($0 < f_S < 1$). At the beginning of the simulation, each cell was given the same initial temperature above the liquidus of the alloy and all the cells were the liquid state. The state transformation from liquid to interface can be achieved through the following ways: stochastic nucleation event, artificially setting certain cell's state as interface or captured by its neighboring solid cells [12]. The present model combines a continuous function CA description of grain growth with a finite difference (FD) computation of solute diffusion. Both the CA and FD components of the model run on the same regular spatial square grid and with the same time step. The temperature at each time-step was given by the cooling curve of the specimen, which was obtained either from a temperature simulation or from a temperature measurement. In order to simulate the microstructure evolution during solidification, the governing equations used to calculate the distribution of concentration and temperature, interface curvature, growth kinetics, solid fraction, and nucleation process will be described below.

3 The nucleation model

Nucleation process was described by a continuous Gaussian nucleation distribution model [22], and the total density of nuclei $n(\Delta T)$ at a given undercooling ΔT can be ex-

pressed as follows:

$$n(\Delta T) = \int_0^{\Delta T} [1 - f_{Solid}(\Delta T')] \frac{dn}{d(\Delta T')} d(\Delta T') \quad (1)$$

$$\frac{dn(\Delta T)}{d(\Delta T')} = \frac{N_{max}}{\sqrt{2\pi}\Delta T_\sigma} \exp\left(-\frac{1}{2}\left(\frac{\Delta T' - \Delta T_N}{\Delta T_\sigma}\right)^2\right), \quad (2)$$

where ΔT is the undercooling, $n(\Delta T)$ is the nucleus density, N_{max} is the maximum nucleus density, ΔT_σ is the standard deviation of the distribution, ΔT_N is the mean nucleation undercooling, $\Delta T'$ is the undercooling integral element, and $f_{Solid}(\Delta T')$ is the fraction of solid phase.

When $T < T_L$, the total γ phase nucleus density can be calculated by Eq. (1) and Eq. (2), and the total number of γ grains in the calculation domain can be obtained. The sites of newly formed γ phase nuclei are randomly selected from liquid cells and a random integer representing a new crystallization orientation is given to the orientation variable. The capture process for a cell begins only when the cell is selected as γ phase nucleus (or totally solidified), it will alter its surrounding liquid neighbors into L/ γ interface state. In the CA model, the nearest four cells orthogonally (Von Neumann neighborhood) and the next nearest four cells diagonally (Moore neighborhood) are generally regarded as the neighboring cells, whose states will be changed with the parent cell in the center.

4 The solute diffusion

The growth process is mainly controlled by solute redistribution during solidification. Initially, the computational domain begins at a uniform composition, and as the solidification proceeds, the growing cells absorb solute from its neighboring liquid cells. Solute diffusion within the entire domain is then calculated based on the following equation:

$$\frac{\partial C_i}{\partial t} = \nabla \cdot (D_i \nabla C_i) + C_i (1 - k_0) \frac{\partial f_S}{\partial t} \quad (3)$$

where C is the composition with its subscript i denoting solid or liquid, D is the solute diffusion coefficient, and k_0 is the solute partition coefficient.

5 The S/L interface growth kinetics

During solidification process, the solute diffusion plays an important role in determining the dendritic growth and

microstructural characteristics. The local interface equilibrium composition at the interface can be calculated by the following equations:

$$C_L^* = C_0 + [T^* - T_L + \Delta T_R] / m_L \quad (4)$$

$$C_S^* = k_0 C_L^* \quad (5)$$

where T^* is the interface temperature, T_L is the liquidus temperature, C_L^* and C_S^* are the equilibrium liquid and solid composition, respectively, C_0 is the initial composition, ΔT_R is the curvature undercooling, and m_L is the slope of the liquidus line.

The curvature undercooling can be calculated by the following expression [9]:

$$\Delta T_R = \Gamma [1 - 15\epsilon (4(\varphi - \theta))] \cdot K \quad (6)$$

where Γ is the Gibbs–Thomson coefficient, ϵ is the degree of anisotropy of the interfacial energy, φ is the inclined angle of the normal to the interface with respect to the coordinate axis, θ is the angle between the preferred growth direction and the coordinate axis, and K is the curvature of the interface.

The average curvature for an interface cell is affected by its neighboring cells, and is calculated with the following expression [23]:

$$K = \left[1 - 2 \left(f_S + \sum_{i=1}^N f_S(i) \right) / (N + 1) \right] / \Delta x \quad (7)$$

where $f_S(i)$ and N are the solid fraction and the number of the neighboring cells. In the present model, N equals 8.

Solute conservation at the L/ γ interface is given by [9]:

$$v_n C_L^* (1 - k_0) = -D_L \left(\frac{\partial C_L}{\partial x} + \frac{\partial C_L}{\partial y} \right) + D_S \left(\frac{\partial C_S}{\partial x} + \frac{\partial C_S}{\partial y} \right) \quad (8)$$

where v_n is the normal velocity of the interface.

The two-dimensional sketch of virtual front-tracking algorithm for orthogonal grid previously proposed was adopted in the present model [9, 13]. Figure 2 illustrates the process of virtual reconstruction of sharp S/L interface. The normal direction of each interfacial cell is determined by:

$$\mathbf{n} = -\nabla f_S / \|\nabla f_S\| = n_x \mathbf{i} + n_y \mathbf{j} \quad (9)$$

with $\mathbf{i}$ and $\mathbf{j}$ defining as the unit vectors along x-axis and y-axis, respectively. The length of the normal vector L is measured from the center of the interfacial cell along its

Figure 2: Schematic of the virtual front-tracking scheme [13].

normal direction and is proportional to the solid fraction f_S :

$$L = M \cdot f_S \cdot \Delta x \quad (10)$$

where M denotes the coefficient to ensure the reconstructed interface continuous. The capturing rules are analyzed in detail in [9].

After calculating v_n , the solid fraction increment of the interface in one time step Δt is calculated using the following equations [9]:

$$\Delta f_S = \frac{(v_n \Delta t)}{L} \quad (11)$$

$$L = \frac{\Delta x}{[\max(|\cos \varphi|, |\sin \varphi|)]}, \quad (12)$$

$$\text{then } f_S^{t_0+\Delta t} = f_S^{t_0} + \Delta f_S, \quad (13)$$

where Δf_S is the increment of solid fraction, $f_S^{t_0}$ and $f_S^{t_0+\Delta t}$ are the solid fraction at the current time step and the next time step, respectively. When f_S equals 1, the L/ γ interface cell becomes solid and captures the neighboring liquid cells, and the captured cells change its state to L/ γ interface.

6 The monotectoid transformation

With the temperature decreasing, the molten metal will transform into γ phase totally. The simulated prior- γ microstructure was used as the initial condition for simulation of monotectoid decomposition process. Monotectoid

decomposition starts to nucleate at inclusions and prior- γ grain boundaries when the temperature drops to T_M . In this work, the effect of inclusions was neglected in the simulation. It is assumed that the nucleation of monotectoid decomposition occurs at some sites on the prior- γ grain boundaries. Based on the classical nucleation theory, the nucleation model for monotectoid decomposition can be described as follows [24, 25]:

$$I = K_1 D_\gamma (kT)^{-\frac{1}{2}} \exp\left(-\frac{K_2}{kT(\Delta G)^2}\right) \quad (14)$$

where K_1 is a constant related to the nucleation site density, K_2 is a constant related to interface energy, D_γ the niobium diffusion coefficient in γ phase, k is the Boltzmann's constant, ΔG is the driving force for the nucleation of monotectoid decomposition. The nucleation density for monotectoid microstructure, n_M , can be expressed as:

$$n_M = \int_T^{T_M} \frac{I(T')}{\varphi(T')} dT' \quad (15)$$

where $\varphi(T')$ is the cooling rate.

The growth velocity of monotectoid decomposition, $v^{\gamma/M}$, can be calculated by [26]:

$$v^{\gamma/M} = a(\Delta T_M)^2 \exp\left(-\frac{Q}{RT}\right) \quad (16)$$

where ΔT_M is undercooling of monotectoid transformation ($\Delta T_M = T_M - T$), a is a parameter, R is the universal gas constant, Q is the activation energy for atom motion at the interface. In this work, a and Q were obtained by fitting the model to the experimental results reported in reference [27]. Once a γ phase cell changes monotectoid microstructure state, it will begin to grow into the prior- γ region at a uniform velocity. The radius of the monotectoid microstructure at time t , R_t , can be obtained:

$$R_t = R_{t-\Delta t} + v^{\gamma/M} \cdot \Delta t \quad (17)$$

where $R_{t-\Delta t}$ is the radius at the previous step.

7 Results and discussion

7.1 The solidification microstructure

The nominal composition of the U-Nb alloys used in this study was 5.5wt.% high purity niobium and 94.5wt.% depleted uranium. The materials were melted in an arc melting furnace under vacuum up to 10 Pa at temperatures of about 1520°C for 30 min, and then the liquid metal was

Figure 3: The temperature change curves at different positions inside the casting during solidification.

Figure 4: Optical and SEM images of as-cast U-5.5Nb alloy at different position. (a) and (b) at top, (c) and (d) at middle.

directly cast into ingots (100mm in diameter and 200mm in height). In order to investigate the influence of cooling rate on the as-cast microstructure of U-5.5Nb alloy, thermocouples were positioned at the top and the middle of the casting to measure the temperature variation during the solidification process. Samples for microstructure observation were taken from the position of the thermocouples. Standard metallographic techniques were adopted for grinding and polishing. The 5% H_3PO_4 aqueous solution was applied for electroetching and constant 2V DC

Table 1: Properties of the U-5.5Nb alloys used in the following simulations.

Definition and symbols	values
Liquidus temperature T_L (K)	1633
Liquidus slope m_L (K·wt.% ⁻¹)	37.3
Maximum nucleus density N_{max} (m ⁻²)	7.0×10^9
Standard deviation of the distribution ΔT_σ (K)	0.5
Mean nucleation undercooling ΔT_N (K)	5
Liquid diffusion coefficient D_L (m ² ·s ⁻¹)	1.05×10^{-9}
Solid diffusion coefficient D_S (m ² ·s ⁻¹)	3.14×10^{-10}
Partition coefficient k_0	2.0
Gibbs-Thomson coefficient Γ (K·m)	1.9×10^{-7}
Monotectoid temperature T_M (K)	920
Growth parameter for monotectoid transformation a (m·s ⁻¹ ·K ⁻²)	3.55×10^8
Activation energy Q (J·mol ⁻¹)	338039

bias was used for the etching. The microstructure of U-Nb alloys was investigated by means of optical microscopy (OM) and scanning electron microscopy (SEM). The fraction of monotectoid decomposition was measured by using Image-pro Plus 6.0 (IPP 6.0) software.

Figure 3 shows the measured cooling curves at different positions inside the casting during solidification. It can be that the top position inside the casting cool faster than the middle position during solidification and consequent cooling process. As-cast microstructure of U-5.5Nb alloy at different position taken from the ingot were shown in Figure 4. Figures 4(a) and 4(c) show optical images of as-cast U-5.5Nb alloy at different positions where thermocouples were placed. Figures 4(b) and 4(d) show the corresponding high-magnification SEM images in high magnification. It can be seen that monotectoid decomposition (dark region in Figures 4(a) and 4(c), while bright region in Figures 4(b) and 4(d)) tends to occur at prior- γ grain boundaries and inclusions and it is consistent with the reported results.

7.2 The numerical simulation

The CA model was used to predict two-dimensional microstructure evolution of U-5.5Nb alloy. The computational domain was divided into 900×900 rectangular cells, and the cell size is 1μm. For the simulation the boundary was adiabatic so the heat flux and the atom flux are zero on the boundary. The parameters used in the simulation are shown in Table 1. The nucleation parameters were determined by adjusting simulated microstructure similar to ac-

Figure 5: The simulated microstructure (a, c) and corresponding Nb concentration (b, d) of U-5.5Nb alloy at the top of the casting during solidification process.

tual microstructure. By using the measured cooling curves in Figure 3 and the developed model, the continuous microstructure evolution of U-5.5Nb alloy during solidification can be simulated, and the as-cast microstructure can be predicted.

The simulated results of γ phase precipitation from liquid metal at different positions where thermocouples were placed inside the casting are shown in Figure 5 and Figure 6. Figure 5(a) and Figure 6(a) show the simulated dendrite morphology during solidification, where the white area represents liquid metal and different orientation γ grains are represented by different colors. Figure 5(b) and Figure 6(b) show the corresponding Nb concentration of U-5.5Nb alloy. Figures 5(c)-5(d) and Figures 6(c)-6(d) show the final microstructure and corresponding Nb concentration when the molten metal has transformed into γ phase totally. It can be seen that nucleation occurs in liq-

uid with the lowering of temperature, and the dendrites will grow with the decreasing temperature until they meet each other. With the increase of the cooling rate, the average grain size decreases and the Nb microsegregation increases. The simulated γ phase microstructure can be used as the initial condition for simulation of monotectoid decomposition process.

The simulated microstructure of monotectoid decomposition at different positions where thermocouples were placed inside the casting is shown in Figure 7. The blue area denotes γ phase region, and the yellow region denotes monotectoid microstructure. It can be seen that monotectoid microstructure nucleate at γ grain boundaries and then grow into γ grain interior. With the proceeding of the transformation, more and more monotectoid microstructure form and the impingement appears. The calculated area fraction of monotectoid microstructure in Fig-

Figure 6: The simulated microstructure (a, c) and corresponding Nb concentration (b, d) of U-5.5Nb alloy at the middle of the casting during solidification process.

Figure 7: The simulated microstructure of monotectoid decomposition at different positions. (a) at top, (b) at middle.

Figure 8: The calculated kinetics of monotectoid decomposition at different positions.

ures 7(a) and 7(b) are 22.4% and 49.7%, respectively, which is basically consistent with the experimental result in Figure 4. The results show that the fraction of monotectoid microstructure of as-cast microstructure of U-5.5Nb alloy decreases with the increase of the cooling rate. The calculated kinetics of monotectoid decomposition during cooling process is shown in Figure 8. It can be seen that the transformation rate increases at first and decreases gradually during the cooling process. The fraction of monotectoid microstructure at a given temperature decreases with the increase of the cooling rate. This is because the time for monotectoid decomposition is short with the increase of the cooling rate.

8 Conclusions

A cellular automaton model has been developed to simulate the microstructure evolution of U-Nb alloys during solidification and consequent cooling process. By using the developed model, nucleation and growth of γ phase, Nb microsegregation behavior and monotectoid decomposition of U-5.5Nb alloy were simulated. The evolution of dendrite morphology, Nb microsegregation behavior and kinetics of monotectoid decomposition under different cooling rate were obtained. The results showed that with the increase of the cooling rate, the average grain size of γ phase decreases and the Nb microsegregation increases. The fraction of monotectoid microstructure of as-cast microstructure decreases with the increase of the cooling rate. The transformation rate of monotectoid decomposition increases at first and decreases gradually during the cooling process, and the fraction of monotectoid microstructure

at a given temperature decreases with the increase of the cooling rate. To validate the model, an ingot was produced and metallographic examination was carried out. It was shown that the simulated as-cast microstructure is in good agreement with the experimental result. The developed CA model can be used to simulate the microstructure evolution of U-Nb alloys during continuous cooling process and predict the final microstructure.

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