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Formation of Nb-rich droplets in laser deposited Ni-matrix microstructures

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ABSTRACT

Ni-rich γ cells and Nb-rich eutectic droplets that form during laser power bed fusion solidification of Ni-Nb alloys are studied using experiments and simulations. Finite element simulations estimate the cooling rates in the melt pool and phase-field simulations predict the resulting cellular microstructures. The cell and droplet spacings are determined as a function of cooling rate and fit to a power law. The formation of Laves phase is predicted for a critical composition of Nb in the liquid droplets. Finally, our simulations demonstrate that anisotropy in the γ orientation influences the Laves fraction significantly.

Ni-based superalloys possess excellent mechanical properties and corrosion resistance up to high temperatures primarily due to the fine precipitation of Nb-rich phases and are therefore used in gas-turbine and jet-engine components [1]. The laser powder bed fusion (LPBF) additive manufacturing (AM) process is used to fabricate or repair these alloys by layer-by-layer application of the alloy powder and subsequent repeated melting, solidification and solid-state phase transformations [2–5]. The solidification in this process often results in a columnar face-centered-cubic γ-Ni matrix and microsegregation of Nb, Mo and Ti in the interdendritic regions [2,6-8]. The regions with high concentration of Nb often transform to intermetallic phases during terminal solidification. One of those phases is the Laves phase. Laves drastically reduces the tensile strength, fracture toughness and low-cycle fatigue properties of the additively manufactured material. An understanding of the formation and control of Laves is therefore essential.

Under nonequilibrium solidification conditions, solute redistribution across a solid-liquid interface during the growth of primary γ phase leads to severe Nb segregation in the liquid molten pool [9]. During terminal solidification, as the roots of the γ cells coalesce in the semisolid mushy zone, Nb-rich liquid channels between the cells are separated into isolated droplets [10]. It is difficult to entirely avoid the formation of these droplets due to the rapid nature of cooling during LPBF solidification. The metastable liquid in the form of droplets could potentially undergo a nonequilibrium reaction below the eutectic temperature and transform to a combination of Laves and γ. Since Laves is brittle and makes the as-deposited microstructures weak, there have been several experiments [11–15] and simulations [16] to suggest approaches to minimize its formation. The most widely used approach is homogenization heat treatment [8,14,17-19]. A manipulation of the solidification conditions in the melt pool by heat input/cooling rate was also found to be effective in controlling the morphology and distribution of Laves. High cooling rates resulted in a fine and discrete Laves network beneficial for mechanical properties, whereas low cooling rates resulted in a coarse and continuous network detrimental for the same [11,16,19,20]. Laves was found to be refined significantly and reduced/eliminated in ultrarapid cooling rates [12,13,19]. The morphological transition of γ phase from columnar to equiaxed, due to an increase in the cooling rate, was found effective for separating a continuous liquid network into isolated droplets/Laves [16,20]. While previous studies considered cooling rates on the order of 10^3 K s^{-1}, the present work uses cooling rates on the order of 10^6 K s^{-1}, consistent with LPBF. The microstructure-property correlation is therefore expected to be different than that reported in the existing literature. In what follows, we present the as-deposited microstructures from LPBF experiments and finite element and phase-field simulations.

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Fifteen millimeter cubes were additively produced from virgin Inconel 625 alloy powder using an EOS M270 LPBF system[1]. The standard EOS parameter set for this alloy was used consisting of a laser power of 195 W, scan speed of 800 mm s^{-1}, nominal powder layer thickness of 20 μm and hatch spacing of 100 μm. The samples were then cut from the build plate with electro-discharge machining in the as-built condition, i.e. the specimens did not undergo any stress-relief heat treatment. Portions of the as-built material were mounted and polished using standard metallographic techniques for scanning electron microscopy (SEM) analysis [21]. The samples were etched via immersion in aqua regia for 10 s to 60 s to reveal the microstructure. A final polishing step using a vibrational polishing system with 0.2 μm colloidal silica was employed to provide a strain-free surface for SEM electron backscatter diffraction and energy dispersive X-ray spectroscopy (EDS). The as-built microstructures from the EDS spectra consist of Ni-rich γ matrix and Nb, Mo, C and other elemental segregation [8,22]. We consider a binary analog of these microstructures, i.e. Ni matrix and Nb segregation, to describe the microstructural evolution. The as-built microstructures are presented in Fig. 1. These consist of primary Ni-rich γ cells/dendrites (average spacing $\approx$ 0.6 μm) and Nb-rich interdendritic regions. Although not clear, the secondary/tertiary sidearms cut the interdendritic space into smaller regions, and left less space for Nb-rich spots to grow in a sphere-like morphology. These spots appear bright and are extremely fine; the average spacing is $\approx$ 0.26 μm, average diameter is $\approx$ 0.13 μm, and average area fraction is $\approx$ 2% to 3%. The concentration of these spots could not be resolved since the beam spot size was quite large with respect to the size of the segregation features.

The solidification conditions in the above experiment were estimated by heat transfer finite element simulations and reported in previous works by us and our collaborators [5,23,24]. Here we show the typical temperature distribution during this LPBF simulation in Fig. 2. Referring to this temperature profile, the γ cells/dendrites solidify directionally and grow perpendicular to the solid-liquid boundary approximated by T_l isotherm in a temperature gradient G and at a solidification velocity V. The solid-liquid growth front represents different G and V. We note that G ranges from 2.4×10^7 K m^{-1} to 0.14×10^7 K m^{-1} and V ranges from 0.01 m s^{-1} to 0.3 m s^{-1} as we move from the bottom to the rear of this boundary. G is translated along the build direction (z) by the pulling velocity V in a directional "frozen temperature" solidification framework for microstructure evolution. G times V is the cooling rate $\dot{T}$.

We use a phase-field model detailed in Refs. [5,10] where the first simulations of γ cells during solidification of a dilute Ni-Nb alloy, a binary approximation of a Ni-based superalloy, were reported. A conserved composition field c and a non-conserved phase-field variable ϕ are used to label the microstructure phases; $\phi = 1$ in the solid, $\phi = -1$ in the liquid, and the solid-liquid interface is automatically extracted by the contour $\phi = 0$. An antitrapping solute flux [27,28] was introduced to minimize the interface-induced solute partitioning at low $\dot{T}$ leading to effective solute rejection in the liquid in front of the advancing cells. The effects of melt convection are not included in this model and solute is transported in the liquid by diffusion only. The time-dependent ϕ and c equations of motion are solved on a uniform mesh, using the finite volume method, explicit time stepping scheme and zero-flux boundary conditions. The size of the simulation box in the growth (z) direction is 40 μm, and a representative domain size, $L_x \times L_y$, 4 μm × 4 μm is used for 3D simulations. Other numerical and thermophysical parameters are detailed in Refs. [10,24]. In this parameter space, our simulation results presented below become virtually independent of the discretization size (8 nm) and the interface thickness (10 nm) values.

The solidification pathway for a Ni–5% Nb[2] alloy is given by $L \rightarrow L + \gamma \rightarrow \gamma +$ Laves [25,26]. These phase transformations occur due to different degrees of undercooling below T_l, and the resultant microstructures are predicted by phase-field simulations in Fig. 3. The microstructures consist of Ni-rich γ cells and Nb-rich intercellular regions and correspond only to a particular position along the melt pool boundary. The average distance between the γ cells remains constant in steady state, which is the cell spacing or the primary dendrite arm spacing. As the cells grow in the liquid, Nb is rejected through the cell-liquid interface in a nonequilibrium partitioning process [5,24]. Nb thus varies in the liquid ahead of the cell tip, in the liquid between cells, and in the solid cell core, resulting in complex segregation features (Fig. 3b). The microsegregation or the composition gradient between the cell core and the periphery of individual cells is extracted by a composition-distance profile across the cells and reported in [24]. The rejection of Nb by the growing cells increases the Nb content in the liquid. During terminal solidification, close to the bottom of the simulation box, as the roots of the solid cells grow toward each other and coalesce in the mushy zone at a low temperature, Nb-rich liquid in the intercellular channels is separated into isolated droplets, as in [24,29-31]. Since the diffusion path is absent at lower temperatures, the Nb content in these droplets increases rapidly with a reduced residual liquid fraction with increasing distance below the cellular growth front. These droplets could undergo eutectic transformation beyond a threshold composition of Nb, resulting in (γ + Laves) eutectic. The present binary model does not represent any phase beyond L and γ. Therefore, the formation of Laves is predicted using a criterion for the threshold composition of Nb in the liquid. Different critical values of Nb were reported in literature to describe the Laves formation. Dupont et al. considered the Laves formation due to a Nb composition in the liquid > 23%, whereas Nastac and Stefanescu [26] and Peng et al. [32] used a value of 19% to describe the same. We consider the criterion used by Nie et al. [16] and Antonsson et al. [13]. In this approach, the liquid with Nb > 20% transforms into Laves phase and the liquid with Nb $\leqslant$ 20% transforms into γ phase.

The spacing between the γ cells as well as between the droplets is related to the yield and tensile strengths of the solidified material. Prediction and control of the spacing between γ and droplets are therefore essential. We extract the average cell and droplet spacings from the simulated cellular microstructures by the calculation of the mean power spectrum: $S(k) = |h(k)|^2$, where $h(k)$ is the Fourier transform of the solid-liquid interface profile $h(z)$ and k is the wave number. From this analysis, λ_c and λ_d are estimated by λ_c (or λ_d) = $2\pi/k_{mean} = \sum_{k>0} kS(k)/\sum_{k>0} S(k)$, as shown in Fig. 4a. The highest peak in this spectrum corresponds to the dominant wavelength in the microstructure, i.e. λ_c. For the simulated cooling rates, the estimated λ_c ranges from 0.1 μm to 0.5 μm in 3D (Fig. 4b) and from 0.2 μm to 1.1 μm in 2D (Fig. 4c). Solute rejection/diffusion at the cell tip is more efficient in 3D than that in 2D; λ_c is therefore smaller in 3D than in 2D. The simulated cell spacing data agree reasonably with our experiment, where λ_c is estimated between 0.5 μm and 1.0 μm. Similar observations were also made by Amato et al. [33] for Inconel alloys.

The second dominant wavelength in Fig. 4a is the droplet spacing λ_d. In our simulations, λ_d ranges from 0.1 μm to 0.2 μm in 3D (Fig. 4b) and from 0.1 μm to 0.6 μm in 2D (Fig. 4c). The droplets are finer in 3D than that in 2D due to the same above reason for λ_c. Note that the average λ_d estimated from the experiment is $\approx$ 0.26 μm, which compares reasonably with our simulation data. Since the droplets form

[1] Any mention of commercial companies or products herein is for information only; it does not imply recommendation or endorsement by NIST.

[2] Concentration is represented in mass fraction.