Three-dimensional Dendritic Needle Network model with application to Al-Cu directional solidification experiments

D. Tourret¹,², A. Karma¹, A. J. Clarke², P. J. Gibbs², S. D. Imhoff²
¹ Northeastern University, Department of Physics and Center for Interdisciplinary Research on Complex Systems, Boston, MA, USA
² Los Alamos National Laboratory, Materials Science and Technology Division, Los Alamos, NM, USA
E-mail: dtourret@lanl.gov

Abstract. We present a three-dimensional (3D) extension of a previously proposed multi-scale Dendritic Needle Network (DNN) approach for the growth of complex dendritic microstructures. Using a new formulation of the DNN dynamics equations for dendritic paraboloid-branches of a given thickness, one can directly extend the DNN approach to 3D modeling. We validate this new formulation against known scaling laws and analytical solutions that describe the early transient and steady-state growth regimes, respectively. Finally, we compare the predictions of the model to in situ X-ray imaging of Al-Cu alloy solidification experiments. The comparison shows a very good quantitative agreement between 3D simulations and thin sample experiments. It also highlights the importance of full 3D modeling to accurately predict the primary dendrite arm spacing that is significantly over-estimated by 2D simulations.

1. Introduction
Since the early descriptions of steel dendrites by Grignon [1] in 1775 and Chernoff [2] in 1880, much has evolved in our understanding of dendritic microstructures, including the physical origin of their morphological complexity, and the laws underlying their growth dynamics [3]. However, the schematic drawings of dendritic microstructures contained in these early works (some of which are reproduced in Fig. 1) already capture salient features of the dendritic network organization, such as dendritic arm spacings, crucial to final mechanical properties.

Figure 1. Schematic drawings of steel dendrites after (a) Grignon [1] (1775) and (b) Chernoff [2] (1880).
Recently, a new multi-scale approach to simulate dendritic solidification was proposed [4,5] that conceptualizes the dendritic network as a hierarchical structure of primary, secondary, and higher order branches, with each arm represented as a thin needle much like in the drawings of Fig. 1. Additionally, this approach provides a rigorous theoretical footing for describing the needle dynamics on the dendritic grain scale by integrating out the interface dynamics on the scale of the dendrite tip radius. In concentrated alloys that solidify at low undercooling, this Dendritic Needle Network (DNN) approach reproduces analytical and phase field predictions of crystal growth dynamics in both transient and steady-state regimes [5], with much faster simulations than the well established phase field approach [6], since it does not explicitly track the evolution of the complex solid-liquid interface pattern on the dendrite tip scale. This new approach aims at bridging the modeling scale gap that exists between phase field [6] at the scale of a dendrite (or a small dendritic array), and continuum averaged approaches, such as cellular automaton based models [7] at the scale of a technological casting process. In this article, we present the first implementation of the multi-scale DNN model in three dimensions (3D) and its application to the directional solidification of an Al-12at.% Cu alloy, which we compare quantitatively to experimental data from in situ X-ray radiography.

2. Modeling

2.1. DNN model in two dimensions

As detailed in Ref. [5], the evolution of a binary alloy solid-liquid interface at a given temperature, \( T_0 \), in a diffusive regime with negligible solid state diffusion follows

\[
\partial_t u = D \nabla^2 u, \tag{1}
\]

\[
[1 - (1 - k)u] = D \partial_n u_i, \tag{2}
\]

\[
u_i = d_0 f(\theta) \kappa, \tag{3}
\]

where \( u \equiv (c_l^0 - c)/(1 - k)c_l^0 \) is a reduced solute field, with \( c \) the solute concentration field, \( c_l^0 \) the equilibrium concentration at the temperature \( T_0 \), and \( k \) is the solute partition coefficient at the solid-liquid interface. Equation (1) represents the diffusion of solute in the liquid with a diffusion coefficient \( D \). Equation (2) is a statement of solute mass conservation at the interface, giving the normal velocity of the solid-liquid interface, \( v_n \), as a function of the value of \( u \) on the liquid side of the interface, \( u_i \), and its normal gradient, \( \partial_n u_i \). Equation (3) is the Gibbs-Thomson condition for equilibrium at the interface, with \( f(\theta) \) the orientation dependence of the capillary length, \( d_0 = \Gamma/|m|(1 - k)c_l^0 \), of the alloy of Gibbs-Thomson coefficient \( \Gamma \) and liquidus slope \( m \), and \( \kappa \) is the local curvature of the interface [8]. For isothermal solidification at \( T_0 \), the far-field condition \( c \to c_\infty \) reads \( u \to \Omega \) with a solute supersaturation \( \Omega \equiv (c_l^0 - c_\infty)/(1 - k)c_l^0 \).

In our specific regime of interest, i.e. at low supersaturation in concentrated alloys, dendritic microstructures exhibit very sharp needle-like branches similar to those schematized in Fig. 1, and one can write a solute mass balance at an intermediate length scale (Fig. 2a) much larger than the dendritic tip radius, \( \rho \), and much smaller than the diffusion length, \( l_D = D/V \), with \( V \) the tip growth velocity. At this scale, the needle crystal is infinitely sharp, so that \( u_i \approx 0 \) from Eq. (3) and the solute locally follows Laplace equation \( \nabla^2 u = 0 \) in place of Eq. (1). This

![Figure 2. Schematics of a dendritic tip at an intermediate scale between the tip radius \( \rho \) and the much larger diffusion length \( D/V \) for a sharp (a) and a parabolic (b) needle crystal.](image)

\[\text{Figure 2. Schematics of a dendritic tip at an intermediate scale between the tip radius } \rho \text{ and the much larger diffusion length } D/V \text{ for a sharp (a) and a parabolic (b) needle crystal.}\]
problem has an analytical solution that exhibits a square-root singularity of amplitude $F$ of the $u$-field close to the tip as $\partial u / \partial y |_{y=0} \sim F / \sqrt{d_0(x_t - x)}$, where $+x$ is the needle growth direction, and $(x, y) = (x_t, 0)$ are the tip coordinates [9]. Since that singularity is similar to that of the stress field in linear elasticity at the tip of a crack in mode III fracture, one can calculate its amplitude $F$ using an integral along a contour $\Gamma_e$ around the tip (see Fig. 2a) similar to the classical J-integral of fracture mechanics [10]. Combining this expression of the normal gradient of $u$ around the tip with the fact that further behind the tip, the normal velocity of the solid-liquid interface is essentially 1D and follows $\partial u / \partial y |_{y=0}$, one can integrate the equation of the solid-liquid interface at this scale, yielding a parabolic shape of radius $\rho$ following [5]

$$\rho V^2 = 2D^2 F^2 / d_0.$$  

Together with the microscopic solvability condition at the scale of the tip radius $\rho$ [11, 12]

$$\rho^2 V = 2Dd_0 / \sigma,$$

with $\sigma$ the tip selection constant of the alloy, those two equations prescribe uniquely the instantaneous tip velocity, $V(t)$, and radius, $\rho(t)$, of a dendritic tip. While Eq. (5) was introduced as a steady-state solution, it was shown later that $\rho^2 V$ reaches a constant value very early during the transient development of a dendritic branch [13]. Hence, the product $\rho^2 V$ is kept constant and the model captures the evolution of the surrounding solute field and the transient interactions between individual dendrite arms through the evolution of $F(t)$.

For directional solidification within a frozen temperature field with $T(x) = T_0 + G(x - V_p t)$ with a pulling velocity $V_p$, if we set $T_0$ as the solidus temperature, Eq. (3) changes to

$$u_i = d_0 f(\theta) \kappa + (x - V_p t) / l_T$$

with a thermal length $l_T = |m|(1 - k)c_i^0 / G$ corresponding to the applied thermal gradient $G$, and the equilibrium along a needle reads

$$u_i = (x - V_p t) / l_T$$

instead of $u_i = 0$ for isothermal solidification at $T_0$. Thus the prefactor of $v_n$ in Eq. (2) is not $\approx 1$ anymore, and the dynamics of a needle tip depends on the value of $u$ at its tip, $u_t$, as [5]

$$\rho V^2 = 2D^2 F^2 / \{[1 - (1 - k)u_t]^2 d_0\}$$  

$$\rho^2 V = 2Dd_0 / \{[1 - (1 - k)u_t] \sigma\}$$

with the far-field condition on $u$ now corresponding to its liquidus value $u \to 1$ as $x \to \infty$.

### 2.2. Extension to a thick needle crystal

The model summarized above, and described in details in Ref. [5], cannot be readily extended to 3D, because no analytical solution exists in 3D for a Laplacian field around an infinitely thin needle abstracted as a line at a fixed concentration, such as the one yielding the square-root singularity of the solute gradient normal to the needle in the tip region in 2D. Therefore, first we developed a new formulation of the 2D DNN model for a needle of finite thickness with $u$ imposed within a parabolic shape of radius $\rho$ (Fig. 2b).

The integration of the solute conservation at the interface $D \partial u / \partial n \approx v_n$ in the tip region over a length $a \ll l_D$, i.e. over a contour $\Gamma_t$ between $x = x_t - a$ and $x = x_t$ as in Fig. 2b, yields

$$D \int_{\Gamma_t} \frac{\partial u}{\partial n} d\Gamma_t = \int_{y_a}^{y_t} V \partial y = 2V \sqrt{2\rho a},$$

$$\int_{\Gamma_f} \frac{\partial u}{\partial n} d\Gamma_f = -2V \sqrt{2\rho a}.$$
with \( y_a = y_i(x_t - a) = \sqrt{2\rho a} \). Thus, Eq. (9) can be rewritten in a form
\[
\rho V^2 = \frac{1}{8a} \left(D \int_{\Gamma_i} \frac{\partial u}{\partial n} \, d\Gamma_i \right)^2,
\]
that relates the product \( \rho V^2 \) to the square of the flux intensity factor defined by
\[
\mathcal{F} = \frac{1}{4\sqrt{a/d_0}} \int_{\Gamma_i} \frac{\partial u}{\partial n} \, d\Gamma_i
\]
(11)
with the identical Eq. (4) defined in the previous section, together with the product \( \rho^2 V \) still following Eq. (5) with a value of \( \sigma \) given by solvability theory \cite{14}. In order to integrate the value of \( \mathcal{F}(t) \), now defined by Eq. (11), we write the Laplace equation in a moving frame of velocity \( V \),
\[
D \nabla^2 u = -V \frac{\partial u}{\partial x},
\]
at a scale \( \ll D/V \). Integrating \( \nabla^2 u \) over a surface \( \Sigma \) bounded by the contours \( \Gamma_e \) and \( \Gamma_i \) shown in Fig. 2b, we can use the divergence theorem to write
\[
\int_{\Gamma_i} \frac{\partial u}{\partial n} \, d\Gamma_i = \int_{\Gamma_e} \frac{\partial u}{\partial n} \, d\Gamma_e + V D \int_{\Sigma} \frac{\partial u}{\partial x} \, d\Sigma,
\]
where, for consistency with the sharp needle model, we have the contour normal \( n \) pointing outward of the closed contour \( (\Gamma_e + \Gamma_i) \) along \( \Gamma_e \) (see Fig. 2a), while pointing outward of the dendrite, i.e. inward the contour, along \( \Gamma_i \) (see Fig. 2b). Finally, the “thick” DNN model follows the exact same growth laws as the sharp needle model, with a constant value of \( \rho^2 V \) and a product \( \rho V^2 \sim \mathcal{F}(t)^2 \). The only difference resides in the new definition of the flux intensity factor \( \mathcal{F}(t) \) as Eq. (11), where the contour integral term can be integrated over a chosen outer contour \( \Gamma_e \) using Eq. (12).

2.3. Extension to three dimensions

The model thus defined can be directly extended for a 3D dendrite growing as a paraboloid of revolution with a circular cross section of radius \( r_i(x) = \sqrt{2\rho(x_t - x)} \). In 3D, \( \Gamma_e \) and \( \Gamma_i \) are surfaces joining at \( x = x_t - a \), and \( \Sigma \) is the volume enclosed within the closed surface \( (\Gamma_e + \Gamma_i) \). The mass conservation \( D\partial u/\partial n \approx v_n \) in the tip region over a length \( a \) now yields
\[
D \int_{\Gamma_i} \frac{\partial u}{\partial n} \, d\Gamma_i = \int_0^{r_a} \int_0^{2\pi} V r \, dv \, d\theta = 2\pi a \rho V,
\]
(13)
with \( r_a = \sqrt{2\rho a} \) the radius of the paraboloid at a distance \( a \) behind the tip, i.e. at \( x = x_t - a \). Thus, we can define the flux intensity factor
\[
\mathcal{F} = \frac{1}{2\pi a} \int_{\Gamma_i} \frac{\partial u}{\partial n} \, d\Gamma_i,
\]
(14)
and write the flux condition at the tip that replaces Eq. (4) as
\[
\rho V = D\mathcal{F},
\]
(15)
which follows from Eqs. (13) and (14). Similarly as in two dimensions, the flux term over the surface \( \Gamma_i \) can be calculated over a surface \( \Gamma_e \) that intersects the paraboloid at \( x = x_t - a \) with
\[
\int_{\Gamma_i} \frac{\partial u}{\partial n} \, d\Gamma_i = \int_{\Gamma_e} \frac{\partial u}{\partial n} \, d\Gamma_e + V D \int_{\Sigma} \frac{\partial u}{\partial x} \, d\Sigma,
\]
(16)
The growth dynamics of the needle tips can thus be calculated by combining the new flux balance Eq. (15) with the solvability condition Eq. (5), which is similar as in 2D with a tip selection parameter $\sigma$ that still only depends on the strength of surface tension anisotropy, but with a different value as compared to 2D [14].

In the case of directional solidification, similarly as in 2D, we replace the equilibrium condition along the needle $u_i = 0$ by $u_i = (x - V_p t)/l_T$ and the non-vanishing prefactor of $v_n$ in Eq. (2) appears in the needle growth laws, now combining Eq. (8) and

$$\rho V = DF / [1 - (1 - k)u_t].$$

(17)

3. Results and Validation

First, we test the steady-state growth regime predicted by the different versions of the DNN model — namely 2D “sharp” (Sec. 2.1), 2D “thick” (Sec. 2.2), and 3D (Sec. 2.3) — against the exact analytical solution for a parabola/paraboloid established by Ivantsov [15, 8], relating uniquely the tip solute supersaturation $\Omega$ to the Péclet number $P \equiv \rho V/(2D)$. In order to do so, we let one needle grow within an imposed $\Omega$ and measure the value of $P$ once $\rho$ and $V$ reach steady values. Additionally, we define $\rho_s$ and $V_s$ the steady-state values of respectively $\rho$ and $V$ that satisfy the solvability condition Eq. (5) or Eq. (8) together with the Ivantsov solution.

In 2D (Fig. 3), as previously explained in Ref. [5], the sharp-needle formulation tends to a first order approximation $\Omega \approx \sqrt{\pi P}$ of the exact Ivantsov solution $\Omega = \text{Iv}_{2D}(P) = \sqrt{\pi P} e^{P} \text{erfc}(\sqrt{P})$, hence limiting the scope of the approach to $\Omega \ll 1$. In contrast, the DNN formulation with a parabolic needle of finite thickness tends to the exact 2D Ivantsov solution. Therefore, in addition to making the extension to 3D possible, the thick needle formulation has the advantage of increasing the range of Péclet number that can be quantitatively modeled. This is because this formulation provides a more accurate description of the solute field, and because the calculation of $F(t)$ by Eq. (12) introduces a correction in $V$ from the moving frame expression of Laplace equation around the tip.

In 3D (Fig. 4), the predictions of the DNN model for a paraboloid follows the steady-state Ivantsov solution $\Omega = \text{Iv}_{3D}(P) = P e^{P} \int_{P}^{\infty} (e^{-s}/s) ds$ — significantly different from $\text{Iv}_{2D}(P)$ plotted for comparison in Fig. 4. This difference between 2D and 3D also has a consequence in terms of appropriate computational parameters. The DNN model is built on the assumption

![Figure 3. Steady-state predictions of 2D isothermal DNN simulations using a sharp needle (Fig. 2a) with $\Delta x = 10\rho_s$, and using a thick parabolic needle (Fig. 2b) with $\Delta x = \rho_s$.](image1)

![Figure 4. Steady-state predictions of 3D isothermal DNN simulations for $r_{max}/\rho_s = 10$ with $l_D/\Delta x = 50, 20$, and for $l_D/\Delta x = 20$ with $r_{max}/\rho_s = 100, 10, 5, 2, 1$.](image2)
of a scale separation between the tip radius $\rho$ and the diffusion length $l_D = D/V$. As seen in Fig. 4, for a given $\Omega$ the ratio $\rho/(D/V) \sim P$ is lower in 2D than in 3D. Thus in 3D, the choice of grid spacing $\Delta x \sim \rho$ is more severely bounded by $\Delta x \ll l_D$ than in 2D. However, this is not such a stringent restriction, as the model reaches accurate results up to $l_D/\Delta x \approx 20$ (Fig. 4).

In the broader context of interactions among multiple branches, when a needle is stopped by diffusive interactions, the constancy of $\rho^2 V$ would imply $\rho \to \infty$ as $V \to 0$. While this is not a problem with a sharp needle as $u$ is only imposed on a sharp line, this may lead to numerical instabilities when $u$ is imposed on the whole thickness of a needle. A way to avoid these instabilities is to bound the thickness of a needle to a given radius $r_{\text{max}}$. Fig. 4 shows that using a $r_{\text{max}}$ as low as the steady-state tip radius $\rho_s$ does not significantly affect the results.

We have also tested the DNN predictions of the early transient growth of an equiaxed crystal. In 2D, the growth of an idealized crystal with four sharp branches has an analytical solution with the branches length following $L \sim t^{3/5}$ [16]. This solution was reproduced by phase-field calculations. In 3D, dimensional analysis validated by phase field calculations yields $L \sim t^{2/3}$ [13]. The results of the 3D DNN simulations in Fig. 5 for $\Omega \approx 0.006$ (i.e. $P \approx 0.02$) show that the model also captures accurately the transient equiaxed growth regime.

**Figure 5.** Early growth of a six-branch equiaxed crystal predicted by 3D DNN simulations, compared to a line of slope $2/3$, according to the analytical law $L \sim t^{2/3}$ [13]. The plot is built by continuity from simulations with $\Delta x = 0.05 \rho_s$ (for $t V_s/\rho_s < 2$), $\Delta x = 0.1 \rho_s$ ($2 < t V_s/\rho_s < 10$), and $\Delta x = 0.5 \rho_s$ ($t V_s/\rho_s > 10$). The insets show snapshots of the isovalues of $u$ with steps of 0.0005.

### 4. Application to Al-Cu directional solidification experiments

#### 4.1. Experiments

Aluminum-copper alloys are good candidate for in situ observation of dendritic solidification in metals and they have been extensively used for X-ray radiography [17, 18]. We prepared an alloy of nominal composition Al-12at.%Cu, i.e. $c_{\infty} = 24.3$wt.%Cu, by arc-melting in an inert argon atmosphere. Thin samples ($\approx 200 \mu m$) were obtained by Electrical Discharge Machining and mechanical grinding. The resulting foil was put into a boron nitride crucible with a $\approx 0.1$ mm thinned window and 2 mm diameter observation hole. The (sample + crucible) assembly was inserted in a steel rod instrumented with thermocouples and independently heated by two induction coils located above and below the sample position. Directional solidification was achieved by stabilizing the temperature gradient with a melted alloy, then reducing the imposed temperatures at the location of the coils at a controlled cooling rate. Synchrotron X-ray imaging was performed at the Sector 32 Insertion Device beamline at Argonne National Laboratory’s Advanced Photon Source with a monochromatic beam at 18 keV. X-rays pass through the metal sample and impinge upon a scintillator that converts them to visible light. A mirror positioned behind the scintillator reflects the images to a CCD camera recording the 1024$\times$1280 pixel images with a pixel size of $\approx 1.34 \mu m$. The experiment presented here and compared to DNN simulations in Fig. 6 corresponds to a measured temperature gradient $\approx 72$ K/cm and a measured isotherm velocity $\approx 100 \mu m/s$, both expressed along the gravity axis (vertical in Fig. 6).
4.2. Simulations

We simulate the Al-Cu directional solidification experiments with DNN simulations, both in 2D (sharp-needle formulation [5]) and in 3D. The alloy parameters are $m = -3.4\ \text{K/wt\%Cu}$, $k = 0.17$, $D = 3.5 \times 10^{-9}\ \text{m}^2/\text{s}$, $\Gamma = 2.36 \times 10^{-7}\ \text{Km}$ [8], and $\sigma = 0.083$ in 2D and $\sigma = 0.06$ in 3D, corresponding to an interfacial anisotropy strength $\approx 1.2\%$ [14]. With a theoretical steady-state radius $r_s \approx 1.4\ \mu\text{m}$ in 2D and $1.8\ \mu\text{m}$ in 3D, we solve the diffusion and needle growth equations on a finite difference grid of spacing $\Delta x = 2r_s$, using an explicit Euler time scheme with time step $\Delta t = \Delta x^2/(4D)$ in 2D and $\Delta x^2/(6D)$ in 3D. The integration contour $\Gamma_e$ in 2D is a square of side $6\Delta x$ centered on each needle tip. In 3D, the integration surface is a cuboid centered on the tip with dimensions $2a = 6\Delta x$ in the growth direction $x$ and $(r_a+4\Delta x)$ in normal directions. The width along the needles is bound to $r_{max} = 3r_s$. Sidebranching events are triggered at a distance $d = (15 \pm 5)\rho$ behind the tip each time a needle grows by a distance $d$, as described in Ref. [5], with an initial length $l_0^{SB} = \sqrt{2pd} + \rho$. An orientation angle $\alpha$ of the crystal with respect to the vertical axis in the $(x, y)$ plane is introduced by adding a $y$-dependent term on the imposed $u_y = (x - V_p)/l_T - y \tan(\alpha)$ along the needles, in order to match the corresponding experimental crystal orientation $\alpha \approx 30^\circ$ (Fig. 6a). Hence, the equivalent pulling velocity and temperature gradient along the growth axis $x$ are input as $G = 72 \times \cos(\alpha) \approx 62\ \text{K/cm}$ and $V_p = 100/\cos(\alpha) \approx 115\ \mu\text{m/s}$. The simulation domain measures $\approx 3000 \times 2500\ \mu\text{m}^2$ in 2D and $\approx 3000 \times 2500 \times 100\ \mu\text{m}^3$ in 3D (half sample thickness with symmetry boundary conditions in $z$), over a total time of 180 (2D) and 120 (3D) seconds, both starting with 100 evenly spaced needles — in the $(z = 0)$ plane in 3D — with their tips located at the liquidus temperature.

4.3. Results

Fig. 6 shows the experimental observation (a) compared to DNN results in 2D (b) and 3D (c); all panels are represented at the same scale. The 2D calculation predicts an average primary spacing $\Lambda_{2D} \approx 1020\ \mu\text{m}$ (b), much larger than the experimentally measured $\Lambda_{exp.} \approx 258\ \mu\text{m}$ (a), while 3D simulations yields an excellent agreement with experiments as $\Lambda_{3D} \approx 237\ \mu\text{m}$ (c), with $\Lambda$ measured normal to the growth direction. The steady-state tip supersaturation of Fig. 6c is clearly shown that the 3D DNN model provides a powerful tool to make quantitative predictions of complex dendritic microstructures with time efficient calculations.

![Figure 6](image-url)

*Figure 6. Directional solidification of an Al-12at.\%Cu (Al-24.3wt\%Cu) alloy at $G = 62\ \text{K/cm}$ and $V_p = 115\ \mu\text{m/s}$: (a) X-ray image; (b) 2D simulation; (c) 3D simulation (all represented at the same scale). [Corresponding movies attached.]*
5. Summary and outlook
We presented the first three-dimensional extension to the previously proposed Dendritic Needle Network (DNN) multi-scale approach to dendritic solidification [5]. The model generally describes both transient and steady-state growth regimes. We compared model predictions with synchrotron X-ray imaging of an Al-Cu alloy during directional solidification. The 3D DNN predictions show excellent agreement with measured primary dendrite arm spacing, whereas the 2D modeling predicts significantly larger spacing than the experiments.

The present modeling approach bridges the length-scale gap between detailed solid-liquid interface pattern formation models, e.g. phase-field, and macroscopic average models of grain growth, e.g. cellular automata. The possibility to model and explore both intragrain and intergrain dynamical phenomena in complex dendritic microstructures should pave the way for a deeper understanding of microstructure selection mechanisms, such as for dendrite arm spacings, grain texture, and casting defects, crucial to the properties of industrial materials.

The works in progress and outlook for DNN modeling include studies on: scaling laws for dynamics of dendritic secondary sidebranches [19]; the Columnar-to-Equiaxed transition [20]; columnar grain growth competition [21]; and coupling the model with solute-driven fluid flow.

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