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Modeling of In-Situ Nitrogen Alloying in SS316 during Laser Directed Energy Deposition

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Abstract

Laser directed energy deposition (LDED) can alloy AISI 316 with nitrogen in situ by replacing part of the argon shielding and carrier gas with N₂. Whether useful nitrogen levels can be reached depends on how fast molecular nitrogen is absorbed during the short life of the melt pool, so equilibrium thermodynamics alone is insufficient. This work develops a coupled thermal–thermodynamic–kinetic model and reduces it to a single dimensionless group. An Eagar–Tsai moving Gaussian heat source supplies the melt-pool free-surface area, volume, area-averaged surface temperature and lifetime. Sieverts' law with temperature-scaled Wagner interaction parameters sets the equilibrium solubility. Because LDED intensities do not produce a nitrogen-dissociating plasma, absorption is treated as reversible dissociative exchange at the free surface, second order in dissolved nitrogen. The rate law has an exact hyperbolic-tangent solution governed by a Damköhler number, $Da = \beta N_{eq} \tau$. Powder-in-flight absorption, layer-to-layer remelting and a Scheil-based N₂ porosity criterion complete the model. For 800 W, 10 mm s⁻¹ and 0.5 atm N₂, the model predicts a mean pool surface temperature of 2897 K, an equilibrium ceiling of 0.085 wt% and $Da=0.22$. The steady-state deposit reaches 0.061 wt% N, i.e., 71% of the ceiling but only 42% of the gap between feedstock (0.043 wt%) and equilibrium, confirming kinetically limited operation for the uncalibrated rate constant. In this regime nitrogen pickup scales almost linearly with N₂ partial pressure rather than with its square root, rises with melt superheat (apparent activation energy 50.5 kJ mol⁻¹), and is strongly reduced by dissolved oxygen. Process maps over 600–1000 W, 5–15 mm s⁻¹ and 0.25–1 atm N₂ give 470–1000 ppm, below the 1410 ppm porosity threshold. Pure-argon deposition is predicted to lose nitrogen. The rate multiplier χ is the only adjustable kinetic parameter and requires experimental calibration.

Keywords: Directed energy deposition; In-situ alloying; Nitrogen; Austenitic stainless steel; Sieverts' law; Melt-pool kinetics; Damköhler number

1 Introduction

Nitrogen is an interstitial solid-solution strengthener in austenitic stainless steels. It also stabilizes austenite and improves localized corrosion resistance through its contribution to the pitting resistance equivalent number, $PREN = \%Cr + 3.3\%Mo + 16\%N$ [1]. Wrought 316LN grades typically contain about 0.10–0.16 wt% N and offer more than 100 MPa higher yield strength than 316L without a significant loss of ductility [2]. However, achieving the same benefit to additively manufactured 316

is difficult [3]. High-nitrogen steel (HNS) powder increases material cost, while gas nitriding of the powder creates a surface nitride layer whose dissolution during melting becomes an additional process variable [4]. In-situ N_2 alloying during DED can provide a viable solution to this problem, in which an $Ar-N_2$ mixture is introduced through the chamber atmosphere, the shielding gas or the powder carrier gas. The feedstock remains unchanged, the N_2 input is controlled through the process gas, and the gas composition can, in principle, be changed during a build to grade the deposit. Fig. 1 illustrates the in-situ nitrogen alloying during LDED.

Previous studies show that gas-based nitrogen alloying is feasible, but the reported pickup varies considerably among processes. Wire-arc DED with N_2 -containing shielding gas has produced high-nitrogen austenitic stainless steel deposits [5], and electromagnetic-field-controlled wire-arc DED of 316L has used in-situ nitrogen alloying to improve strength, ductility and corrosion resistance [6]. In LDED of 316L, in-situ interstitial nitrogen alloying improved wear and corrosion resistance [7], and $Ar-N_2-He$ shielding gases changed the microstructure and mechanical response of laser-MIG hybrid deposits [8]. In laser powder bed fusion, replacing argon with nitrogen produces only a small change in composition and properties [9], and gas-nitrided powder is used when higher nitrogen levels are required [4]. In fiber-laser welding of 304 under pure N_2 , the fusion-zone nitrogen increased from about 360 to 490 ppm [10], a pickup of roughly 130 ppm even though the equilibrium ceiling is much higher.

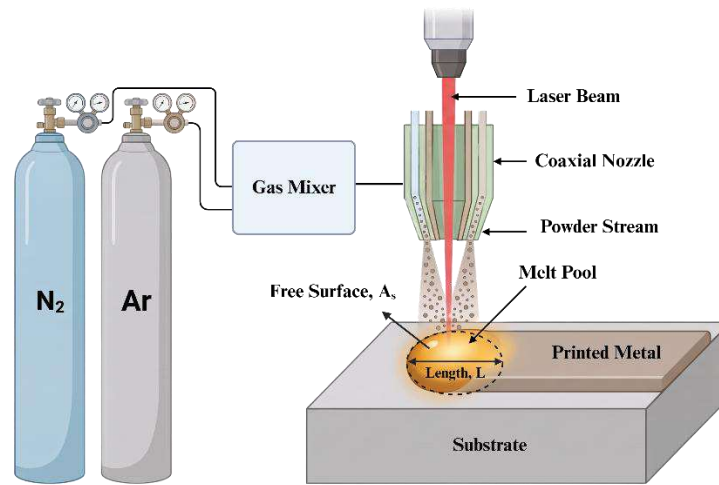


Fig. 1 Schematic of in-situ nitrogen alloying during LDED under an $Ar-N_2$ atmosphere. Nitrogen is supplied through shielding and powder-carrier gas and absorbed by the powder in flight and at the melt-pool free surface. The melt-pool geometry and thermal quantities - surface area A_s , volume V , length L , residence time τ , and mean surface temperature $\bar{T}_s$ - from the thermal model are used as inputs to the chemistry model.

These differences are consistent with a change in the gas–metal reaction mechanism. In arc processes, the plasma dissociates molecular nitrogen and exposes the pool to monatomic nitrogen, whose chemical potential is far higher than that of N_2 ; absorption is then fast and can exceed Sieverts' law [11, 12]. Classical models for nitrogen in stainless steel welds, including those of du Toit and Pistorius [13, 14] and Varbai and Májlínger [15], were developed for this situation. LDED operates differently. At typical DED intensities of about 10^4 – 10^5 $W\,cm^{-2}$ in conduction mode, the plume is not expected to dissociate nitrogen to a significant extent [16]. The melt pool is therefore exposed mainly to molecular N_2 , and nitrogen transfer requires dissociative chemisorption at the liquid surface [17, 18]. The reverse reaction is second order in dissolved nitrogen, so the overall approach to equilibrium is slow.

Applying an arc-welding nitrogen model directly to laser DED can therefore overpredict pickup, and so can equilibrium thermodynamics alone, because the pool may not live long enough to approach equilibrium. A laser DED formulation should (i) use an interfacial rate law appropriate for molecular N_2 , (ii) use a melt-pool thermal state predicted from the process conditions rather than an assumed temperature, and (iii) account for DED-specific effects, in particular powder exposure during flight and repeated remelting during layer deposition.

On this basis, the present work makes four contributions:

1. a closed-form solution for reversible dissociative nitrogen absorption, reduced to a Damköhler number that combines laser power, travel speed, N_2 partial pressure and alloy chemistry;
2. the prediction that, for the uncalibrated rate constant ($\chi = 1$), laser DED of 316 operates at $Da \approx 0.1$ – 0.6 , below equilibrium, which explains the modest nitrogen pickups reported for laser processes;
3. the prediction of a reversal in temperature sensitivity, in which hotter pools increase pickup when $Da \ll 1$ but lower it when $Da \gg 1$, together with an analytical apparent activation energy;
4. process maps with a Scheil-based N_2 porosity criterion and a sensitivity analysis showing that dissolved oxygen is a controllable variable of similar importance to the remelt fraction.

Section 2 presents the model, Section 3 the inputs, implementation and verification, and Section 4 the results and discussion, including five predictions that can discriminate experimentally between the kinetically limited and equilibrium regimes. Conclusions are given in Section 5.

2 Model formulation

2.1 Model structure and assumptions

Figure 2 shows the model framework. Nitrogen enters the deposit through two sequential routes: absorption by powder particles during flight and absorption at the melt-pool free surface. The melt-pool stage is then repeated as previously deposited material is partially remelted by subsequent passes. The following assumptions are made.

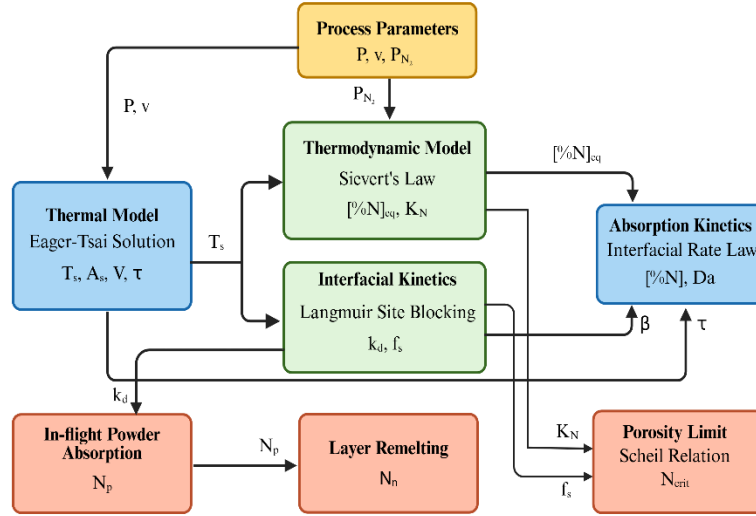


Fig. 2 Architecture of the coupled model. The thermal model provides the melt-pool quantities required by the thermodynamic and kinetic calculations; these are combined in the reversible nitrogen rate law and then applied to powder exposure, layer-to-layer remelting and the porosity screen

A1. The temperature field is quasi-steady in a frame moving with the laser beam. Heat transfer is by conduction with an effective thermal conductivity that accounts approximately for Marangoni convection.

A2. The melt pool is well mixed. Marangoni turnover times are of order 1–10 ms compared with a pool lifetime $\tau \approx 170$ ms, so composition gradients in the liquid are neglected.

A3. The gas above the pool contains molecular N_2 without a significant monatomic contribution from plasma dissociation [16].

A4. Dissolved nitrogen remains below about 0.1 wt%, so f_N is evaluated at the nominal alloy composition and nitride formation in the liquid is neglected.

A5. Gas-side transport is not rate limiting. With an N_2 diffusivity of $5 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$, a 2 mm pool length scale, $Sh \approx 4$ and a film temperature of 1500 K, the gas-phase supply flux is about 60 times the peak nitrogen consumption flux at the baseline condition of Section 4.

A6. Evaporative losses of Mn and Cr and the resulting changes in nitrogen activity are neglected.

A7. Carbide and nitride precipitation during layer reheating is not modeled. At the carbon level of AISI 316, $M_{23}C_6$ precipitation in the approximate 700–1100 K range is possible during repeated thermal cycling and would remove chromium from solid solution; the model tracks bulk dissolved nitrogen only.

2.2 Thermal model

The thermal field is calculated with the Eagar–Tsai solution for a Gaussian heat source of absorbed power ηP , distribution parameter σ and travel speed v on a semi-infinite body [19] (Fig. 3):

$$T = T_0 + \frac{2\eta P}{\rho c_p (4\pi\alpha)^{3/2}} I \quad (1)$$

$$I = \int_0^\infty \frac{G(t')}{t'^{1/2}(t' + t_\sigma)} dt' \quad (2)$$

$$G = \exp \left\{ -\frac{(\xi + vt')^2 + y^2}{4\alpha(t' + t_\sigma)} - \frac{z^2}{4\alpha t'} \right\} \quad (3)$$

where $\xi = x - vt$ is the moving-frame coordinate, $\alpha = k/\rho c_p$ is the thermal diffusivity and $t_\sigma = \sigma^2/2\alpha$ is the time constant associated with the source width. As $\sigma \rightarrow 0$ the solution reduces to the Rosenthal moving point source [20], which is used for verification in Section 3.3.

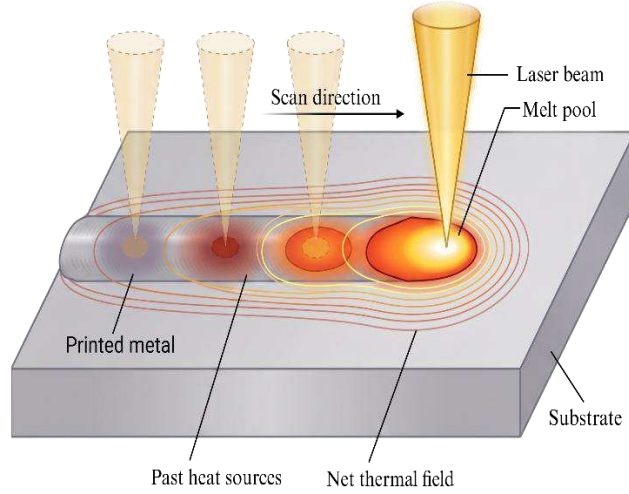


Fig. 3 Superposition of heat pulses from the moving laser: past and present pulses integrate to give the local temperature, Eqs. (1)-(3)

The temperature field is thresholded at the liquidus temperature. On the free surface $z = 0$ we define $\Omega_s = \{(x, y): T(x, y, 0) \geq T_L\}$ and $A_s = \iint_{\Omega_s} dA$. The chemistry model then uses A_s , the melt volume V , the pool length L , the residence time $\tau = L/v$ and the area-averaged free-surface temperature

$$\bar{T}_s = \frac{1}{A_s} \iint_{\Omega_s} T(x, y, 0) dA \quad (4)$$

The area average is used rather than the peak temperature because both the solubility and the interfacial rate constant depend on conditions over the whole exchanging surface.

2.3 Nitrogen solubility

Nitrogen dissolution, $N_2(g) \rightleftharpoons 2[N]$, follows Sieverts' law (Fig. 4):

$$[\%N]_{eq} = \frac{K_N(T)}{f_N} \sqrt{p_{N_2}} \quad (5)$$

$$\log_{10} K_N = -\frac{247}{T} - 1.22 \quad (6)$$

Equation (6) gives 0.0445 wt% N for pure liquid iron at 1873 K and 1 atm, consistent with the classical measurements of Pehlke and Elliott [21] and the pure-iron term of Wada and Pehlke [22]. In Eqs. (5)–(8), p_{N_2} is in atm, consistent with the empirical definition of K_N . The activity coefficient is calculated with first-order Wagner interaction parameters tabulated at 1873 K [23, 24] and scaled with temperature following Varbai and Májlínger [15]:

$$\log_{10} f_N(T) = \left(\frac{2538}{T} - 0.355 \right) \sum_i e_N^i [\%i] \quad (7)$$

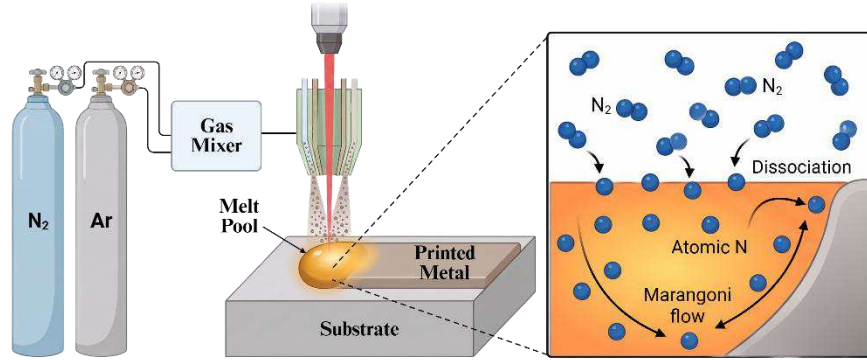


Fig. 4 Dissociation of N_2 at the melt pool surface into the dissolved atomic nitrogen

The temperature-scaling term equals unity at 1873 K, so the tabulated parameters are used directly there: $e_N^i = -0.047$ (Cr), $+0.010$ (Ni), -0.011 (Mo), -0.020 (Mn), $+0.047$ (Si), $+0.130$ (C) and $+0.007$ (S) [23, 24]. For the reference chemistry of Table 1, $f_N = 0.182$ at 1873 K, with chromium providing the dominant contribution, and the solubility at 1 atm is 0.244 wt%, 5.5 times that of pure iron. Substituting Eqs. (6) and (7) into Eq. (5) gives

$$\log_{10} [\%N]_{eq} = \frac{A}{T} + B + 1/2 \log_{10} p_{N_2}, \quad A = 1631K, B = -1.483 \quad (8)$$

where $A = -247 - 2538 \sum_i e_N^i [\%i]$ and $B = -1.22 + 0.355 \sum_i e_N^i [\%i]$ are evaluated for the chemistry of Table 1. Because $A > 0$, the equilibrium ceiling falls with melt superheat: the temperature dependence of the Cr–N interaction outweighs that of K_N . The coefficients must be recalculated for lot-specific chemistry.

2.4 Absorption kinetics

Under assumption A3, absorption and desorption occur through the dissociative reaction $2[N] \rightleftharpoons N_2(g)$ at the free surface (Fig. 5) [17, 18]. The reverse reaction is second order in dissolved nitrogen. Writing the net transfer as a reversible lumped interfacial rate law, with the forward pressure dependence embedded through N_{eq}^2 , gives the nitrogen mass balance of the pool:

$$\frac{d[\%N]}{dt} = \beta ([\%N]_{eq}^2 - [\%N]^2) \quad (9)$$

$$\beta = \frac{100A_s k_d}{\rho V} \quad (10)$$

The factor 100 converts mass fraction to weight percent, so β has units of $\text{s}^{-1} (\text{wt}\%)^{-1}$ when k_d is in $\text{kg m}^{-2} \text{s}^{-1} (\text{wt}\%)^{-2}$. The interfacial rate constant follows the Arrhenius relation used for stainless steel weld pools, with Langmuir site blocking by the surface-active elements [14, 15, 17]:

$$k_d = \chi \cdot \frac{10^{(-6340/T+1.85)}}{1 + 130f_S[\%S] + 260[\%O]} \quad (11)$$

$$\log_{10}f_S = [\%Cr] \left(-\frac{94.2}{T} + 0.040 \right) \quad (12)$$

Equation (11) is an empirical dimensional correlation in which $[\%S]$ and $[\%O]$ are entered in wt%. The dimensionless factor χ lumps effects not resolved explicitly: Marangoni surface renewal, differences between the real free surface and the flat-cap approximation, and the scatter among published interfacial rate constants. It is the only adjustable kinetic parameter in the formulation. The oxygen term is a provisional extension ($\delta_O = 0$ at baseline, $\delta_O = 1$ in the oxygen sensitivity study), because its original pressure-based formulation still requires reconciliation; oxygen is known to retard nitrogen exchange at stainless steel weld-pool surfaces [25].

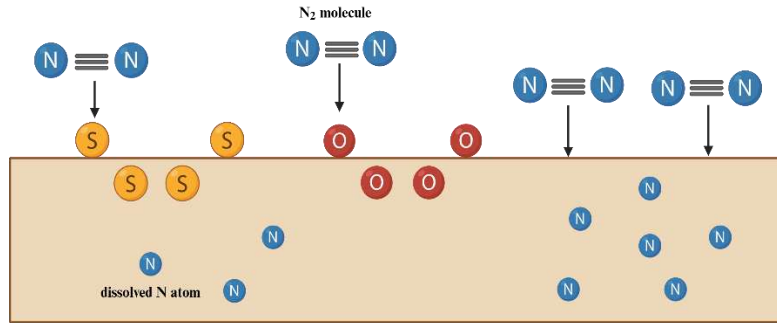


Fig. 5 Absorption kinetics of N_2 at the melt-pool surface: site blocking by surface-active sulfur and oxide films

The oxygen term requires the dissolved oxygen content of the melt. Inert-gas-fusion analysis of powder returns total oxygen, most of which resides in Cr, Mn and Si oxide films on the particle surfaces rather than in solution. The dissolved fraction is set by equilibrium with those oxides and is generally lower than the measured total. When the oxygen term is active it can dominate the site-blocking denominator, so the assumed $[\%O]$ and a calibrated χ are not independent; a calibrated χ should always be reported together with the dissolved oxygen level assumed in obtaining it.

Equation (9) can be integrated analytically. For $[\%N] = N_0$ at $t = 0$,

$$N(t) = N_{eq} \frac{N_0 + N_{eq} \tanh(\beta N_{eq} t)}{N_{eq} + N_0 \tanh(\beta N_{eq} t)} \quad (13)$$

The argument of the hyperbolic tangent defines the controlling dimensionless groups. The Damköhler number is the ratio of the pool residence time to the characteristic absorption time $t_k = 1/(\beta N_{eq})$:

$$Da = \beta N_{eq} \tau = \frac{\tau}{t_k}, \quad r_0 = \frac{N_0}{N_{eq}} \quad (14)$$

The normalized pass-exit concentration then depends on Da and r_0 only:

$$\frac{N_f}{N_{eq}} = \frac{r_0 + \tanh Da}{1 + r_0 \tanh Da} \quad (15)$$

Equation (15) is the central analytical result. For $Da \gg 1$ the pool approaches equilibrium and thermodynamics controls the nitrogen content. For $Da \ll 1$ it reduces to

$$\Delta N \approx \beta N_{eq}^2 \tau (1 - r_0^2) = \frac{100 A_s k_d N_{eq}^2 \tau (1 - r_0^2)}{\rho V} \quad (16)$$

In the kinetically limited regime, the pickup is therefore proportional to $k_d N_{eq}^2$ rather than to N_{eq} . Two consequences follow. First, because $N_{eq}^2 \propto p_{N_2}$, the pickup becomes linear in N_2 partial pressure instead of following Sieverts' square root. Second, from Eqs. (8) and (11) and neglecting site blocking, $k_d N_{eq}^2 \propto 10^{-3078/T}$, which increases with temperature even though N_{eq} decreases. With sulfur (and oxygen) site blocking, the additive denominator of Eq. (11) prevents reduction to a single Arrhenius exponent, so a local apparent activation energy is defined from the logarithmic temperature sensitivity:

$$E_{app} = -R \frac{d \ln(k_d N_{eq}^2)}{d(1/T)} \quad (17)$$

Without site blocking $E_{app} = 2.303R \times 3078K = 58.9 \text{ kJ mol}^{-1}$; with sulfur blocking at the baseline $\bar{T}_s$ it is 50.5 kJ mol^{-1} (Section 4.4). The positive value implies a sign change in temperature sensitivity as the process crosses $Da \approx 1$, which provides a direct experimental test of the kinetic formulation.

2.5 Powder absorption

For a spherical particle of diameter d_p , $A/V = 6/d_p$, and Eq. (10) becomes

$$\beta_p = 100 \left(\frac{6}{\rho d_p} \right) k_d(T_p) \quad (18)$$

For $d_p = 89.4 \mu\text{m}$ the particle surface-to-volume ratio is $6.7 \times 10^4 \text{ m}^{-1}$, 22 times the pool value $A_s/V = 3.1 \times 10^3 \text{ m}^{-1}$. However, the molten exposure time of a particle is only a few milliseconds, compared with about 170 ms in the pool, and the particle is cooler. Equation (13) is applied with β_p , T_p and the exposure time t_f , and the resulting powder nitrogen content N_p becomes the inlet composition for the pool stage.

2.6 Layer accumulation

Each pass is represented by an effective remelt/dilution mass fraction λ , defined as the fraction of the inlet pool mass contributed by previously deposited material (Fig. 6). The inlet concentration for pass n is

$$N_n(0) = (1 - \lambda)N_p + \lambda N_{n-1} \quad (19)$$

with N_0 replaced by the substrate nitrogen content for the first pass, and $N_n = \mathcal{K}(N_n(0))$ is the single-pass map of Eq. (13). For $\text{Da} \ll 1$, each pass adds an approximately constant increment ΔN_1 , giving the fixed point

$$N_\infty \approx N_p + \frac{\Delta N_1}{1 - \lambda} \quad (20)$$

Repeated remelting increases pickup because previously deposited material is exposed to the reactive gas more than once. The amplification factor $(1 - \lambda)^{-1}$ is moderate for typical DED dilutions: 1.5 at $\lambda = 0.33$ and 4 at $\lambda = 0.75$. Equation (20) is only an estimate; all results below use the exact recursion of Eqs. (13) and (19).

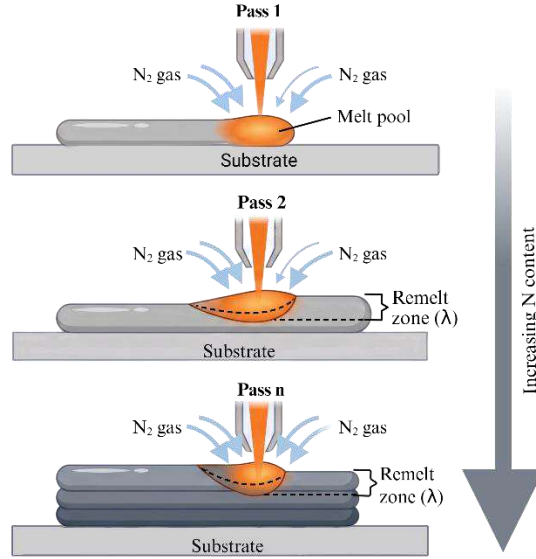


Fig. 6 Layer accumulation: each new pass remelts a fraction λ of the previously deposited material, which is re-exposed to the reactive gas

2.7 Porosity criterion

The upper useful nitrogen level may be set by gas porosity during solidification rather than by liquid solubility [26]. As solidification proceeds, nitrogen is rejected into the interdendritic liquid according to the Scheil relation

$$C_L(f_s) = C_0(1 - f_s)^{k_p^{-1}} \quad (21)$$

where k_p is the nitrogen partition coefficient. The N_2 pressure supported by the enriched liquid follows from inverting Sieverts' law at the solidus temperature:

$$p_{N_2}^* = \left[\frac{f_N(T_s)C_L}{K_N(T_s)} \right]^2 \quad (22)$$

A pore can form when this pressure exceeds the ambient pressure plus the capillary back-pressure on a pore nucleus of radius r (Fig. 7). Because pressures in Eqs. (5) and (22) are in atm, the capillary term is converted from Pa:

$$p_{back} = p_{amb} + \frac{2\gamma_{lv}}{101325r} \quad (23)$$

Setting $p^* = p_{back}$ at a selected terminal solid fraction f_s^* gives the critical bulk nitrogen concentration

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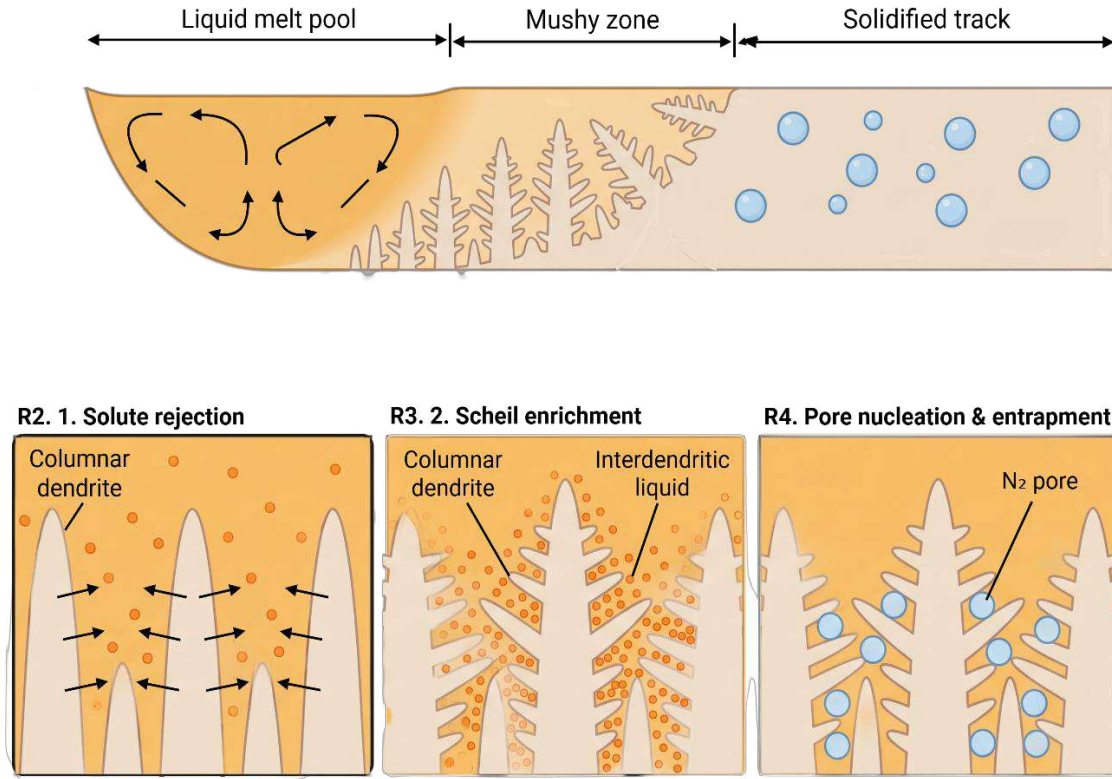
$$N_{crit} = \left[\frac{\sqrt{p_{back}} K_N(T_S)}{f_N(T_S)} \right] (1 - f_s^*)^{1-k_p} \quad (24)$$

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Equations (21)–(24) treat N_2 as the only gas-forming species. Carbon and oxygen are also rejected into the interdendritic liquid, so CO formation is a second route to porosity; it is neglected here, and its importance grows with feedstock carbon content.



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Fig. 7 Pore formation mechanism: Scheil enrichment of the interdendritic liquid raises the supported N_2 pressure until it exceeds the ambient plus capillary back-pressure

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3 Inputs, implementation and verification

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3.1 Reference inputs

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The reference alloy is NIST SRM 1155a (AISI 316) [27] (Table 1); the same material underlies the liquid thermophysical property measurements of Pichler et al. [28, 29]. Applying 316L properties to 316 is an approximation. Material, process, powder, layer and porosity inputs are listed in Table 2. The baseline process (800 W , 10 mm s^{-1}) is a representative blown-powder laser DED condition. Parameters marked “assumed” are not measured and are covered by the sensitivity study.

247 **Table 1** Reference chemistry (NIST SRM 1155a, AISI 316) used in all calculations, wt%

Cr	Ni	Mo	Mn	Si	C	S	N ₀ ^a	O ^b	Fe
17.803	12.471	2.188	1.593	0.521	0.026	0.0020	0.0428	0.0010	bal.

248 ^a Feedstock and substrate nitrogen adopted from the reference material; not a powder-lot measurement.

249 ^b Assumed dissolved oxygen, used only in the provisional oxygen-blocking extension.

250 **Table 2** Baseline model inputs

Symbol	Value	Unit	Basis
ρ	6827	kg m ⁻³	Liquid at 1873 K [28]
c_p	847	J kg ⁻¹ K ⁻¹	Liquid [28]
k	30.36	W m ⁻¹ K ⁻¹	Liquid at 1873 K [29]
T_L, T_S	1708, 1675	K	[29]
P, v	800, 0.010	W, m s ⁻¹	Representative condition
η	0.35	–	Assumed
σ	3.0×10^{-4}	m	Beam diameter $4\sigma = 1.2$ mm (assumed)
T_0, p_{amb}	300, 1.0	K, atm	Assumed
p_{N_2}	0.50	atm	N ₂ mole fraction 0.5 (assumed)
χ	1.0	–	Uncalibrated
d_p	8.94×10^{-5}	m	Median particle size (assumed)
T_p, t_f	1800, 0.003	K, s	Effective molten exposure (assumed)
λ	0.33	–	Assumed
k_p	0.50	–	Assumed screening value
γ_{lv}	1.59	N m ⁻¹	Near-liquidus value [29]
r, f_s^*	$1.0 \times 10^{-5}, 0.95$	m, –	Assumed

251 3.2 Numerical implementation

252 The model was implemented in Python (NumPy/SciPy). The Eagar–Tsai integral is evaluated by adaptive
253 vector quadrature after the substitutions $t' = t_\sigma u^2$ and $u = s/(1 - s)$, which remove the square-root
254 singularity at $t' = 0$ and map the infinite interval to $[0,1]$ (relative tolerance 10^{-5} , 200 subdivisions). The
255 field is sampled on a $150 \times 100 \times 50$ cell-centered grid with 0.1 mm spacing over $-10 \leq \xi \leq 5$ mm,
256 $|y| \leq 5$ mm and $0 \leq z \leq 5$ mm, with the free surface evaluated separately at $z = 0$. Area and volume are

midpoint sums over cells at or above T_L ; a run is rejected if molten cells touch a side or bottom boundary. Passes are iterated until the successive change in exit nitrogen falls below 10^{-5} wt% (0.1 ppm), with ten passes always reported. Sensitivity studies vary one input at a time about the baseline, and the process map is a 45-case Cartesian grid over $P \in \{600, 800, 1000\}$ W, $v \in \{5, 10, 15\}$ mm s $^{-1}$ and $p_{N_2} \in \{0, 0.25, 0.5, 0.75, 1.0\}$ atm.

3.3 Verification

Three checks were made (Fig. 8). First, with σ reduced to 2 μ m, the Eagar–Tsai field reproduces the analytical Rosenthal point-source solution [20] along the centerline at depths of 0.5 and 1.0 mm with a maximum relative error in temperature rise of 2.2×10^{-5} (Fig. 8a). Second, direct numerical integration of Eq. (9) agrees with the closed-form solution, Eq. (13), to within 1.4×10^{-13} wt% over five pool lifetimes (Fig. 8b). Third, halving the grid spacing to 0.05 mm changed A_s by +0.5%, V by -0.2% , $\bar{T}_s$ by -0.2% and the first-pass and steady-state nitrogen by -0.4% and -0.5% ; the pool length changed by one cell (-2.9%). Tightening the quadrature tolerance tenfold changed no reported quantity by more than 10^{-5} relative. The baseline discretization is therefore adequate for the conclusions drawn here.

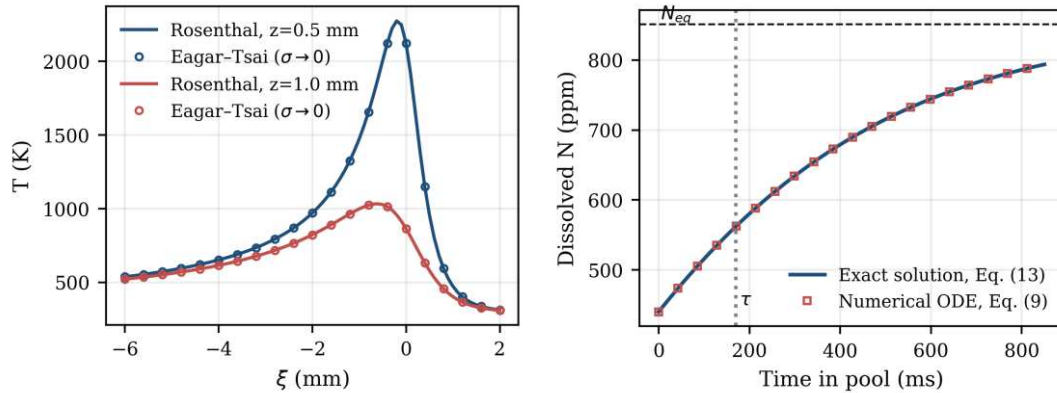


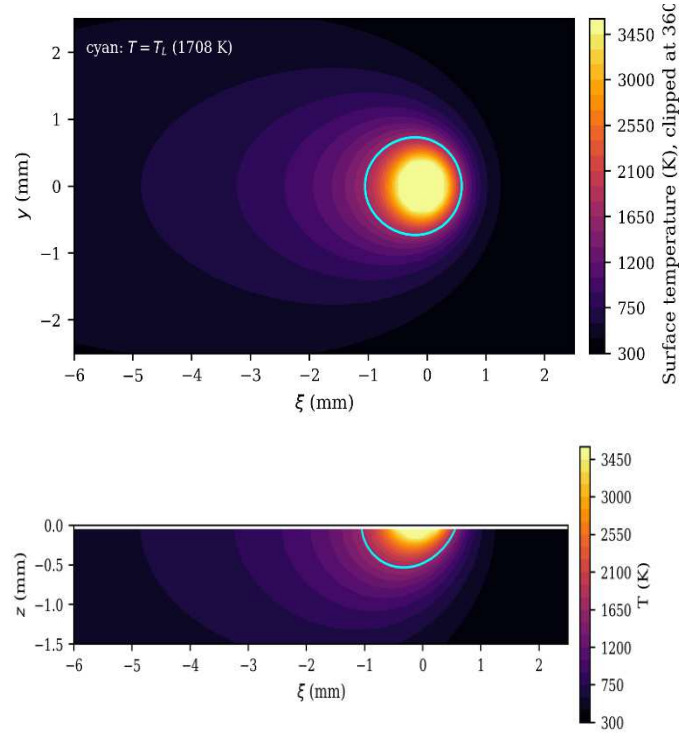
Fig. 8 Verification. (a) Eagar–Tsai solution with $\sigma = 2$ μ m against the Rosenthal point-source solution along $y = 0$ at two depths. (b) Exact solution, Eq. (13), against numerical integration of Eq. (9) for the baseline first pass

4 Results and discussion

4.1 Thermal state of the melt pool

Figure 9 shows the baseline surface and longitudinal temperature fields. The liquidus isotherm encloses a pool 1.7 mm long, 1.4 mm wide and 0.5 mm deep, with $A_s = 1.88$ mm ^{2} , $V = 0.61$ mm ^{3} ($A_s/V = 3.08 \times 10^3$ m $^{-1}$) and a residence time $\tau = 0.17$ s (Table 3). The area-averaged surface temperature is

278 $\bar{T}_s = 2897$ K. The peak sampled temperature, 5348 K, is unphysical because the conduction-only solution
 279 has no latent heat, evaporation or convective redistribution; this is why the area average rather than the
 280 peak is passed to the chemistry model. Even so, $\bar{T}_s$ is probably an upper estimate, and a convective pool
 281 would be cooler and larger. Over the process map, $\bar{T}_s$ ranges from 2536 to 3145 K.



282 **Fig. 9** Baseline Eagar–Tsai temperature field (800 W, 10 mm s^{−1}, $\eta = 0.35$): free surface $z = 0$ (top) and
 283 longitudinal section $y = 0$ (bottom). The cyan contour is the liquidus (1708 K); the color scale is clipped at 3600 K

284 4.2 Baseline nitrogen balance

285 Table 3 summarizes the baseline chemistry. At $\bar{T}_s = 2897$ K and 0.5 atm N₂, $f_N = 0.411$ and the pool
 286 ceiling is $N_{eq} = 0.0851$ wt%, much lower than the 1873 K value because the ceiling falls with superheat
 287 (Eq. (8)). The interfacial rate constant is $k_d = 0.339$ kg m^{−2} s^{−1} (wt%)^{−2} and $\beta = 15.3$ s^{−1} (wt%)^{−1},
 288 giving a characteristic absorption time $t_k = 0.77$ s, 4.5 times the pool lifetime, and $Da = 0.22$.

289 During its 3 ms molten exposure in flight, the powder rises from 428 to 446 ppm; its local ceiling at 1800
 290 K (0.188 wt%) is high, but the exposure is short. The first pass enters the pool at 440 ppm (including
 291 substrate dilution) and leaves at 562 ppm, i.e., at $N_f/N_{eq} = 0.66$. The small-Da approximation, Eq. (16),
 292 predicts a first-pass increment of 138 ppm against 122 ppm from the exact solution; the 13% overestimate
 293 shows that $Da = 0.22$ is already outside the strict $Da \ll 1$ limit. Repeated remelting raises the deposit to a
 294 converged 607 ppm after seven passes (Fig. 10); the fixed-point estimate of Eq. (20) gives 650 ppm

(+7%), because at $Da = 0.22$ the increment per pass already shrinks as N approaches N_{eq} . Of the total pickup of 179 ppm, the powder stage contributes about 10%, and remelting amplifies the first-pass pickup by a factor of 1.33, below the $(1 - \lambda)^{-1} = 1.49$ limit.

The steady-state deposit reaches 71% of the pool ceiling, but this figure flatters the process because the feedstock already supplies 50% of it. Measured relative to the starting gap, the deposit closes only 42% of the distance between N_0 and N_{eq} . For the uncalibrated rate constant, the baseline process is therefore kinetically limited, consistent with the modest pickups reported for laser processing under nitrogen [9, 10].

Table 3 Baseline model outputs (800 W , 10 mm s^{-1} , 0.5 atm N_2 , $\chi = 1$, sulfur-only site blocking)

Quantity	Value	Quantity	Value
$L \times W \times D$ (mm)	$1.7 \times 1.4 \times 0.5$	N_{eq} at $\bar{T}_S$ (wt%)	0.0851
A_s (mm ²)	1.88	k_d (kg m ⁻² s ⁻¹ wt% ⁻²)	0.339
V (mm ³)	0.61	β (s ⁻¹ wt% ⁻¹)	15.3
τ (s)	0.17	Da (–)	0.221
$\bar{T}_S$ (K)	2897	E_{app} (kJ mol ⁻¹)	50.5
K_N (wt% atm ^{-1/2})	0.0495	N_p , powder exit (wt%)	0.0446
f_N, f_S (–)	0.411, 1.359	First-pass exit (wt%)	0.0562
$t_k = 1/\beta N_{eq}$ (s)	0.77	Steady state N_∞ (wt%)	0.0607
C_L at f_S^* (wt%)	0.271	Eq. (20) estimate (wt%)	0.0650
p^*, p_{back} (atm)	0.77, 4.14	N_{crit} (wt%)	0.141

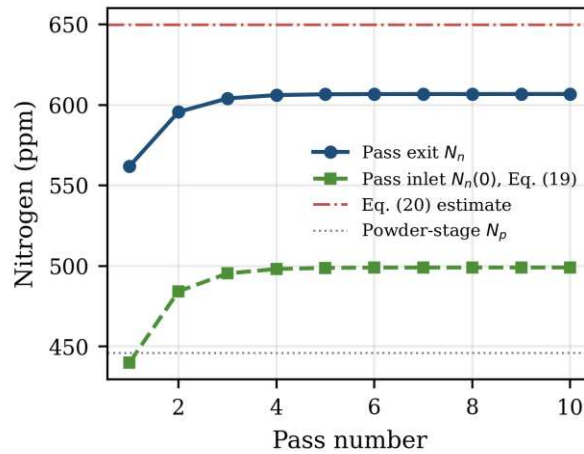


Fig. 10 Pass-by-pass nitrogen at baseline: inlet (Eq. (19)) and exit concentrations converge to 607 ppm after seven passes; the fixed-point estimate (Eq. (20)) overpredicts by 7%

4.3 Kinetic regime and the role of the rate multiplier

Figure 11a places the baseline on the universal curve of Eq. (15). With $r_0 \approx 0.52$, the baseline ($\chi = 1$) lies on the rising part of the curve; $\chi = 0.1$ ($Da = 0.022$) leaves the pool almost at its inlet concentration, and $\chi = 10$ ($Da = 2.2$) brings it to 99% of equilibrium. Figure 11b shows the corresponding steady-state deposit nitrogen over four decades of χ . The transition to equilibrium control ($Da = 1$) occurs at $\chi \approx 4.5$, and the deposit saturates at the pool ceiling of 851 ppm for $\chi \gtrsim 10$. The ceiling remains below the porosity threshold for all χ at this condition.

Because χ shifts the process across the whole regime map, its calibration is the main experimental requirement of the model. A single measurement of deposit nitrogen at one condition fixes χ (for a stated dissolved oxygen level), after which the model is predictive. The fiber-laser welding result of a roughly 130 ppm pickup under pure N_2 [10] is of the same order as the 179 ppm predicted here at $\chi = 1$, which suggests that χ is of order unity for laser processing; a DED-specific calibration is nevertheless required.

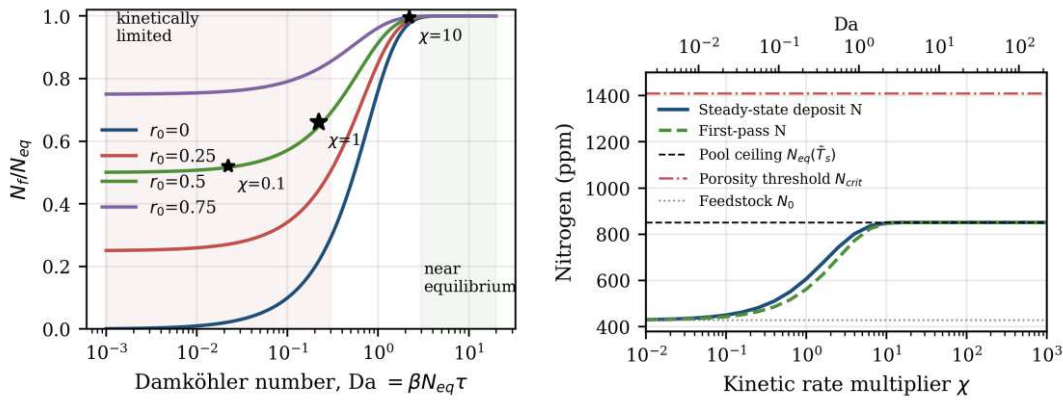


Fig. 11 Kinetic regime. (a) Normalized pass-exit nitrogen, Eq. (15), against Da for several r_0 ; stars mark the baseline thermal state with $\chi = 0.1$, 1 and 10. (b) First-pass and steady-state deposit nitrogen against χ (bottom axis) and Da (top axis), compared with the pool ceiling, the feedstock level and the porosity threshold

4.4 Reversal of temperature sensitivity

Figure 12a isolates the effect of pool temperature by holding the baseline geometry and lifetime fixed and varying $\bar{T}_s$ from 1900 to 3400 K. In the equilibrium limit ($\chi = 10^3$), the pass-exit nitrogen follows the ceiling and falls from 1680 to 702 ppm. In the kinetic limit ($\chi = 0.1$), the single-pass pickup instead rises from 5.6 to 15.2 ppm, a factor of 2.7, because $k_d N_{eq}^2$ increases with temperature. At $\chi = 1$, the pickup rises from 55 to 122 ppm and then levels off near 3100–3200 K, where the falling ceiling begins to offset faster kinetics. The local apparent activation energy (Fig. 12b) decreases from 54.1 to 49.3 kJ mol⁻¹ over

this range as sulfur site blocking strengthens with temperature, and equals 50.5 kJ mol^{-1} at the baseline. It stays below the unblocked value of 58.9 kJ mol^{-1} throughout.

This reversal has a practical consequence: at the baseline the process sits close to the broad maximum of the $\chi = 1$ curve, where temperature sensitivity nearly vanishes. Laser power and absorptivity, which act mainly through $\bar{T}_s$, are therefore weak levers at $\chi \approx 1$ (Section 4.5). Measuring deposit nitrogen as a function of superheat at a fixed residence time would reveal which side of the crossover a real process occupies.

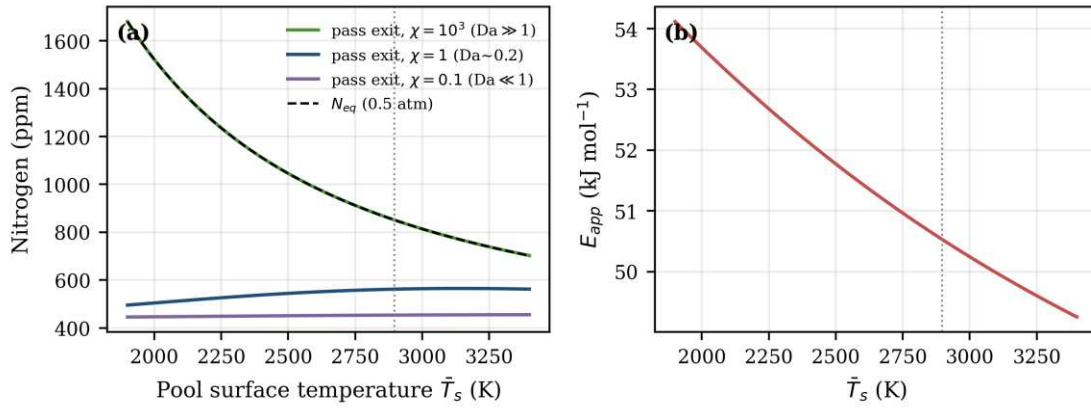


Fig. 12 Temperature sensitivity at fixed baseline geometry and lifetime. (a) Pass-exit nitrogen against $\bar{T}_s$ in the equilibrium ($\chi = 10^3$), intermediate ($\chi = 1$) and kinetic ($\chi = 0.1$) regimes, with the equilibrium ceiling. (b) Local apparent activation energy, Eq. (17). The dotted line marks the baseline $\bar{T}_s$

4.5 Process maps and sensitivity

Figure 13 shows the steady-state deposit nitrogen over the 45-case process map, and Fig. 14 resolves the power–speed dependence at 0.5 and 1.0 atm. For $p_{N_2} \geq 0.25$ atm, the accessible range is 473–996 ppm, with Da between 0.08 and 0.62; no case reaches equilibrium control, and none exceeds the porosity threshold. Under pure argon ($p_{N_2} = 0$), the deposit loses nitrogen by desorption and ends at 320–400 ppm, below the 428 ppm feedstock level.

Three trends stand out. First, at 10 mm s^{-1} and 800 W the increments per 0.25 atm step are 121, 117, 112 and 108 ppm, so the deposit nitrogen is nearly linear in p_{N_2} , as Eq. (16) predicts for $\text{Da} < 1$; at equilibrium the same steps would follow $\sqrt{p_{N_2}}$, with the first increment more than three times the last. Second, travel speed is the strongest process-side lever after p_{N_2} : slowing from 15 to 5 mm s^{-1} lengthens τ , raises Da from 0.15 to 0.39 and increases the deposit nitrogen from 568 to 672 ppm at 800 W and 0.5 atm . Third, at fixed speed, higher power slightly lowers the deposit nitrogen (e.g., 697 , 671 and 651 ppm

at 600, 800 and 1000 W and 5 mm s^{-1}), because the hotter pool has a lower ceiling and a smaller surface-to-volume ratio (A_s/V falls from 3.5×10^3 to $1.9 \times 10^3 \text{ m}^{-1}$ between 600 and 1000 W at 5 mm s^{-1} , while τ rises from 0.28 to 0.42 s); together these outweigh the faster kinetics near the crossover identified in Section 4.4.

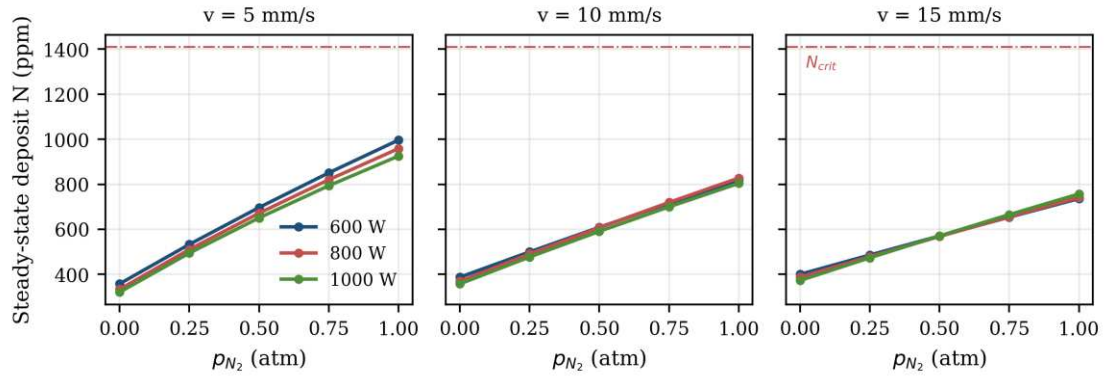


Fig. 13 Steady-state deposit nitrogen against N_2 partial pressure for three powers at each travel speed. The dash-dotted line is the baseline porosity threshold $N_{crit} = 1410 \text{ ppm}$

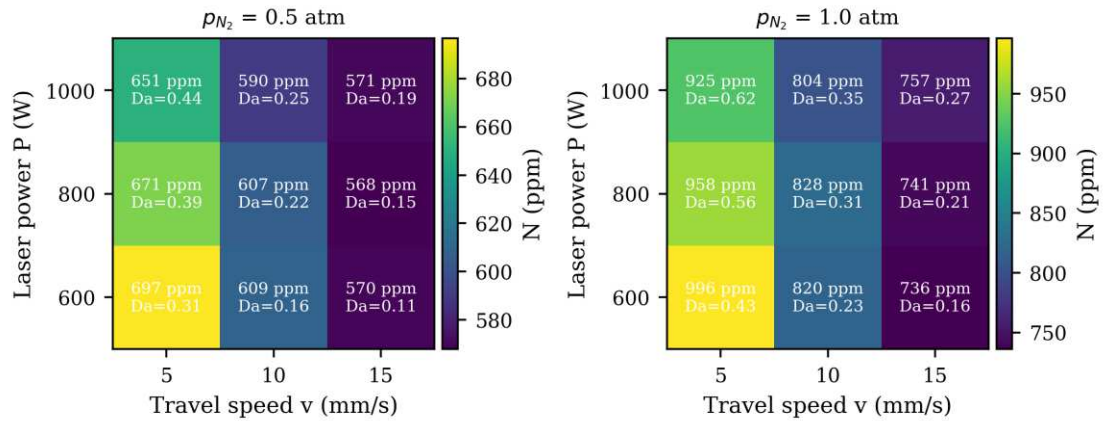


Fig. 14 Steady-state deposit nitrogen and Damköhler number over laser power and travel speed at $p_{N_2} = 0.5 \text{ atm}$ (left) and 1.0 atm (right)

Fig. 15 ranks the one-at-a-time sensitivities of the steady-state nitrogen. N_2 partial pressure ($-238/+221 \text{ ppm}$ over $0-1 \text{ atm}$) and the rate multiplier ($-157/+242 \text{ ppm}$ over $\chi = 0.1-10$) dominate. They are followed by the remelt fraction ($-40/+107 \text{ ppm}$ over $\lambda = 0-0.75$) and dissolved oxygen: activating oxygen site blocking at 10, 30 and 100 ppm dissolved O lowers the deposit nitrogen from 607 to 584, 552 and 499 ppm and Da from 0.22 to 0.076. Dissolved oxygen is therefore a controllable variable of the same order as the remelt fraction, which argues for low-oxygen powder and tight shielding when nitrogen

pickup is the goal. Travel speed (−39/+65 ppm) follows; absorptivity, power, powder exposure time and powder temperature each change the result by less than 20 ppm.

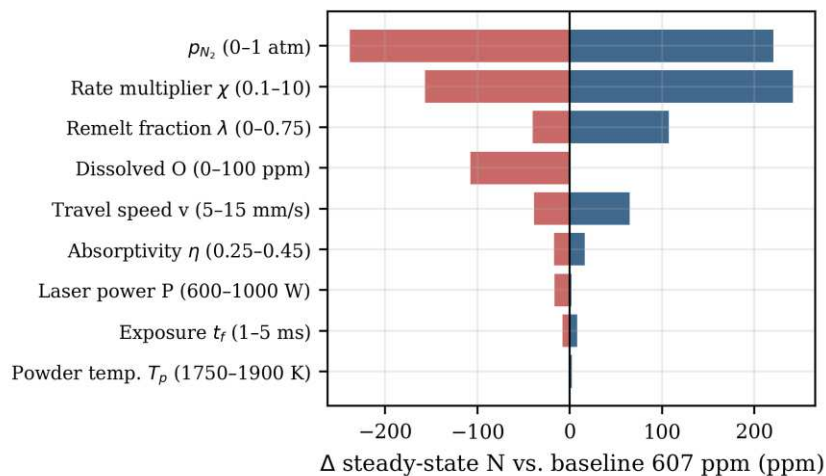


Fig. 15 One-at-a-time sensitivity of the steady-state deposit nitrogen about the 607 ppm baseline. Ranges are those of Table 2 variations; dissolved oxygen activates the provisional oxygen-blocking term

4.6 Porosity screening

At baseline, Scheil enrichment to $f_s^* = 0.95$ raises the interdendritic liquid to 0.271 wt% N, which supports an N_2 pressure of 0.77 atm against a back-pressure of 4.14 atm for a 10 μm nucleus; the critical bulk concentration is $N_{crit} = 0.141$ wt%, and the deposit is at 43% of it. The criterion is, however, sensitive to the poorly known solidification parameters (Fig. 16). Across $k_p = 0.3$ –0.7 and $r = 5$ –50 μm , N_{crit} spans 486 to 3403 ppm. Only the most pessimistic combination, $k_p = 0.3$ with $r = 50$ μm , places the baseline deposit (607 ppm) above the threshold. Because N_{crit} falls steeply as $f_s^* \rightarrow 1$, the choice of terminal solid fraction matters as much as k_p . Measured partition coefficients for the actual solidification mode (primary austenite or primary ferrite) are needed before the porosity limit can be used quantitatively; within the present ranges, porosity is unlikely to limit in-situ nitrogen alloying of 316 at $\chi \approx 1$.

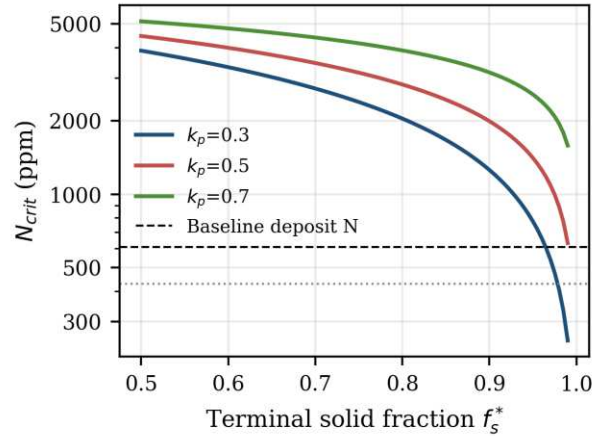


Fig. 16 Critical bulk nitrogen for N_2 porosity, Eq. (24), against terminal solid fraction for three partition coefficients ($r = 10 \mu\text{m}$), compared with the baseline deposit nitrogen (dashed) and the feedstock level (dotted)

4.7 Experimentally testable predictions

The model yields five predictions that can distinguish the kinetically limited regime from the equilibrium regime in a single DED campaign with bulk nitrogen analysis:

1. *Pressure scaling.* For $Da < 1$, deposit nitrogen increases almost linearly with p_{N_2} ; for $Da \gg 1$ it follows $\sqrt{p_{N_2}}$.
2. *Residence-time scaling.* At fixed power, pickup increases as travel speed decreases (longer τ) in the kinetic regime, but becomes insensitive to speed near equilibrium.
3. *Temperature sign.* At fixed τ , increasing superheat raises pickup when $Da \ll 1$ and lowers it when $Da \gg 1$ (Fig. 12).
4. *Surface-active elements.* Sulfur and dissolved oxygen strongly reduce pickup in the kinetic regime but affect the equilibrium ceiling only weakly through f_N .
5. *Argon baseline.* Under pure argon, deposits lose nitrogen relative to the feedstock, at a rate set by the same β that governs absorption.

Predictions 1 and 5 are the simplest to test because they require only a gas-composition series at one laser condition, and together they also calibrate χ .

4.8 Limitations

The conduction-only thermal model neglects latent heat, evaporation, radiation and Marangoni convection, so it overpredicts the peak temperature and $\bar{T}_S$; a convective pool would be cooler, which at

$\chi \approx 1$ would raise the ceiling and lower k_d with little net change in pickup (Section 4.4). Constant 316L liquid properties are applied to 316. The powder thermal history is represented by an effective temperature and exposure time rather than computed. The rate correlation, Eq. (11), was developed for weld pools; its oxygen term is tentative, and χ is uncalibrated. The porosity limit considers N_2 only and uses assumed solidification parameters. Solid-state precipitation during reheating and Mn/Cr evaporation are not modeled. All predicted nitrogen levels remain below 0.1 wt%, consistent with assumption A4. Nitride formation is neglected in the study

5 Conclusions

This study developed and verified a coupled thermal–thermodynamic–kinetic model of in-situ nitrogen alloying of AISI 316 during LDED. The main conclusions are as follows.

1. Treating absorption of molecular N_2 as reversible dissociative exchange gives an exact hyperbolic-tangent solution whose behavior is governed by a single Damköhler number, $Da = \beta N_{eq} \tau$, together with the initial ratio $r_0 = N_0/N_{eq}$.
2. For the reference condition (800 W, 10 mm s⁻¹, 0.5 atm N_2 , $\chi = 1$), the pool surface averages 2897 K, the ceiling is 0.085 wt%, $Da = 0.22$, and the steady-state deposit reaches 0.061 wt% N, closing only 42% of the gap between feedstock and equilibrium. Powder-in-flight absorption contributes about 10% of the pickup and remelting amplifies the first-pass pickup by a factor of 1.33.
3. In the kinetic regime, pickup scales almost linearly with N_2 partial pressure and increases with superheat (apparent activation energy 49–54 kJ mol⁻¹), opposite to the equilibrium trend. At $\chi \approx 1$ the process operates in the kinetically limited regime ($Da < 1$). Within the investigated parameter range, laser power has a relatively weak influence on nitrogen content, whereas travel speed and nitrogen partial pressure provide more effective process control.
4. Over 600–1000 W, 5–15 mm s⁻¹ and 0.25–1 atm N_2 , the predicted accessible range is 473–996 ppm, below the baseline Scheil-based porosity threshold of 1410 ppm. Pure-argon deposition is predicted to lower the nitrogen content below that of the feedstock.
5. Dissolved oxygen reduces pickup by up to 108 ppm over 0–100 ppm oxygen, that must be reported together with calibrated rate multiplier.
6. The rate multiplier χ is the only adjustable kinetic parameter. A gas-composition series at a single laser condition is sufficient to calibrate it and to test the predicted linear pressure scaling.

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Declarations

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Author contributions. **Md Al Amin Meia:** Conceptualization, Methodology, Mathematical modeling, Software, Validation, Formal analysis, Investigation, Data curation, Visualization, Writing—original draft, Writing—review & editing. **Shafin Ahmed:** Methodology, Programming, Investigation, Data Visualization, Formal analysis, Validation, Writing—review & editing. **Md Shahriar Forhad:** Methodology, Investigation, Formal analysis, Validation, Writing—review & editing. **Monsuru Ramoni:** Supervision, Project administration, Conceptualization, Writing—review & editing. **Jianzhi Li:** Supervision, Project administration, Conceptualization, Writing—review & editing.

Data availability. The model inputs and generated results supporting this study are available from the corresponding author on reasonable request.

Code availability. The Python implementation of the model and the scripts used to generate the figures are available from the corresponding author on reasonable request.

Appendix A Nomenclature

Table A1 Nomenclature

Symbol	Meaning (unit)	Symbol	Meaning (unit)
A_s	pool free-surface area (m^2)	p_{N_2}	N_2 partial pressure (atm)
c_p	specific heat ($\text{J kg}^{-1} \text{K}^{-1}$)	p^*	N_2 pressure supported by liquid (atm)
C_L	interdendritic liquid N (wt%)	p_{back}	ambient + capillary pressure (atm)
Da	Damköhler number, $\beta N_{eq} \tau$ (–)	PREN	%Cr + 3.3%Mo + 16%N (–)
d_p	powder diameter (m)	r	pore-nucleus radius (m)
e_N^i	Wagner parameter of i on N (wt% $^{-1}$)	r_0	N_0/N_{eq} (–)
E_{app}	apparent activation energy (J mol^{-1})	R	gas constant ($\text{J mol}^{-1} \text{K}^{-1}$)

Symbol	Meaning (unit)	Symbol	Meaning (unit)
f_N, f_S	activity coefficients of N, S (–)	T, T_0	local, ambient temperature (K)
f_S, f_S^*	solid fraction, terminal value (–)	T_L, T_S	liquidus, solidus (K)
k	thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)	$\bar{T}_S$	area-averaged surface temperature (K)
k_d	interfacial rate constant ($\text{kg m}^{-2} \text{s}^{-1} \text{wt}\%^{-2}$)	T_p	effective powder temperature (K)
k_p	N partition coefficient (–)	t_f, t_k	powder exposure, absorption time (s)
K_N	Sieverts constant ($\text{wt}\% \text{atm}^{-1/2}$)	t_σ	source-width time constant (s)
L, W, D	pool length, width, depth (m)	v, V	travel speed (m s^{-1}), pool volume (m^3)
N_0	feedstock nitrogen (wt%)	α	thermal diffusivity ($\text{m}^2 \text{s}^{-1}$)
N_{eq}	equilibrium ceiling (wt%)	β, β_p	rate coefficient, pool/powder ($\text{s}^{-1} \text{wt}\%^{-1}$)
N_{crit}	critical bulk N for porosity (wt%)	γ_{lv}	surface tension (N m^{-1})
N_f, N_∞	pass-exit, steady-state N (wt%)	η, σ	absorptivity (–), source width (m)
N_p	powder-stage nitrogen (wt%)	λ	remelt/dilution fraction (–)
P	laser power (W)	ξ, ρ	moving coordinate (m), density (kg m^{-3})
$[\%N]$	dissolved nitrogen (wt%)	τ, χ	pool lifetime L/v (s), rate multiplier (–)

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