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High-fidelity phase-field simulation of solid-state sintering enabled by Bayesian data assimilation using *in situ* electron tomography data

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ABSTRACT

Experimental observation methods for understanding industrially important solid-state sintering are essential for the development of new materials and devices. To experimentally characterize solid-state sintering, the limitations posed by the complexity of target materials, experimental equipment, and observation conditions must be overcome. Therefore, hybrid techniques for predicting sintering behavior based on experimental datasets and physics-based simulation models are highly sought after. Herein, we propose a new technique for combining a physics-based model and experimental observation results from solid-state sintering using a nonsequential Bayesian data assimilation (DA) method. The proposed technique assimilates experimental data—obtained using *in situ* electron tomography/scanning transmission electron microscopy—into the corresponding phase-field (PF) model to enable high-fidelity PF simulations by estimating multiple material parameters included in the PF model. This study demonstrates the inverse estimation of seven parameters, including temperature-dependent diffusion coefficients, from the time-series information on the morphology of sintered nanoparticles observed *in situ*. The estimated parameters provide the high-fidelity PF simulation to capture the observed solid-state sintering of copper nanoparticles. Thus, this study contributes to the construction of digital twins for solid-state sintering based on DA-integrated PF simulations and *in situ* observation datasets and deepens our understanding of the sintering process.

1. Introduction

Owing to the importance of solid-state sintering for the production of bulk polycrystalline materials, *in situ* observations using scanning/transmission electron microscopy have been widely used to elucidate the details of this process [1–4]. For example, the solid-state sintering of Cu particles has been observed *in situ* using scanning electron microscopy [1], that of Cu and alumina particles has been observed using electron and X-ray tomography [2,3], and that of Cu nanoparticles (NPs) has been observed in four dimensions using electron tomography/scanning transmission electron microscopy (STEM) [4]. However, even though the aforementioned state-of-the-art techniques shed light on

some solid-state sintering phenomena, they do not provide deeper insights into the sintering process because of the complexity of the target material and observation uncertainty (*i.e.*, experimental noise and errors). Therefore, a coupling methodology to combine experimental observations and physics-based simulation models is required to effectively elucidate the mechanism of solid-state sintering and predict the evolution of sintered materials [5,6].

The phase-field (PF) method [7] is a powerful physics-based model for simulating physical phenomena occurring at the nanoscale and mesoscale. Various PF models of sintering have been proposed, and some of them have been experimentally validated through investigations of neck growth and densification [8–10]. However,

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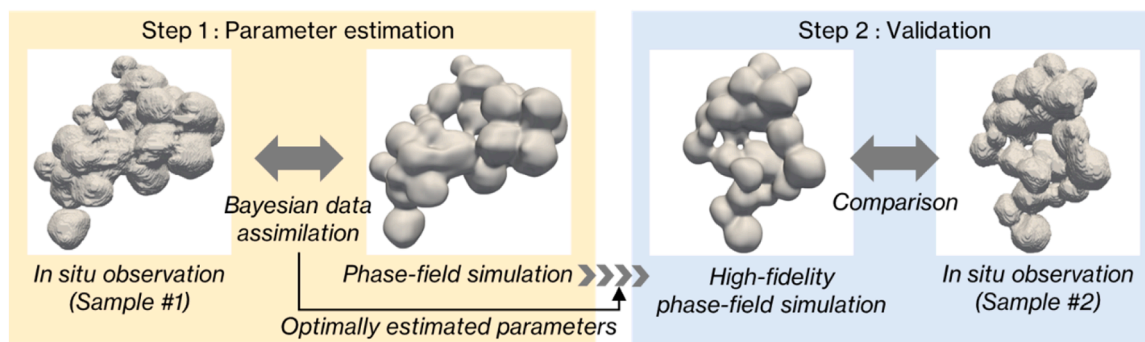


Fig. 1. Schematic of the PF model combined with *in situ* observation results for the solid-state sintering of Cu NPs.

considering that the PF model is a continuum one, the included physical and material parameters, such as interfacial energy and mobility, must be accurately identified to simulate the target physical phenomena, even if the PF model itself provides highly accurate prediction. The direct experimental identification of all material parameters required for quantitative PF simulation is difficult. Despite the multitude of material parameters reported to date (e.g., interfacial energies and diffusion coefficients), most are calibrated under ideal conditions. Therefore, an accurate prediction of solid-state sintering is impossible even if the PF simulation of solid-state sintering is performed using values available in literature.

Bayesian data assimilation (DA) is a powerful statistical method for *assimilating* experimental data into numerical simulations based on Bayes' theorem. It can also be used to estimate unmeasurable information from available experimental datasets [11], with successful applications, including more accurate weather [12] and typhoon [13] forecasting. Moreover, Bayesian DA can simultaneously and inversely identify unknown material parameters from small experimental datasets. Previous numerical studies have demonstrated that the inverse estimation of material parameters required for high-fidelity PF simulations can be performed using numerous DA methods such as the ensemble Kalman filter [14–16] and ensemble four-dimensional variational method [17–19]. Rosenthal et al. [20] used the bootstrap particle filter to assimilate the *in situ* observation data for sintered polyamide-12 (nylon), obtained via X-ray contrast tomography, into the corresponding PF model. However, to the best of our knowledge, no study has reported the inverse estimation of multiple material parameters for the PF modeling of solid-state sintering from four-dimensional electron tomography datasets.

To bridge the aforementioned gap and perform high-fidelity PF simulations, we propose a new method for combining a PF simulation and *in situ* observation datasets obtained via four-dimensional electron tomography/STEM and inversely estimating multiple material parameters from *in situ* observation data, which is essential for a high-fidelity PF simulation. Furthermore, the proposed method was used to perform a

high-fidelity PF simulation of the solid-state sintering of Cu NPs. Fig. 1 illustrates the concept of combining a PF model and *in situ* observation results of the solid-state sintering of Cu NPs. In the first step, a PF model for solid-state sintering was selected. The proposed method identifies optimal parameters that can be utilized in PF simulations based on arbitrary models to reproduce the experimental data. The more advanced and accurate the PF model used, the closer the estimated values are to the real ones. Conversely, by verifying the accuracy of the estimated values, the proposed method can contribute to the experimental validation of PF models. To demonstrate the ability of the proposed method to enable a primitive PF model to capture the observed sintering behavior, in this study, we adopted a commonly used PF model (proposed by Wang [21]) that can simulate the rigid-body motion of Cu NPs during the sintering process. The physical values and temperature-dependent material parameters included in the PF model were estimated using the Bayesian DA method from the time-series experimental data of nonisothermal solid-state sintering. The morphological information on Cu NPs obtained via an *in situ* experiment was used as the observation data for the DA. The use of optimally estimated parameters enabled the high-fidelity PF simulation of the solid-state sintering of Cu NPs. In the second step, the fidelity of the PF simulation was validated by performing a PF simulation of nonisothermal solid-state sintering using an initial Cu NP configuration different from that adopted for parameter estimation. For practical utility, we demonstrated that seven parameters, including the temperature-dependent surface and grain-boundary diffusion coefficients, of Cu atoms can be inversely estimated from the morphological information on sintered Cu NPs. Finally, we investigated the extrapolative predictability of the estimated parameters.

2. *In situ* observation

2.1. Experimental conditions

Two clusters of Cu NPs with a bimodal size distribution (Mitsui Mining & Smelting Co., Ltd.; average diameter of the core Cu NPs = 150 nm, diameter of the shell Cu NPs < 10 nm) with different initial configurations (sample #1 and #2) were observed using aberration-corrected STEM (Titan Cubed G2, Thermo Fisher Scientific). The numerous shell Cu NPs, which function as sintering aids, were adsorbed and covered the entire surface of the core Cu NPs. Notably, the STEM images only captured the behavior of the core Cu NPs because the signal intensity of the shell Cu NPs was much smaller than that of the core Cu NPs [4]. Therefore, the discussions hereafter focus on the core Cu NPs.

In this study, an acceleration voltage of 300 kV and a convergence semi-angle of 1.2 mrad were used. The tilt angle was changed from -62° to 62° in increments of 2° . The probe current was set to 10 pA or less. The number of NPs in the field of view was 27 for sample #1 and 24 for sample #2. Atmospheric contamination was prevented by using an argon-filled glove box and a heating TEM holder with an atmospheric

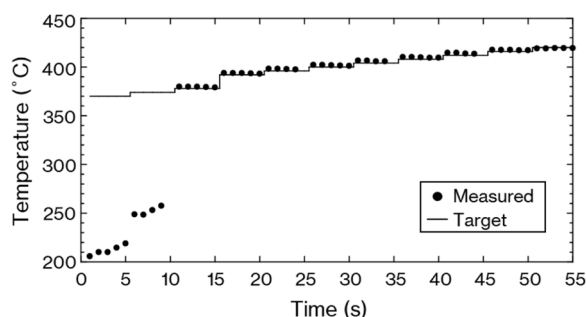


Fig. 2. Sintering temperature variation during the *in situ* observation of the Cu NPs.

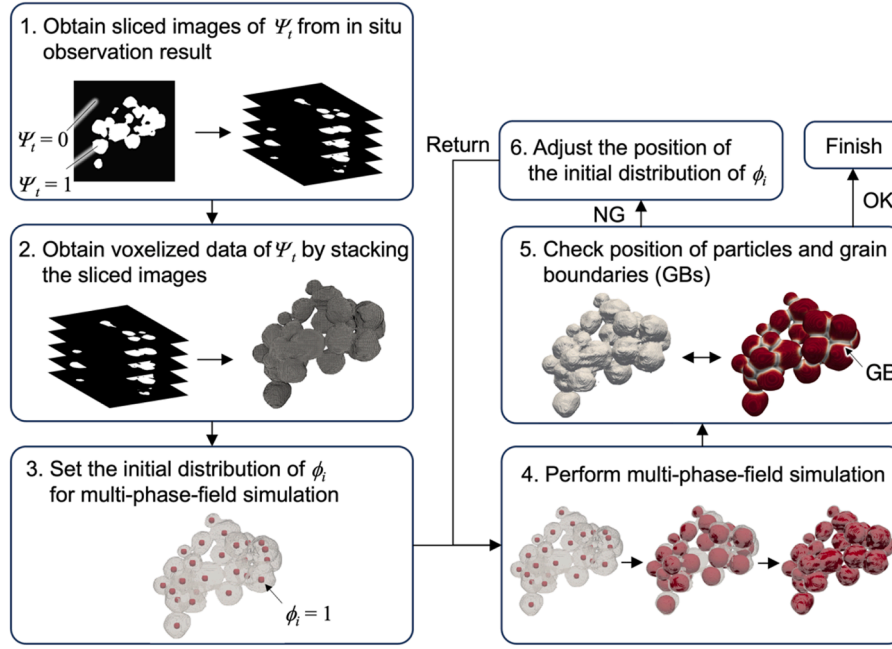


Fig. 3. Flowchart for obtaining observation data (distributions of the variable Ψ_t) and the initial distributions of the PF variables.

isolation function (Mel-Build Co. & NORCADA Inc.). The sample was initially maintained at 200 °C for 1 h and subsequently heated to the target temperature (370–420 °C). After heating for 5 s, the sample was cooled to 200 °C and the tilt series of high-angle annular dark field images was acquired. Because the *in situ* observation data were acquired at low temperatures, no morphological change occurred in the Cu NPs during the observation. Once the observation data were acquired, the sample was reheated. The cooling and heating times were considerably shorter than the sintering time. Ihara et al. [4] confirmed the absence of a significant difference between neck formation via pulse and continuous heating. The heating–observation cycle was iterated up to a heating time of 55 s. The temperature profile for sintering is shown in Fig. 2. Although the target temperature was increased stepwise, the sintering occurred under isothermal conditions in each 5-s heating interval. The tilt series images were denoised using a blocking-matching and three-dimensional (3D) filtering algorithm [22]. Thereafter, a compressed sensing implemented algorithm (iterative series reduction; ISER) [23,24] was used to reduce the artifacts produced by the 3D reconstruction process. The parameters for the ISER process were set as follows: iterations = 300, application times = 63×1 , and sensitivity = 9999. A detailed description of the denoising techniques can be found in the literature [4].

2.2. Postprocessing methods for *in situ* observation

The 3D morphology of the sintered Cu NPs was used as the observation data for the DA. As shown in Fig. 3, the observation data, which included the distribution of Ψ_t and initial distributions of the PF variables were obtained as follows: (i) The *in situ* observation results were binarized to obtain sliced image data. The binarized value obtained from the image data at time t was defined as Ψ_t , where $\Psi_t = 1$ inside the Cu NP and $\Psi_t = 0$ in other regions. (ii) The sliced images of Ψ_t were stacked in 3D to obtain the voxelized data of Ψ_t with $256 \times 256 \times 128$ regularly divided grid points. (iii) The PF variables ϕ_i ($i = 1, 2, \dots, N$) were manually set at the center of Cu NPs based on the voxelized data of Ψ_t . (iv) The multi-phase-field (MPF) simulation of grain growth [25] was performed, where the interfacial mobility was defined as zero in the region $\Psi_t = 0$. (v) The consistency of the position of the grain boundaries obtained via MPF simulation with that of the *in situ* observation was

checked. (vi) Steps (iv)–(v) were repeated with an adjusted initial distribution of ϕ_i until the grain boundary positions obtained via MPF simulation agreed with that of the observation data. The observation data used in this study consisted of 11 time-frame datasets obtained at 5-s intervals.

To quantitatively evaluate the migration of Cu NPs observed *in situ*, their centers of mass, $\mathbf{r}_{c,i}$, were calculated using the distribution of ϕ_i obtained after the MPF simulation as

$$\mathbf{r}_{c,i} = \frac{1}{V_i} \int_V \mathbf{r} \phi_i d^3 r, \quad (1)$$

where $\mathbf{r}$ denotes the position vector. Particle migration was evaluated based on the change in the particle center as follows:

$$\Delta \mathbf{r}_{c,i}^t = \mathbf{r}_{c,i}^{t+1} - \mathbf{r}_{c,i}^t, \quad (2)$$

where the upper superscript denotes time. The approximate volume of the sintered Cu NP clusters and their densification rates were quantitatively evaluated based on the volume of the convex hull [26] with the centers of mass of the particles. The approximate volume of the sintered Cu NP clusters was calculated by applying the following steps: (i) The coordinates of the center of mass of the particle for the *in situ* observation data were calculated using Eq. (1). (ii) Using the convex hull method from the Scipy library [26], the volume of the convex hull was calculated from the 3D coordinate information of the center of mass of the particle obtained in (i). Note that the approximated volume is not the total volume of the sintered particles, but the approximate volume of the particle cluster as a single sintered body.

3. Numerical framework

3.1. Phase-field model

As described in Section 2.1, our experiment was designed to maintain a constant temperature in each 5-s sintering interval. In addition, temperature-distribution analysis confirmed that a spatially uniform isothermal condition could be assumed in the Cu NPs during sintering (see the Supplementary Material). Therefore, the PF model of solid-state sintering used in this study was based on those proposed in the previous

studies [21,27,28]; the PF model was based on the isothermal assumption. The evolution of the Cu NPs during sintering was simulated using two types of PF variables, ρ and η_i ($i = 1, 2, \dots, N$; where N is the number of particles) [21]. Both PF variables were defined to change smoothly from zero to unity in the interface region. The total free energy of the target system, F , was defined as

$$F = \int_V \left\{ f_{\text{bulk}}(\rho, \eta_i) + \frac{\kappa_\rho}{2} |\nabla \rho|^2 + \sum_i \frac{\kappa_\eta}{2} |\nabla \eta_i|^2 \right\} dV, \quad (3)$$

where f_{bulk} is the chemical free-energy density in the bulk defined as

$$f_{\text{bulk}} = A\rho^2(1 - \rho)^2 + B \left[\rho^2 + 6(1 - \rho) \sum_i \eta_i^2 - 4(2 - \rho) \sum_i \eta_i^3 + 3 \left(\sum_i \eta_i^2 \right)^2 \right]. \quad (4)$$

The coefficients A and B are defined as [27,28]

$$A = \frac{12\gamma_{\text{surf}} - 7\gamma_{\text{gb}}}{\delta}, \quad (5)$$

$$B = \frac{\gamma_{\text{gb}}}{\delta}, \quad (6)$$

where γ_{surf} and γ_{gb} are the surface and grain-boundary energies, respectively, and δ is the thickness of the diffuse interface. The second and third terms on the right-hand side of Eq. (3) represent gradient free-energy densities. κ_ρ and κ_η are the gradient energy coefficients calculated as follows [27,28]

$$\kappa_\rho = \frac{3}{4} (2\gamma_{\text{surf}} - \gamma_{\text{gb}}) \delta, \quad (7)$$

$$\kappa_\eta = \frac{3}{4} \gamma_{\text{gb}} \delta. \quad (8)$$

Owing to the conservation of mass, ρ was treated as a conserved variable, and the time evolution equation of ρ was based on the Cahn–Hilliard equation [29]:

$$\frac{\partial \rho}{\partial t} = \nabla \cdot \left(M_\rho \nabla \frac{\delta F}{\delta \rho} - \rho \sum_i \mathbf{v}_i \right), \quad (9)$$

where M_ρ is the mobility of ρ , and $\mathbf{v}_i$ is the advection velocity of the i th particle ($i = 1, 2, \dots, N$). Assuming that the rigid-body motions of sintered particles consist of translation and rotation, $\mathbf{v}_i$ is defined as [21]

$$\mathbf{v}_i = \mathbf{v}_i^{\text{tr}} + \mathbf{v}_i^{\text{ro}}, \quad (10)$$

where $\mathbf{v}_i^{\text{tr}}$ and $\mathbf{v}_i^{\text{ro}}$ are the translational and rotational velocities of the i th particle, respectively. Generally, the advection-induced rigid-body motions of sintered particles occur when the particles are in contact with each other. Therefore, the effective local force, $d\mathbf{F}_i$, that leads to rigid-body motion can be formulated as [21]

$$d\mathbf{F}_i = k_{\text{st}} \sum_{j \neq i} (\rho - \rho_0) \langle \eta_i \eta_j \rangle [\nabla \eta_i - \nabla \eta_j] d^3 \mathbf{r}, \quad (11)$$

where k_{st} is the stiffness constant, ρ_0 is the equilibrium value of ρ in the grain-boundary region, and $\langle \eta_i \eta_j \rangle$ is defined as

$$\langle \eta_i \eta_j \rangle = \begin{cases} 1 & (\eta_i \eta_j \geq c) \\ 0 & (\eta_i \eta_j < c) \end{cases}, \quad (12)$$

where c is the threshold for identifying grain-boundary regions. The force acting on the i th particle, $\mathbf{F}_i$, is defined as

$$\mathbf{F}_i = \int_V d\mathbf{F}_i. \quad (13)$$

In NP sintering, small forces, such as the van der Waals and liquid

bridge adhesion forces, that act on the noncontacting particles cannot be neglected [30]. Because the surfaces of the experimentally observed Cu NPs were assumed to have no liquid phase, only the van der Waals forces were considered. The van der Waals force acting on the i th particle, $\mathbf{F}_i^{\text{vdw}}$, is defined as [31]

$$\mathbf{F}_i^{\text{vdw}} = \sum_j \frac{A}{6z^2} \left(\frac{d_i d_j}{d_i + d_j} \right) \left(\frac{1}{1 + 11.12z/\lambda} \right) \frac{\mathbf{r}_{c,j} - \mathbf{r}_{c,i}}{|\mathbf{r}_{c,j} - \mathbf{r}_{c,i}|}, \quad (14)$$

where A is the Hamaker constant, λ is the characteristic wavelength (~ 100 nm), d_i is the radius of the i th particle, z is the separation distance, and $\mathbf{r}_{c,i}$ is the center position of the i th particle. Using the forces calculated using Eqs. (11), (13), and (14), $\mathbf{v}_i^{\text{trans}}$ and $\mathbf{v}_i^{\text{rot}}$ can be defined as

$$\mathbf{v}_i^{\text{tr}} = \frac{m_{\text{tr}}}{V_i} (\mathbf{F}_i + \mathbf{F}_i^{\text{vdw}}) \eta_i, \quad (15)$$

$$\mathbf{v}_i^{\text{ro}} = \frac{m_{\text{ro}}}{V_i} \mathbf{T}_i \times [\mathbf{r} - \mathbf{r}_{c,i}] \eta_i, \quad (16)$$

where m_{tr} and m_{ro} are the translational and rotational mobilities, respectively; V_i is the i th particle volume; and $\mathbf{T}_i$ is the torque calculated as

$$\mathbf{T}_i = \int_V [\mathbf{r} - \mathbf{r}_{c,i}] \times d\mathbf{F}_i. \quad (17)$$

The center position, $\mathbf{r}_{c,i}$, of the particle was calculated as

$$\mathbf{r}_{c,i} = \frac{1}{V_i} \int_V \mathbf{r} \eta_i d^3 \mathbf{r}. \quad (18)$$

In Eq. (9), M_ρ is defined as [32]

$$M_\rho = M_{\text{vol}} h(\rho) + M_{\text{vap}} \{1 - h(\rho)\} + M_{\text{surf}} \rho^2 (1 - \rho)^2 + \sum_i \sum_{j \neq i} M_{\text{gb}} \eta_i \eta_j, \quad (19)$$

where $h(\rho)$ is an interpolation function defined as $h(\rho) = \rho^3(10 - 15\rho - 6\rho^2)$. M_{vol} , M_{vap} , M_{surf} , and M_{gb} are the mobilities of the volume, vapor, surface, and grain-boundary diffusion, respectively, calculated using the corresponding diffusion coefficients D_X as $M_X = D_X V_m / RT$ ($X = \text{vol, vap, surf, gb}$), where V_m is the molar volume, R is the universal gas constant, and T is the absolute temperature. The diffusion coefficients obtained via the Arrhenius law: $D_X = D_X^0 \exp(-Q_X/RT)$, where D_X^0 is the pre-exponential factor and Q_X is the activation energy. Notably, M_ρ depends on the temperature-dependent diffusion coefficient. Thus, the PF model can address the stepwise increase in temperature after the observation.

The PF variable, η_i , is a nonconserved variable because particle size can change as a result of grain growth during sintering. Therefore, the time evolution equation for η_i was derived based on the Allen–Cahn equation [33]:

$$\frac{\partial \eta_i}{\partial t} = -M_\eta \frac{\delta F}{\delta \eta_i} - \nabla \cdot (\eta_i \mathbf{v}_i), \quad (20)$$

where M_η is the mobility related to grain-boundary migration. In the PF simulation of sintering, Eqs. (9) and (20) were solved via the finite difference scheme. Spatial discretization was conducted via the second-order central finite difference method. Time integration was calculated by using the first order Euler scheme. To strictly satisfy conservation of mass, ρ in the interfacial region ($0.1 \leq \rho \leq 0.9$) was corrected using $\Delta \rho / n_p$, where $\Delta \rho$ is the amount of change in the total ρ per computational time step and n_p is the number of computational grids in the interfacial region (more details are provided in the Supplementary Material).

In this study, the seven parameters in the PF model were simultaneously estimated through nonsequential DA. Four of these parameters (D_{surf}^0 , D_{gb}^0 , Q_{surf} , and Q_{gb}) were used to determine the temperature-dependent surface and grain-boundary diffusion coefficients. The

Table 1

To-be-estimated parameters, ranges of their possible values, background values, and standard deviations used to determine the uncertainty of initially estimated parameters.

Parameter	Range	Background mean	Standard deviation
D_{surf}^0 [$\text{m}^2 \text{s}^{-1}$]	$10^{-7} \leq D_{\text{surf}}^0 \leq 10^3$	1.0×10^{-7}	25 %
Q_{surf} [J mol^{-1}]	$5 \times 10^4 \leq Q_{\text{surf}} \leq 2 \times 10^5$	1.0×10^5	10 %
D_{gb}^0 [$\text{m}^2 \text{s}^{-1}$]	$10^{-7} \leq D_{\text{gb}}^0 \leq 10^3$	1.0×10^{-7}	25 %
Q_{gb} [J mol^{-1}]	$5 \times 10^4 \leq Q_{\text{gb}} \leq 2 \times 10^5$	1.0×10^5	10 %
m_{tr} [$\text{m}^5 \text{J}^{-1} \text{s}^{-1}$]	$0 \leq m_{\text{tr}} \leq 10^{-20}$	5.0×10^{-21}	1 %
M_{η} [$\text{m}^3 \text{J}^{-1} \text{s}^{-1}$]	$0 \leq M_{\eta} \leq 10^{-9}$	5.0×10^{-10}	10 %
k_{st} [N m^{-2}]	$10^6 \leq k_{\text{st}} \leq 10^8$	1.0×10^7	10 %

other three parameters were m_{tr} , k_{st} , and M_{η} . The sensitivity analysis results on k_{st} and m_{tr} are shown in the Supplementary Material.

3.2. Data assimilation

A nonsequential DA method [19], which minimizes the four-dimensional cost function using a tree-structured Parzen estimator (DMC-TPE), was used in this study. The DMC-TPE method relied on Bayesian optimization to determine the minimum value of the cost function, which represents the time integral of the mismatch between the time-series observation data and numerical simulation results. The set of material parameters minimizing this function was defined as that

of optimally estimated parameters leading to experimental data reproduction [19]. To perform the DA using DMC-TPE, the augmented state vector, $\mathbf{x}_t$, and observation vector, $\mathbf{y}_t$, were defined. $\mathbf{x}_t$ consisted of all PF variables on all computational grids at time t ($0 \leq t \leq t_{\text{end}}$) and the parameters to be estimated. $\mathbf{y}_t$ was composed of Ψ_t . Therefore, $\mathbf{x}_t$ was defined as:

$$\mathbf{x}_t = (\rho_t[0], \rho_t[1], \dots, \rho_t[N_{\text{grid}}], \eta_{1t}[0], \eta_{1t}[1], \dots, \eta_{1t}[N_{\text{grid}}], \eta_{2t}[0], \eta_{2t}[1], \dots, \eta_{2t}[N_{\text{grid}}], \dots, \eta_{Nt}[0], \eta_{Nt}[1], \dots, \eta_{Nt}[N_{\text{grid}}], \boldsymbol{\theta})^T. \quad (21)$$

Here, $\rho_t[j]$ and $\eta_{it}[j]$ ($i = 1, 2, 3, \dots, N$) are the order parameters on the finite difference grids j ($j = 1, 2, 3, \dots, N_{\text{grid}}$) at time t , where N_{grid} is the number of finite difference grids. The vector $\boldsymbol{\theta}$ contains the seven parameters to be estimated: $\boldsymbol{\theta} = (D_{\text{surf}}^0, D_{\text{gb}}^0, Q_{\text{surf}}, Q_{\text{gb}}, m_{\text{tr}}, k_{\text{st}}, M_{\eta})$. $\mathbf{T}$ denotes a transposition. $\mathbf{y}_t$ was defined as

$$\mathbf{y}_t = (\Psi_t[0], \Psi_t[1], \dots, \Psi_t[N_{\text{grid}}])^T, \quad (22)$$

where $\Psi_t[j]$ ($j = 1, 2, 3, \dots, N_{\text{grid}}$) denotes the value of Ψ_t on the finite difference grids j . Both $\mathbf{x}_t$ and $\mathbf{y}_t$ were considered stochastic variables because the numerical simulation results and observation data contained numerical and experimental errors. The state and parameters were estimated by optimizing the initial state vector $\mathbf{x}_0$ by minimizing the cost function, which represents the data misfit between $\mathbf{x}_t$ and $\mathbf{y}_t$ from time $t = 0$ to $t = t_{\text{end}}$, as discussed in the literature [18,19].

$$J(\mathbf{x}_0) = \frac{1}{2}(\mathbf{x}_0 - \mathbf{x}_0^b)^T \mathbf{B}^{-1}(\mathbf{x}_0 - \mathbf{x}_0^b) + \sum_{t=0}^{t_{\text{end}}} \frac{1}{2}(H_t(\mathbf{x}_t) - \mathbf{y}_t)^T \mathbf{R}_t^{-1}(H_t(\mathbf{x}_t) - \mathbf{y}_t), \quad (23)$$

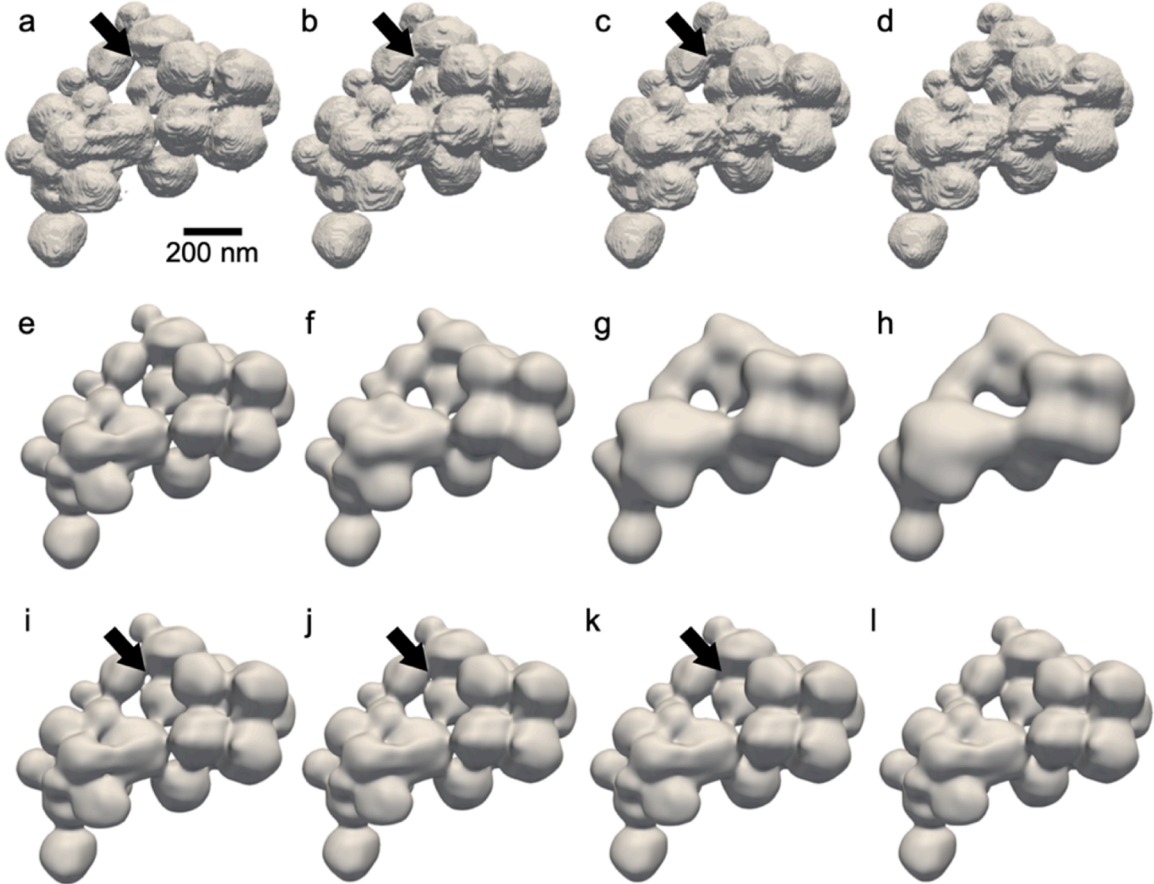


Fig. 4. Results of the (a–d) *in situ* observation of solid-state Cu NPs sintering for sample #1 and (e–h) PF simulation based on previously reported diffusion coefficients [35,37], (i–l) PF simulation performed by assimilating the *in situ* observation data using the DA method. Black arrows indicate pore annihilation locations. The temperatures for each image (from left to right) are 200, 378, 404, and 420 °C and correspond to sintering times of 0, 15, 35, and 55 s, respectively.

where $\mathbf{x}_0^b$ denotes the background state vector containing the initially estimated state variables and parameters. To extract only ρ_t from $\mathbf{x}_t$ and compare them to the components of $\mathbf{y}_t$, an experiment operator, H_t , can be defined as

$$H_t = \text{diag.}[1, 1, \dots, 1, 0, 0, \dots, 0] \quad (24)$$

Here, the first N_{grid} components of Eq. (24) are unity, whereas the others are zero. Therefore, $H_t(\mathbf{x}_t)$ corresponds to $H_t(\mathbf{x}_t) = (\rho_t[0], \rho_t[1], \dots, \rho_t[N_{\text{grid}}])^T$. The matrices $\mathbf{B}$ and $\mathbf{R}_t$ in Eq. (23) represent the background and observation error covariance matrices, respectively, representing the uncertainties in $\mathbf{x}_0^b$ and $\mathbf{y}_t$.

The maximum number of minimization iterations of the cost function was set to 400. The optimally estimated values were contained in $\mathbf{x}_0$, which provided the minimum cost function. For the estimation of Cu atom diffusion coefficients, the relationship $D_{\text{surf}} > D_{\text{gb}} > D_{\text{vol}}$ was assumed to be satisfied regardless of the sintering temperature because this relationship generally holds at high temperatures [34,35]. Table 1 summarizes the ranges of possible values, background values, and standard deviations determined in a preliminary study and used to determine $\mathbf{B}$. The background (i.e., initially estimated) values of D_{surf}^0 and D_{gb}^0 were set to the lower bounds of the respective ranges to prevent numerical divergence caused by large diffusion coefficients. The standard deviations were set to smaller values for parameters more sensitive to state changes [17]. Therefore, $\mathbf{B}$ is defined by the diagonal matrix as

$$\mathbf{B} = \text{diag.} \left[0, \dots, 0, \left(\sigma_{D_{\text{surf}}^0} \right)^2, \left(\sigma_{Q_{\text{surf}}} \right)^2, \left(\sigma_{D_{\text{gb}}^0} \right)^2, \left(\sigma_{Q_{\text{gb}}} \right)^2, \left(\sigma_{m_{\text{tr}}} \right)^2, \left(\sigma_{M_{\eta}} \right)^2, \left(\sigma_{k_{\text{st}}} \right)^2 \right]. \quad (25)$$

Here, the first $N_{\text{grid}} + NN_{\text{grid}}$ components of $\mathbf{B}$ were zero because the initial distribution of ρ_t and η_{it} were obtained from the *in situ* observation results. σ_k ($k = D_{\text{surf}}^0, Q_{\text{surf}}, D_{\text{gb}}^0, Q_{\text{gb}}, m_{\text{tr}}, M_{\eta}$, and k_{st}) are the standard deviations of initially estimated parameters listed in Table 1.

Assuming that the correlation between the observation data at different locations is zero, $\mathbf{R}_t$ is defined by the following diagonal matrix:

$$\mathbf{R}_t = \text{diag.} \left[(\sigma_{\psi})^2, \dots, (\sigma_{\psi})^2 \right] \quad (26)$$

where the dimension of Eq. (26) is N_{grid} and all the components are σ_{ψ} , which is the standard deviation of ψ_t that represents the uncertainty of *in situ* observation data. In this study, we used $\sigma_{\psi} = 0.1$.

DA and PF simulations incur relatively high computational costs. Therefore, the supercomputer Wisteria at the University of Tokyo in Japan [36] was used in this study. We performed parallel computation with eight GPUs, which allowed the execution of 400 iterations within approximately 60 h.

4. Results and discussion

4.1. In situ observation of Cu NP sintering

To obtain the data to be assimilated into the PF simulation (step 1 in Fig. 1), we observed solid-state sintering in two samples with different initial Cu NP configurations (samples #1 and #2). The data obtained for sample #1 were used for the inverse estimation of material parameters using DA, whereas those obtained for sample #2 were used to validate the fidelity of the PF simulation using the estimated parameters (step 2 in Fig. 1). The main findings obtained from the *in situ* observation of Cu NPs are reported in the literature [4].

Fig. 4a–d shows the morphological changes of Cu NPs in sample #1 during the sintering experiment obtained via STEM; the NP cluster morphology is depicted using the distributions of a binarized variable, ψ_t . Neck formation between the NPs was detected during the solid-state sintering, and the shrinkage and disappearance of interparticle voids or pores (indicated by arrows in Fig. 4a–c) caused macroscopic shrinkage and densification. Because conventional theories regarding solid-state

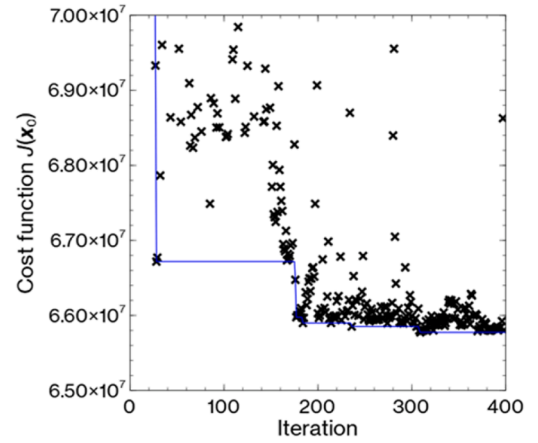


Fig. 5. Evolution of cost function calculated through DA using the *in situ* observation data for sample #1. The blue line shows the evolution of the cost function minimum updated during iterative minimization.

sintering deal with one or two particles, they do not provide sufficient insights into the observed sintering process and are unsuited for the inverse identification of material parameters from observation data. In contrast, the DA method enables the identification of material parameters from the morphological information on sintered Cu NPs, as described below.

4.2. Estimation of material parameters using DA

To estimate the seven material parameters, including the temperature-dependent diffusion coefficients of Cu atoms, we assimilated the observation data (i.e., the distribution of ψ_t obtained from the *in situ* observation results shown in Fig. 4a–d) into the PF simulation [21] using DMC-TPE, as described in Section 3.2. Note that both the observation data and PF model used in this study had certain limitations. The observation data did not contain the shell Cu NPs that affect neck growth [4], and the PF model was primitive (e.g., the simulation results depended on interface thickness). These limitations were treated as uncertainties in the DA. Therefore, estimated values were provided to accurately predict the experimentally observed sintering behavior in the PF simulation performed under the conditions described below. The computational domain had dimensions of $1.14 \mu\text{m} \times 1.14 \mu\text{m} \times 0.57 \mu\text{m}$ for sample #1 and $1.44 \mu\text{m} \times 1.44 \mu\text{m} \times 0.72 \mu\text{m}$ for sample #2. Because both domains were discretized with $128 \times 128 \times 64$ finite-difference grids, the grid spacings were $\Delta x = \Delta y = \Delta z = 8.90 \text{ nm}$ for sample #1 and $\Delta x = \Delta y = \Delta z = 11.27 \text{ nm}$ for sample #2. The measured temperature profile, shown in Fig. 2, was used for the PF simulation. For the first 10 s of sintering, the measured temperatures continuously increased with lower temperatures than the target temperatures. However, we assume that the isothermal PF model could be used, despite this temperature change, because negligible sintering occurred during the low-temperature period. The surface energy was set as $\gamma_{\text{surf}} = 1.35 \text{ J m}^{-2}$ [38]. The grain-boundary energy was assumed to depend on temperature and was set as $\gamma_{\text{gb}} = 0.783 - 1.34 \times 10^{-4} T \text{ J m}^{-2}$ [39]. The parameters related to the advection velocity were $\rho_0 = 0.98$, $c = 0.14$, and $m_{\text{r0}} = m_{\text{tr}}/100$ [21]. The time increment was $\Delta t = 1 \times 10^{-3} \text{ s}$. The volume diffusivity pre-exponential factor and activation energy were $D_{\text{vol}}^0 = 3.5 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ and $Q_{\text{vol}} = 2.04 \times 10^5 \text{ J mol}^{-1}$ [34], respectively. The vapor diffusion coefficient, D_{vap} , was assumed to be one-tenth of D_{vol} because mass transport in the vapor phase is substantially smaller than the sintering temperature is significantly lower than the melting point of Cu. The initial distributions of the Cu NPs were determined based on experimental data for $t = 0 \text{ s}$. Note that previous work [40] revealed the significant influence of ρ_0 on sintering behavior. Therefore, we conducted DA to estimate nine parameters, including

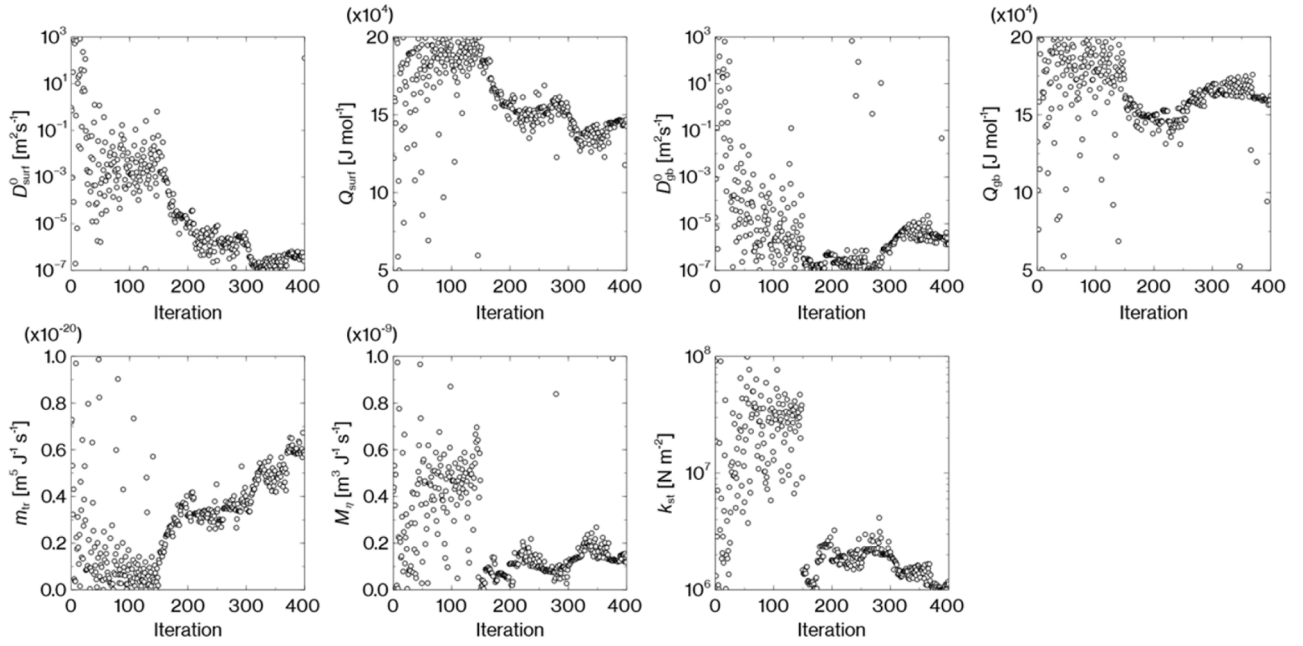


Fig. 6. Parameters variations during cost function minimization.

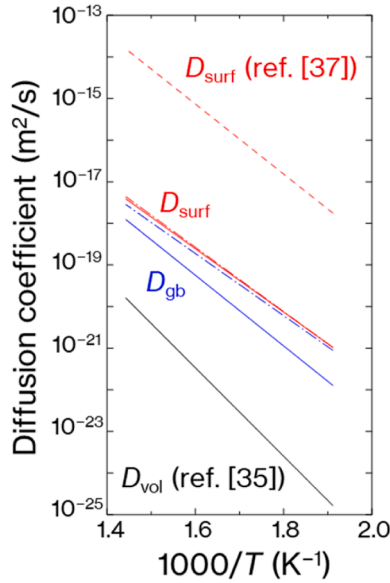


Fig. 7. Temperature dependence of volume, surface, and grain-boundary diffusion coefficients calculated using the optimally estimated parameters.

ρ_0 and m_{ro} , in the preliminary investigation. The results showed that although the estimation of ρ_0 and m_{ro} affected the parameter estimation results, this process did not have a distinct effect on the PF simulation results obtained using the estimated values. The details of the preliminary investigation are presented in the Supplementary Material.

Fig. 5 shows the evolution of the cost function obtained through DA calculations using the observation data for sample #1. Evidently, this function decreases with an increase in the number of iterative minimizations. Because convergence was almost complete after 300 iterations, we concluded that the minimum value of this function during the 400 iterative calculations could be obtained in the 308th iterative step. The optimal material parameters minimizing the cost function were as follows: $D_{surf}^0 = 3.71 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$, $Q_{surf} = 1.46 \times 10^5 \text{ J mol}^{-1}$, $D_{gb}^0 = 2.26 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$, $Q_{gb} = 1.63 \times 10^5 \text{ J mol}^{-1}$, $m_{tr} = 3.78 \times 10^{-21} \text{ m}^5 \text{ J}^{-1} \text{ s}^{-1}$,

$M_\eta = 1.28 \times 10^{-10} \text{ m}^3 \text{ J}^{-1} \text{ s}^{-1}$, and $k_{st} = 1.74 \times 10^6 \text{ N m}^{-2}$. Fig. 6 shows the variation of the seven parameters during cost function minimization. In the early stage, the estimated parameters have a large variation up to approximately the 150th iterative step. However, in the later stages, the variation decreases and there is a tendency toward convergence. To verify the optimally estimated parameters, the PF simulation was carried out using the estimated parameters obtained at the last (399th) iterative step. The result shows that the morphological changes of the particles were almost the same as those obtained using the best optimally estimated parameters (see Fig. S7 in the Supplementary Material).

Fig. 7 shows the temperature dependence of the three diffusion coefficients (i.e., volume, surface, and grain-boundary diffusion coefficients) to obtain the former parameters. The surface and grain-boundary diffusion coefficients were calculated using the estimated parameters. Although not all estimated parameters could be compared with previously reported values, the estimated surface diffusion coefficient was approximately two orders of magnitude smaller than that reported by Bowden and Balluffi [37]. However, the sintering behavior of the Cu NPs simulated using previously reported diffusion coefficients [35,37] differed from that obtained through the *in situ* observation. The sintering process was overpredicted when the previously reported diffusion coefficients were used (Fig. 4e–h), whereas the material parameters estimated using the DA method accurately reproduced the sintering behavior, including pore annihilation and the overall shrinkage of the sintered NPs (Fig. 4i–l). This result demonstrates that the estimated values are helpful for improving the fidelity of PF simulation, even if they differ from the values reported in literature. A more detailed comparison between the *in situ* observation data (Fig. 4a–d) and the results of the data-assimilated PF simulation (Fig. 4i–l) is described below.

Fig. 8a shows the morphological changes in the Cu NPs obtained through the *in situ* observation and PF simulation with the optimally estimated parameters for the rectangular region shown in Fig. 8b. As indicated by the arrows in Fig. 8a, the PF simulation reproduced the observed pore annihilation and rigid-body motion of Cu NPs with relative accuracy. To investigate the discrepancy between the experiment data and simulation results in more detail, we compared the center-of-mass migration behavior predicted using the PF simulation with that observed *in situ* (Fig. 8c). In the simulation results, the direction of Cu NP migration was parallel to the sum of the vectors along the direction of

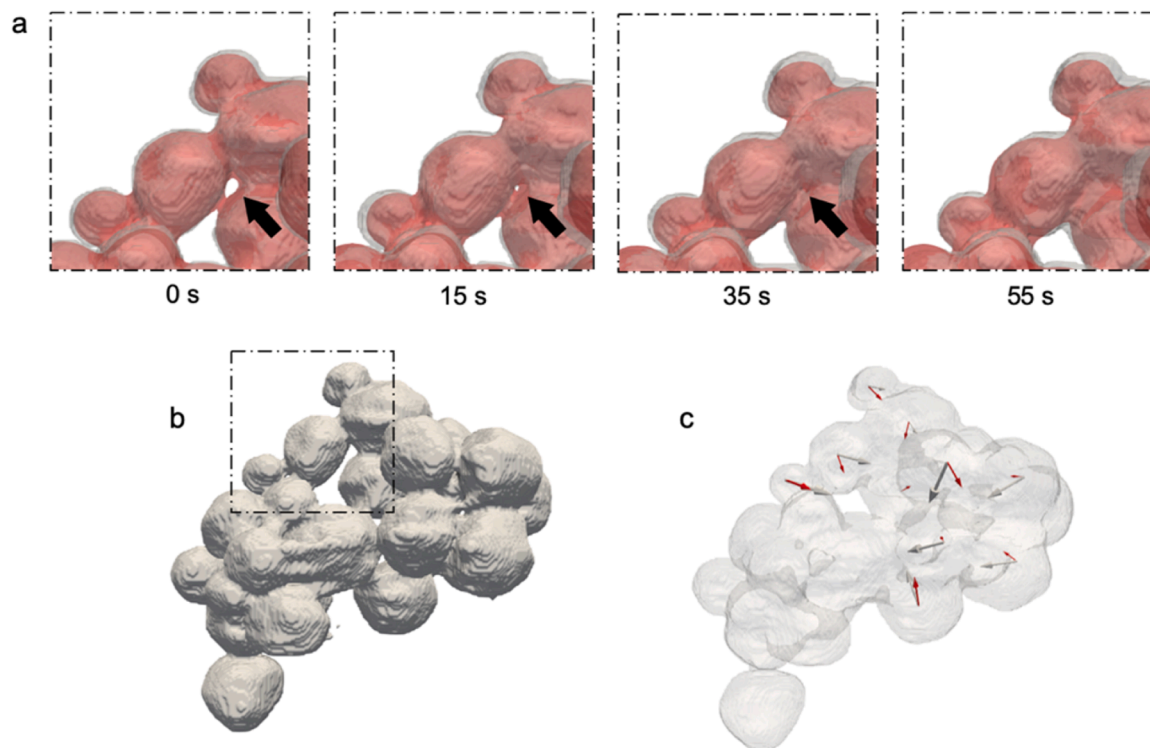


Fig. 8. (a) Morphological evolution of Cu NPs in sample #1 calculated via the PF simulation and experimentally observed for the rectangular domain shown in (b). White and red surfaces refer to the *in situ* observation data and PF simulation results, respectively. (c) Vectors show the variation in the NP center of mass, with white and red arrows corresponding to *in situ* observation and PF simulation, respectively. The vector length represents the particle migration distance.

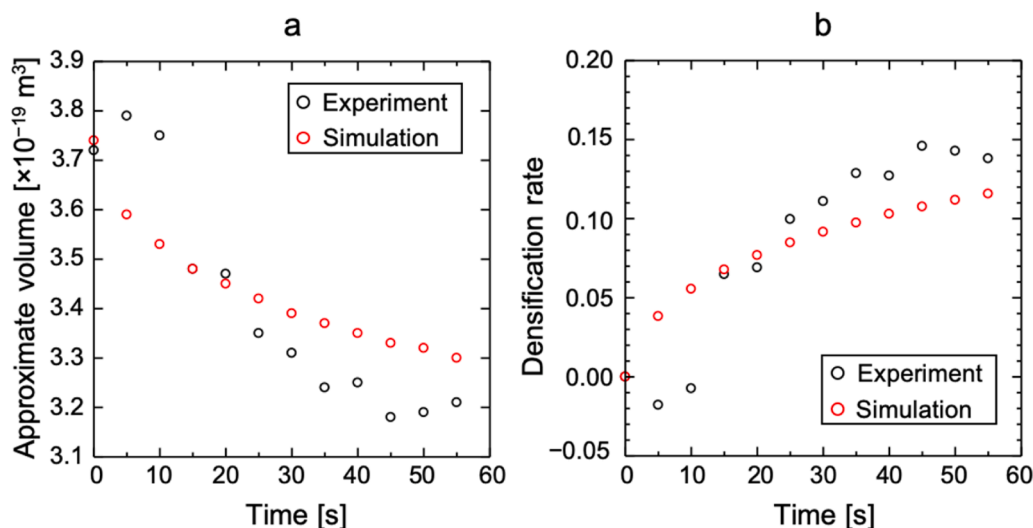


Fig. 9. Variations in (a) approximated volume and (b) densification rate of the NP cluster for sample #1 during solid-state sintering.

the interface between the neighboring NPs, whereas the NPs migrated in the direction of the center of the NP cluster in the *in situ* observations. Although the measured amounts and directions of the NP migration included errors caused by experimental noise, postprocessing using techniques such as segmentation and 3D reconstruction suggested that our PF simulation did not perfectly reproduce the actual sintering process even when the material parameters were accurately identified. However, we should emphasize that the assimilation of the *in situ* observation data into the PF simulation enables one to obtain the insightful information necessary for model improvement, as described in Fig. 1. The important insight highlighted here is the introduction of van der Waals forces into the PF model. To clarify the influence of van der

Waals forces on simulated sintering behavior, we performed PF simulation using optimally estimated parameters with and without van der Waals forces. The result shown in Fig. S8 in the Supplementary Material reveals that the introduction of van der Waals forces enables us to reproduce the experimentally observed pore annihilation.

Fig. 9 compares the time-dependent approximated volume and densification rate of Cu NP clusters extracted from *in situ* observation data with those determined using the PF simulation. The approximate volume obtained from observation data unexpectedly increased in the initial stage of sintering and subsequently decreased with small fluctuations. In contrast, the volume calculated using the PF simulation monotonically decreased with time. This discrepancy was ascribed to

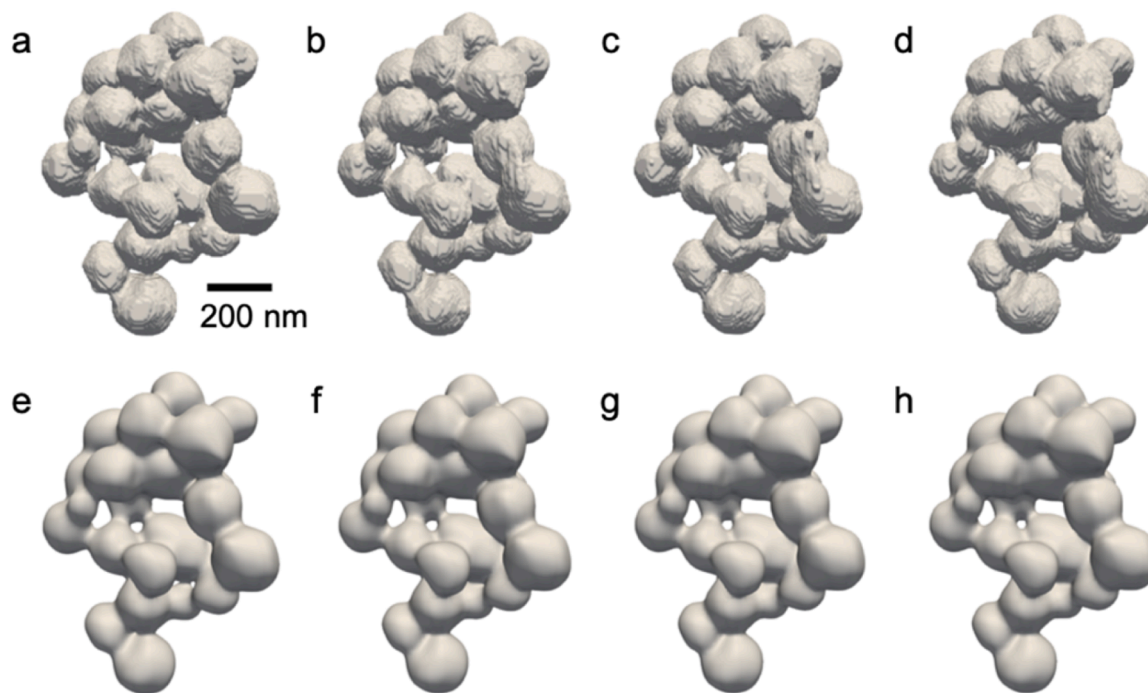


Fig. 10. (a–d) Results of the *in situ* observation of solid-state Cu NP sintering for sample #2 and (e–h) predictions obtained via high-fidelity PF simulation using optimally estimated parameters. The temperatures for each image (from left to right) are 200, 378, 404, and 420 °C, which correspond to sintering times of 0, 15, 35, and 55 s, respectively.

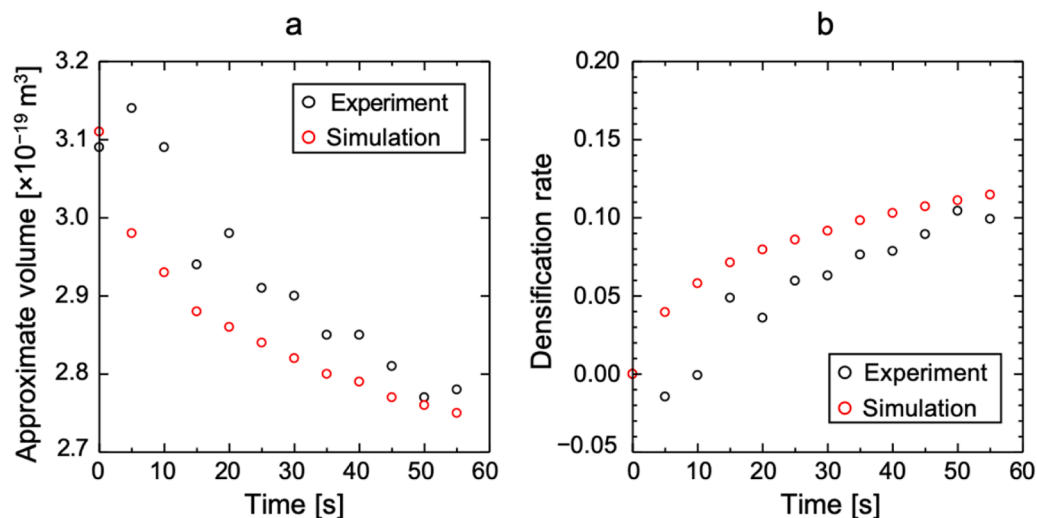


Fig. 11. Variations in (a) approximated volume and (b) densification rate for sample #2.

errors in the *in situ* observation data, as illustrated in Fig. S9. The conventional theory of solid-state sintering using two particles suggests that the densification rate follows a logarithmic function [41,42]. Therefore, the densification rate of NPs calculated using the PF simulation (Fig. 9b) was qualitatively consistent with that of the conventional theory. Notably, the good agreement between the densification rates obtained experimentally and using the PF simulation does not necessarily indicate that the simulation accurately reproduced the experimentally observed morphological and state changes in the sintered NPs (see Fig. S10).

Considering the results obtained for sample #1, we arrived at the following conclusions. First, even if the material parameter values are known from literature, they cannot be used “as is” to perform high-fidelity PF simulations because the PF simulation results obtained using these values differ substantially from the experiment data. Second,

even if the experiment results contain errors, a high-fidelity PF simulation of solid-state sintering can be performed by estimating the optimal material parameters using the DA method. Furthermore, we concluded that the DA framework facilitates on-demand identification of physical parameters in a specific target system. In the following section, we discuss the direct comparison between the *in situ* observation and PF simulation data for sample #2 to validate the PF simulation using the optimally estimated parameters.

4.3. Validation of PF simulation with optimally estimated parameters

For validation of the proposed method to obtain high-fidelity PF simulation results, we performed PF simulation of the sintering behavior of Cu NPs in sample #2 using the material parameters estimated through

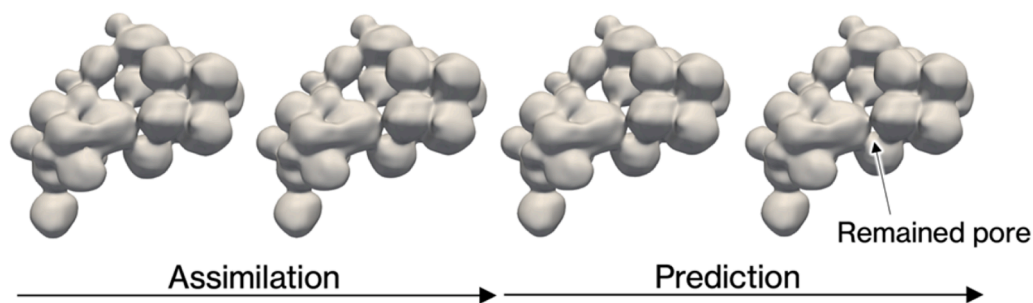


Fig. 12. Results of the PF simulation of solid-state sintering for sample #1 using the parameters estimated from the *in situ* observation data from 0 to 25 s. Compared to the PF simulation result show in Fig. 4i–l, the PF model predicts that a small pore remains after sintering for 55 s. The temperatures for each image (from left to right) are 200, 378, 404, and 420 °C and correspond to sintering times of 0, 15, 35, and 55 s, respectively.

DA using the *in situ* observation data for sample #1 and compared the results with the corresponding *in situ* observation data. Fig. 10 shows the *in situ* observation and simulation results for sintering sample #2. Unlike in sample #1, no clear pore annihilation was experimentally detected in sample #2. The high-fidelity PF simulation captured the experimentally observed sintering behavior. This result indicated that the optimal estimated parameters were reasonable for reproducing Cu NP sintering. Fig. 11a and b show variations in the approximate volume and densification rate, respectively, for sample #2. At the initial stage of sintering, the PF simulation underestimated the approximate volume; however, the extent of this underestimation decreased with time. At the final stage of sintering, the predicted volume and densification rate agreed well with the experimental values. Notably, the sintering behavior of sample #2 could not be reproduced by the PF simulation using previously reported diffusion coefficients (see Fig. S11).

The validation of the PF simulations of solid-state sintering was conducted by comparing experimentally observed macroscopic quantities such as densification rate. Note, there are no studies comparing PF simulation results with *in situ* observations of neck growth, particle rigid-body motion, or pore annihilation behavior. Furthermore, the results obtained in this study clearly demonstrated that the material parameters estimated using the proposed method help to reproduce real sintering behavior, even when a primitive PF model of solid-state sintering is used. This fact indicates that the DA method proposed in this study is expected to enable the identification of actual physical values through further improvements in PF simulation models, leading to deeper insight into the sintering process.

In this study, we estimated the parameters from the observation data in a limited temperature range and sintering time. Therefore, the applicability of the estimated parameters is not universal but currently limited. In future studies, the applicability of the PF model and estimated parameters must be demonstrated for a wider range of temperature, sintering time, heating and cooling rates, and particle geometries.

4.4. Extrapolative prediction performance

To investigate the extrapolative predictability of the estimated parameters, parameter estimation was performed using half of the observed data for sample #1 (from 0 to 25 s). The estimated parameters were used to predict the sintering behavior for the remaining 35 s. The result of the PF simulations with the estimated parameters (i.e., $D_{\text{surf}}^0 = 6.97 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$, $Q_{\text{surf}} = 1.56 \times 10^5 \text{ J mol}^{-1}$, $D_{\text{gb}}^0 = 1.68 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$, $Q_{\text{gb}} = 1.95 \times 10^5 \text{ J mol}^{-1}$, $m_{\text{tr}} = 4.00 \times 10^{-21} \text{ m}^5 \text{ J}^{-1} \text{ s}^{-1}$, $M_{\eta} = 7.40 \times 10^{-10} \text{ m}^3 \text{ J}^{-1} \text{ s}^{-1}$, and $k_{\text{st}} = 1.01 \times 10^6 \text{ N m}^{-2}$) is shown in Fig. 12. Compared to the experimental results, the reproducibility of the pore annihilation in the area indicated by the arrow is degraded. However, the overall sintering behavior is almost the same as that shown in Fig. 4i–l. Therefore, we conclude that the prediction accuracy of the PF model can be improved by performing DA using part of the experiment

data, demonstrating the extrapolative predictability of the estimated parameters.

5. Conclusions

A new technique is proposed to perform high-fidelity PF simulations by combining a PF model and *in situ* observation data of nonisothermal solid-state sintering using a nonsequential DA method. This method can estimate temperature-dependent material parameters that cannot be directly identified via experiments, thus enabling the quantitative prediction of time-series changes in sintered particles. Seven parameters to reproduce experimentally observed sintering behavior could be inversely estimated from the morphological information on sintered Cu NPs. The results of this study clearly showed that the PF simulation using the estimated parameters could quantitatively simulate the sintering behavior of Cu NPs. Thus, this study offers a new framework for constructing digital twins of solid-state sintering using Bayesian DA-integrated PF modeling and *in situ* electron tomography observations.

CRediT authorship contribution statement

Akimitsu Ishii: Writing – review & editing, Writing – original draft, Validation, Software, Methodology, Formal analysis, Data curation, Conceptualization. **Akinori Yamanaka:** Writing – original draft, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **Mizumo Yoshinaga:** Writing – original draft, Investigation, Data curation. **Shunsuke Sato:** Writing – original draft, Investigation, Data curation. **Midori Ikeuchi:** Writing – original draft, Investigation, Data curation. **Hikaru Saito:** Writing – review & editing, Investigation, Data curation, Conceptualization. **Satoshi Hata:** Writing – review & editing, Resources, Project administration, Funding acquisition, Data curation, Conceptualization. **Akiyasu Yamamoto:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

The raw data that support the findings of this study will be available from the corresponding author upon reasonable request. The source codes used in this study cannot be shared at this time as these form part of an ongoing study.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.actamat.2024.120251](https://doi.org/10.1016/j.actamat.2024.120251).

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