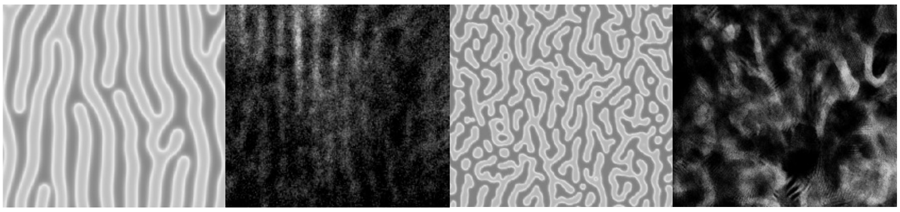

SUPPORTING INFORMATIONS

Phase Textures of Metal-Oxide Nanocomposites Self-Orchestrated by Atomic Diffusions through Precursor Alloys

Nasrat Hannah Shudin, [a, b] Ryuto Eguchi [a, c], Takeshi Fujita,[*d] Tomoharu Tokunaga, [e] Ayako Hashimoto [a] and Hideki Abe [*a, b]

a. *National Institute for Materials Science, Namiki 1-1, Tsukuba, Ibaraki 305-00443, Japan*
b. *Graduate School of Science and Engineering, Saitama University, Shimo-Okubo 255, Saitama 338-8570, Japan*
c. *TsukubaUniversity,1-chome-1-1Tennodai,Tsukuba,Ibaraki.305-8577, Japan*
d. *Kohchi University of Technology, Kami, Kochi, Japan*
e. *Institute of Materials and Systems for Sustainability, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8601, Japan*

The Turing model is generally described as a set of non-linear differential equations (*eq.*1):

$$\frac{\partial u_1}{\partial t} = D_1 \Delta u_1 + f_1 \left(u_1,\ldots\ldots,um\right)$$

$$\frac{\partial u_2}{\partial t} = D_2 \Delta u_2 + f_2 \left(u_1,\ldots\ldots,u_m\right)$$

$$\vdots \quad \vdots$$

$$\frac{\partial u_m}{\partial t} = D_m \Delta u_m + f_m \left(u_1,\ldots\ldots,u_m\right) \qquad \textit{eq. } 1.$$

For our work, we consider interaction of two chemical species of u and v;

$$\frac{\partial u}{\partial t} = f_u(u,v) + D_u \Delta u$$

$$\frac{\partial v}{\partial t} = f_v(u,v) + D_v \Delta v \qquad \textit{eq. 2.}$$

Further derived to;

$$\frac{\partial u}{\partial t} = \frac{\partial f(u)}{\partial (u)} u + \frac{\partial f(u)}{\partial v} v + D_u \Delta u$$

$$\frac{\partial v}{\partial t} = \frac{\partial f(v)}{\partial u} u + \frac{\partial f(v)}{\partial v} v + D_v \Delta v \qquad \textit{eq. 3.}$$

Simplified to;

$$\begin{matrix} \frac{\partial u}{\partial t} \\ \frac{\partial v}{\partial t} \end{matrix} = \begin{pmatrix} \partial f(u) / \partial (u) & \partial f(u) / \partial v \\ \partial f(v) / \partial u & \partial f(v) / \partial v \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} + \begin{pmatrix} D_u & o \\ o & D_v \end{pmatrix} \begin{pmatrix} \Delta u \\ \Delta v \end{pmatrix} + \begin{pmatrix} c_u \\ c_v \end{pmatrix} \qquad \textit{eq. 4.}$$

Where $\partial f(u) / \partial (u) = a_u$, $\partial f(u) / \partial v = b_u$, $\partial f(v) / \partial u = a_v$ and $\partial f(v) / \partial v = b_v$, giving;

$$\begin{matrix} \frac{\partial u}{\partial t} \\ \frac{\partial v}{\partial t} \end{matrix} = \begin{pmatrix} a_u & b_u \\ a_v & b_v \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} + \begin{pmatrix} D_u & o \\ o & D_v \end{pmatrix} \begin{pmatrix} \Delta u \\ \Delta v \end{pmatrix} + \begin{pmatrix} c_u \\ c_v \end{pmatrix} \qquad \textit{eq. 5.}$$

This set of non-linear equations can be simplified in the form of a set of linear equations within a rational boundary condition to describe the pattern formation of nanophase-separated alloy systems as followed;

$$\frac{\partial u}{\partial t} = a_u u + b_u v + c_u + D_u \Delta u$$

$$\frac{\partial v}{\partial t} = a_v u + b_v v + c_v + D_v \Delta v \qquad \textit{eq. 6.}$$

with limits of;

$$\frac{\partial u}{\partial t} = 0.2 \; and \; \frac{\partial v}{\partial t} = 0.5$$

The coefficients a_v, and b_u in *eq.* 2 denote a cross coupling of the metal- and oxygen atoms (*eq*. 4);

$$\begin{pmatrix} \frac{\partial u}{\partial t} \\ \frac{\partial v}{\partial t} \end{pmatrix} = \begin{pmatrix} a_u & b_u \\ a_v & b_v \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} \qquad \textit{eq. 7.}$$

$$\begin{pmatrix} \frac{\partial u}{\partial t} \\ \frac{\partial v}{\partial t} \end{pmatrix} = \begin{pmatrix} 0 & + \\ + & 0 \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} ; \begin{pmatrix} \frac{\partial u}{\partial t} \\ \frac{\partial v}{\partial t} \end{pmatrix} = \begin{pmatrix} 0 & - \\ - & 0 \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} ; \begin{pmatrix} \frac{\partial u}{\partial t} \\ \frac{\partial v}{\partial t} \end{pmatrix} = \begin{pmatrix} 0 & + \\ - & 0 \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} \qquad \textit{eq. 7 (a), (b), and (c)}$$

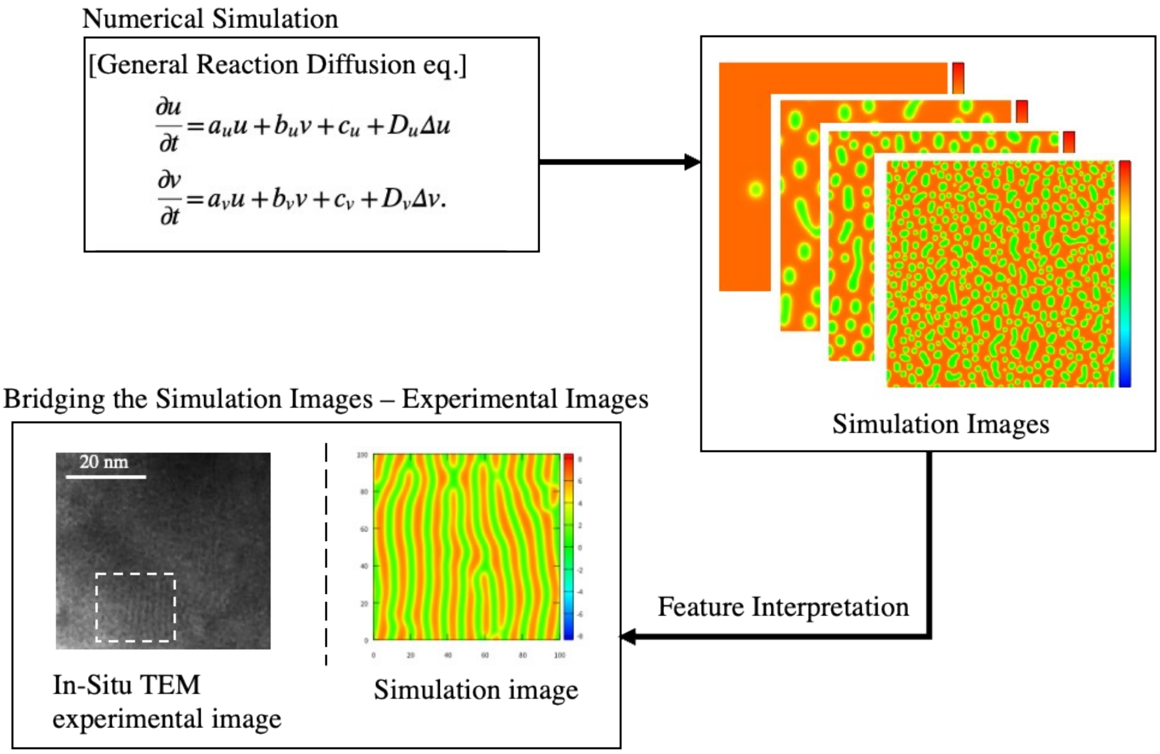

Figure S1: Framework for structure and process in obtaining simulation visuals. Pattern emerges visually generated by calculating interacting parameters of the mathematical model. Computed images from the mathematical model visually assessed to matched the experimental images.

SUPPORTING VIDEO

Video S2(i): *In-Situ* TEM video that recorded the emergence and propagation of lamella pattern of Pt-CeO_2. See the text for details on the experimental conditions.

Video S2(ii): *In-Situ* TEM video that recorded the emergence and propagation of a maze pattern of Pt-CeO_2. See the text for details on the experimental conditions.

Video S2(iii). *In-Situ* TEM video of Pt_5Ce that was subjected to oxygen atmosphere at 400 ^oC. Noticed that there is no pattern emerged which likely due to not enough kinetic energy to initiate oxidation.

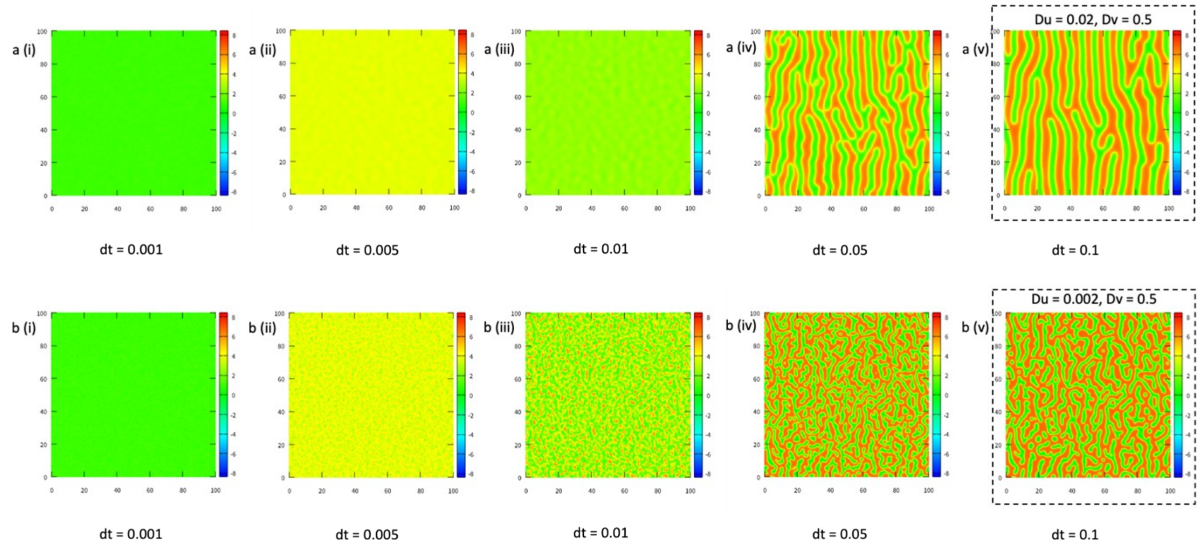

Figure S3: Patterns obtained from simulation by increasing the iteration time, dt from 0.001 to 0.1. Note that a (*v*) and b (*v*) correspond to the Figure 3 (*a* and *b*) in article. Formation of lamellae pattern requires only a slight increase in iteration time from *dt* = 0.01 to *dt* = 0.05 instead of maze pattern which requires a slightly longer iteration time from *dt* = 0.005 to *dt* = 0.05. Keeping the stochastic nature of experiments, in-situ TEM agrees that lamella pattern emerges at faster rate, 5 sec than maze pattern, 120 sec (refer to article).

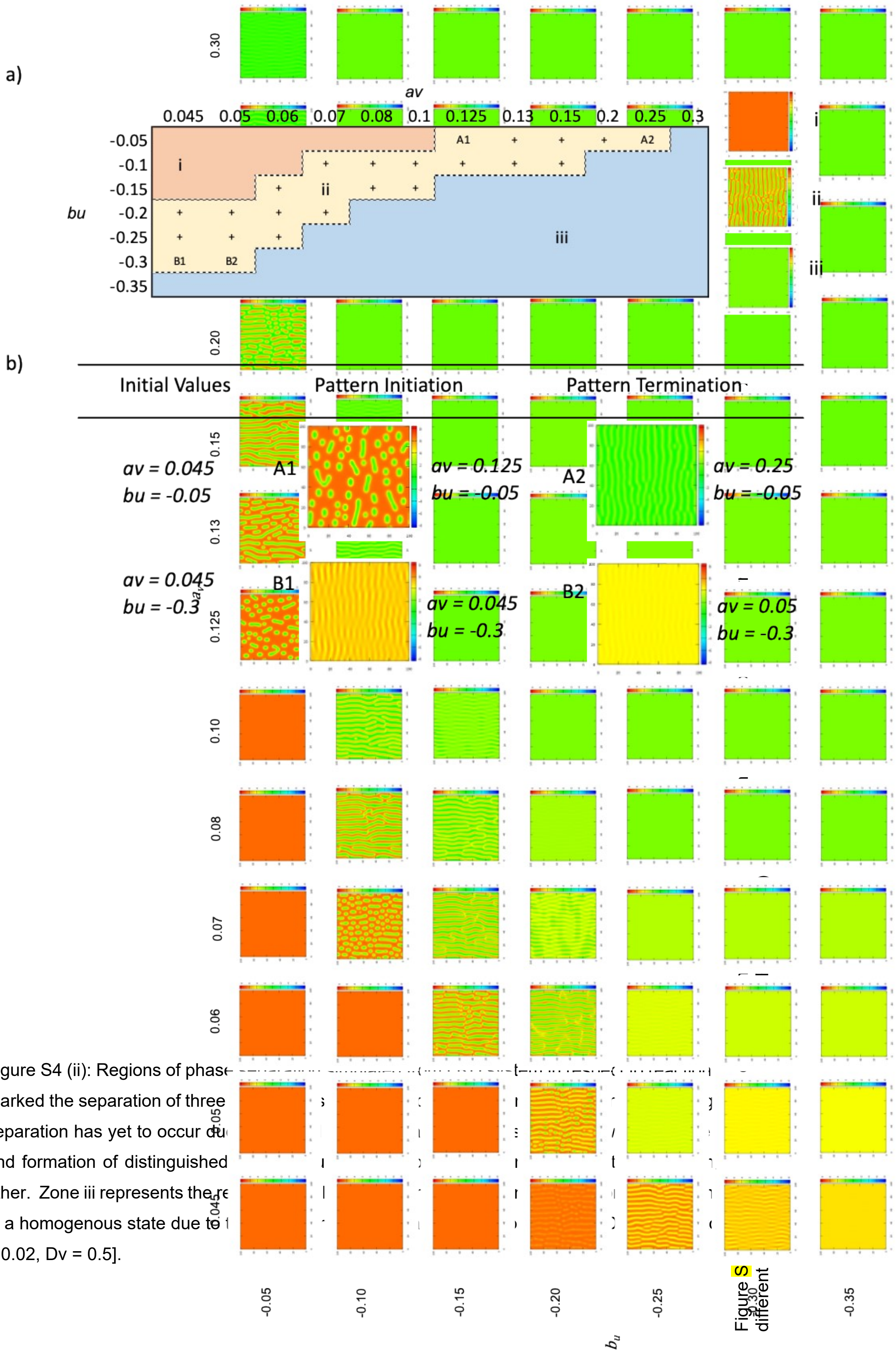

Figure S4 (ii): Regions of phase
marked the separation of three
separation has yet to occur du
and formation of distinguished
other. Zone iii represents the re
in a homogenous state due to
= 0.02, Dv = 0.5].

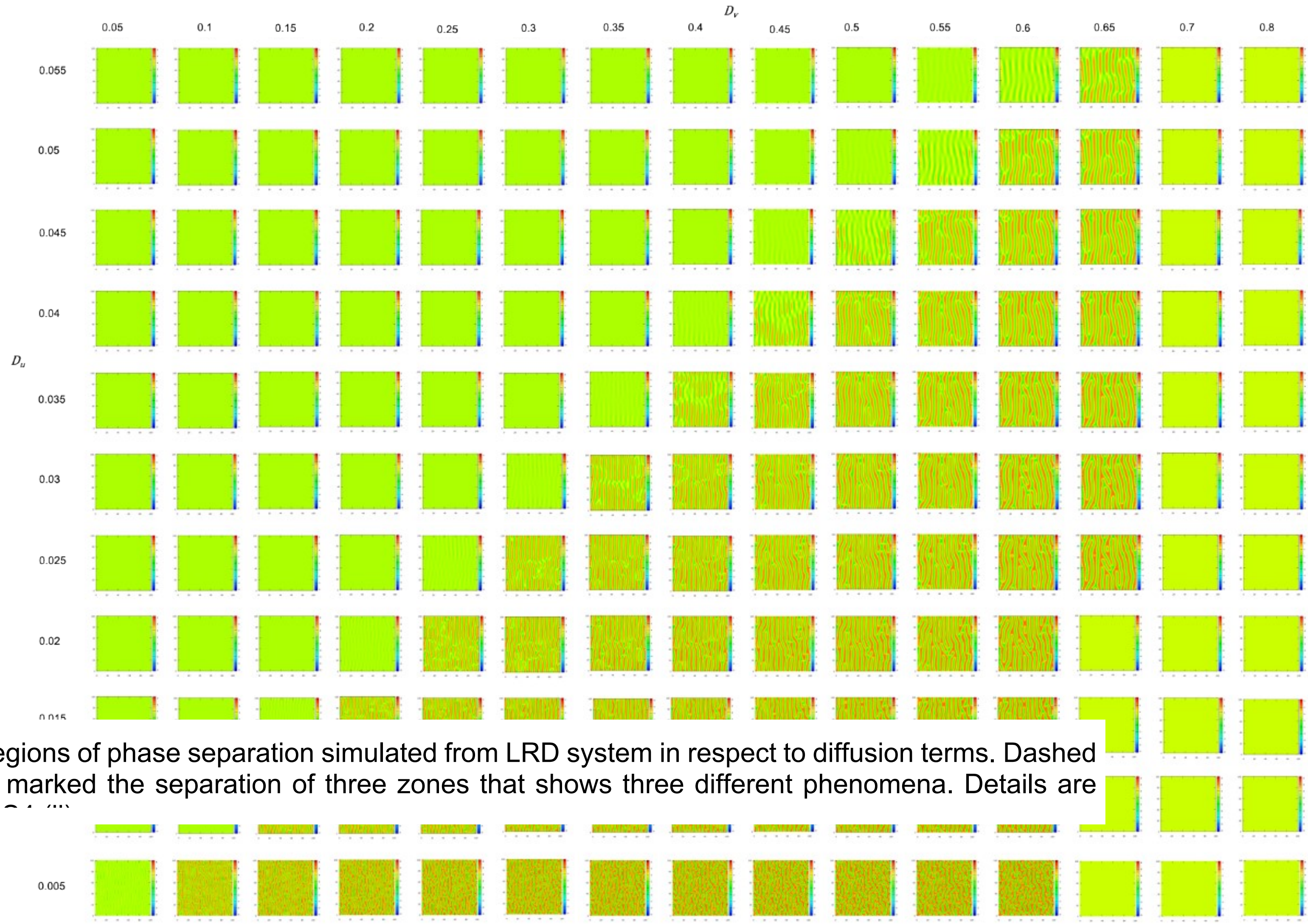

Figure S5 (i): Regions of phase separation simulated from LRD system in respect to diffusion terms. Dashed lines Fig S4 (ii) marked the separation of three zones that shows three different phenomena. Details are

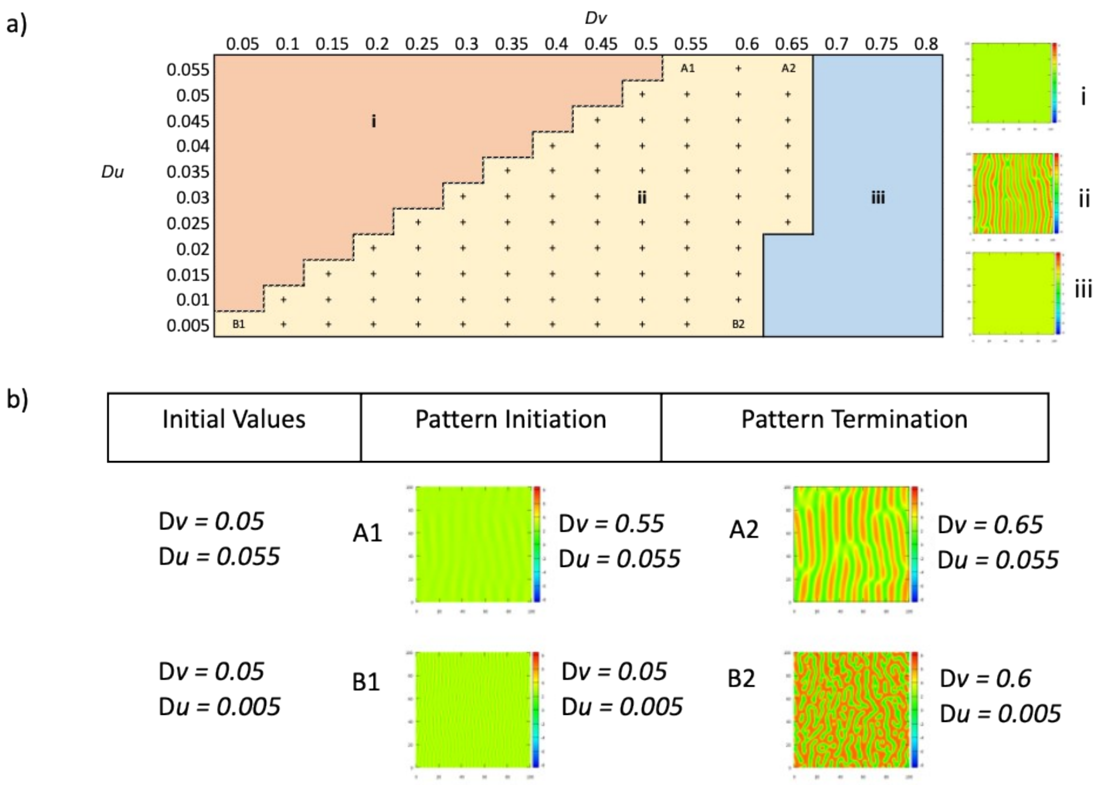

Figure S5(ii): Regions of phase separation simulated from LRD system. Dashed lines marked the separation of three zones that shows three different phenomena. Zone i is the region where the phase separation has yet to occur due to perturbation is not enough to initiate pattern formation. Zone ii is where any perturbation to the system's stability gave a distinguished pattern/structure due to the diffusion of one of the interacting species ousts the other. Zone iii represents the region where large differences between the diffusion rate of *u* and *v* species resulting in a homogenous state due to the rapid conversion/phase separation.

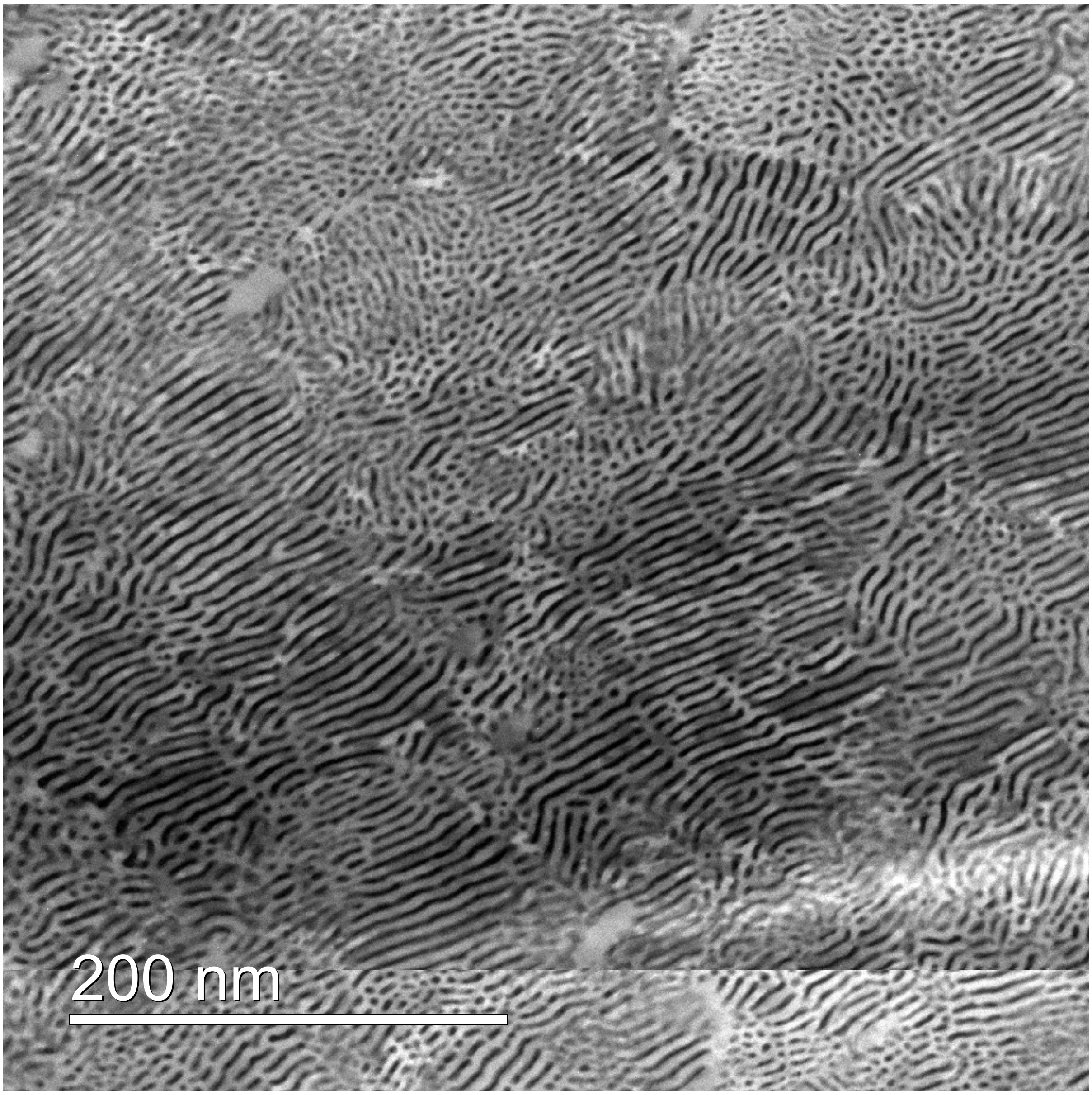

Figure S6: Scanning transmission electron microscope (STEM) image that shows formation of the lamella pattern when Pt_5Ce alloy was subjected to O_2 environment at 600 ^oC. Acquired with JEM-2100F1 at an acceleration voltage of 200 kV.