Supplementary Information for

Optimal thermodynamic conditions to minimize kinetic byproducts in aqueous materials synthesis

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This file includes:

Detailed description of the software code (Synthesis Condition Optimizer) functionality.

1 Software: Synthesis Condition Optimizer

As described in the Results and Methods sections, we defined thermodynamic competition as the difference between the free energy of the target phase and the minimum free energy that can be achieved by combining competing phases, and proved thermodynamic competition is convex considering the linearity of a phase's thermodynamic potential in an aqueous electrochemical system[1, 2]. Thus, we can use gradient-based method to optimize synthesis conditions by minimizing thermodynamic competition. Here, we provide the pseudocode to optimize $\Delta\Phi(Y)$ to find optimum condition Y^*, where Y is an intensive variable such as, pH, in Table 1.

Algorithm: Gradient Descent Method to minimize $\Delta\Phi(Y)$ to find thermodynamic optimal condition

Choose initial Y_0, learning rate η, and maximum step number T

For step $t = 0$ to T, do

$Y_{t+1} \leftarrow Y_t - \eta g(\Delta\Phi_t)$, where $g(\Delta\Phi_t)$ is gradient of the objective function $\Delta\Phi(Y)$ at Y_t

$\Delta\Phi_{t+1} = \Delta\Phi(Y_{t+1})$

End for

Return $Y^* = Y_T$, $\Delta\Phi^* = \Delta\Phi_T$,

where Y^* is the predicted optimal synthesis condition and $\Delta\Phi^*$ is the minimized thermodynamic competition

Table 1: The Pseudocode of gradient descent method

All of the codes used for analyzing coordination environment were based on pymatgen software and its Pourbaix diagrams module[3]. We provide code to calculate the thermodynamic competition and optimize synthesis conditions by minimizing thermodynamic competition here: https://github.com/zherenwang/synthesis_condition_optimizer

References

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[3] Persson, K. A., Waldwick, B., Lazic, P. & Ceder, G. Prediction of solid-aqueous equilibria: Scheme to combine first-principles calculations of solids with experimental aqueous states. *Phys. Rev. B* **85**, 235438 (2012).