Supplementary Information

Simulation Parameters

Table S1. Lock-and-key interaction parameters

Parameters	Lock	Key	Lock-Key
σ	0.5	0.5	0.5
α	6.0	6.0	6.0
Δ	0.33σ	0.0σ	0.165σ
ε	$1.0k_BT$	$1.1k_BT$	$1.75k_BT$
Z	250	520	380

Simulation parameters used for our potential are summarized above. (Table S1) The definition of these parameters are covered in the main text.

Table S2 Overview of Geometric Parameters for the quasi-2D simulation

N_L	D_L	δ	Crystal Structure
1	1.3	0.5	Dis
1	1.3	0.3	Dis
1	1.5	0.3	Dis
1	1.5	0.4	PbO
1	1.5	0.5	PbO
1	1.7	0.5	PbO
2	1.3	0.4	$L_{HH}K_D$
2	1.3	0.5	$L_{HH}K_D$
2	1.5	0.3	Dis
2	1.5	0.4	Dis
2	1.5	0.5	$L_{HH}K_D$
2	1.7	0.3	Dis
2	1.7	0.4	Dis
2	1.7	0.5	$L_{HH}K_D$
3	1.3	0.1	Dis
3	1.3	0.2	Dis
3	1.3	0.3	Dis
3	1.5	0.1	Dis
3	1.5	0.2	Dis
3	1.5	0.3	Dis
3	1.7	0.1	Dis
3	1.7	0.2	Dis
3	1.7	0.3	Dis
4	1.2	0.4	$L_H^4K_D$
4	1.3	0.0	Dis
4	1.3	0.1	Dis
4	1.3	0.2	Dis
4	1.3	0.3	$L_H^4K_D$
4	1.3	0.4	$L_H^4K_S$
4	1.5	0.0	Dis
4	1.5	0.1	Dis
4	1.5	0.2	Dis
4	1.7	0.0	Dis
4	1.7	0.1	Dis
4	1.7	0.2	Dis

Geometric parameters for the Quasi-2D simulation are summarized above. (Table S2) Simulations were performed for the following parameter ranges: lock number (N_L = 1 - 4), lock-key bond distance ($0.0 < \delta < 0.6$), and size ratio ($1.2 < D_L < 1.7$). The majority of the geometric state points shown above are disordered (Dis) The crystals isostructural to PbO, and $L_{HCP}K_D$ are summarized in Figure 8.