Electronic supplementary information (ESI)

Molecular simulation study on the flexibility in the interpenetrated metal–organic framework LMOF-201 using reactive force field

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Table S1 L-J parameters for LMOF-201 atoms.

Atom	ε/k_B	σ(Å)	Reference
Zn	62.4	2.46	UFF[1]
C	47.8562	3.47	Dreiding[2]
N	38.94	3.26	Dreiding[2]
O	48.15	3.03	Dreiding[2]
H	7.64	2.84	Dreiding[2]

Table S2 L-J parameters and charges for adsorbate atoms.

Atom	ε/k_B (K)	σ(Å)	q(e)	Reference
C_CO_2	27.0	2.8	0.7	3
O_CO_2	79.0	3.05	−0.35	3

Additional simulation details

The reactive force field (ReaxFF) is a bond order dependent force field, which is capable of providing accurate modeling of chemical reactions with relatively low calculation cost. In their seminal paper, van Duin et al. demonstrated that ReaxFF did not require any connectivity parameters and the cost of computation was low compared to that of typical first-principles calculations.[4] The bond order is calculated instantaneously based on inter-atomic distances; furthermore, it includes both van der Waals and Columbic interactions. The ReaxFF parameters are trained based on rigorous comparison with density functional theory (DFT) results.

Molecular dynamics (MD) simulations were performed to obtain the structural change of LMOF-201 due to CO_2 adsorption in a quasi-static process at fixed temperature and pressure. It was found that the system was not at adsorption equilibrium under this condition, that is, the calculated volume change was not exactly the same as the volume change along the adsorption isotherm. However, we assumed that the unit cell dimension of LMOF-201 was sensitive to the CO_2 amount, while it was not as sensitive to the pressure, unless the pressure was extremely high. Under this assumption, it was expected that the equilibrium unit cell dimension calculated at fixed CO_2 amount, pressure and temperature was similar to that calculated at the same CO_2 amount and temperature, but at a different pressure.

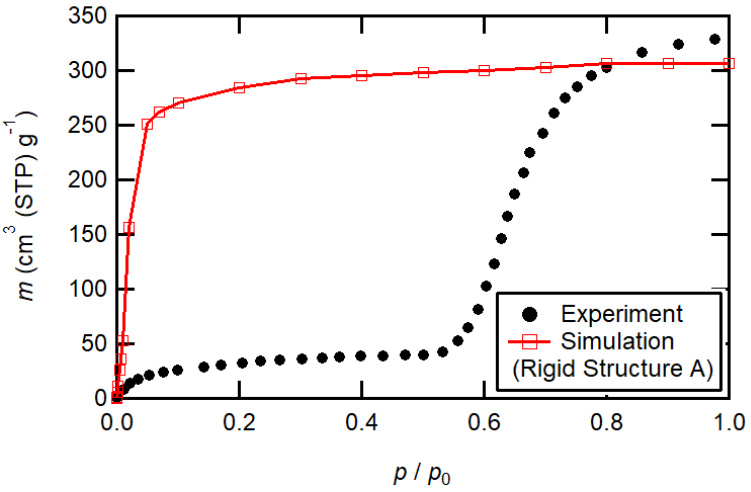

Fig. S1 Calculated adsorption isotherm of CO_2 in LMOF-201 of rigid structure A at 195 K and the experimental adsorption isotherm under the same condition.

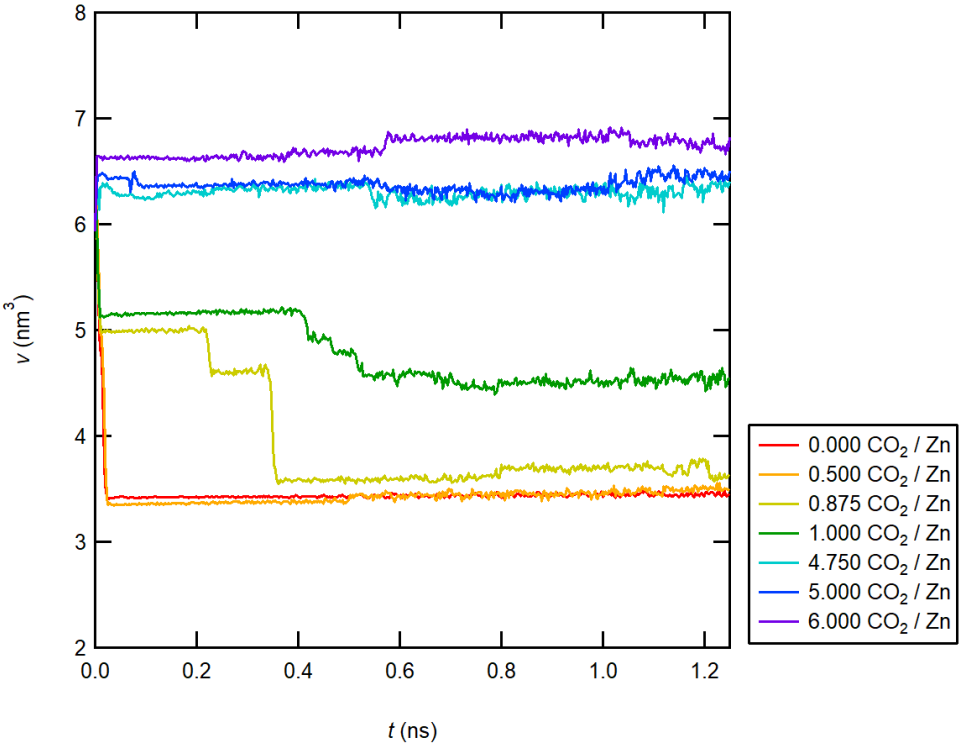

Fig. S2 Variation of the unit cell volume of LMOF-201 for different of CO_2 amounts.

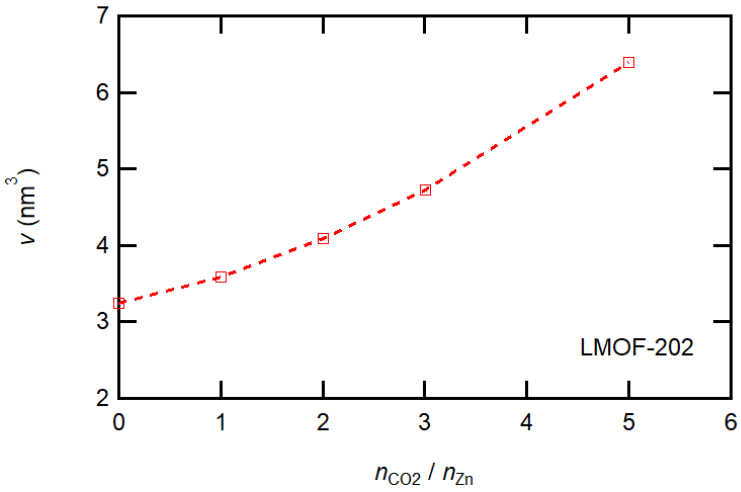

Fig. S3 Progression of the LMOF-202 unit cell volume with CO_2 loading at 195 K.

References

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