Supplementary Information

1 Langmuir and Freundlich equations fitting comparison

Table 1
Langmuir parameters of CO at different temperatures

Temperature/ K	KCK model			TriK model		
	a/(mL·g^{-1})	*b*/(MPa^{-1})	R^2	*a*/(mL·g^{-1})	*b*/(MPa^{-1})	R^2
293.15	44.09	0.06	0.9985	25.99	0.09	0.9983
303.15	40.24	0.05	0.9975	23.67	0.09	0.9992
313.15	35.10	0.06	0.9992	23.20	0.08	0.9976
323.15	35.06	0.06	0.9984	23.12	0.08	0.9971
333.15	32.55	0.06	0.9984	22.28	0.07	0.9990

Table 2
Freundlich parameters of CO at different temperatures

Temperature/ K	KCK model			TriK model		
	a/(mL·g^{-1})	*b*/(MPa^{-1})	R^2	*a*/(mL·g^{-1})	*b*/(MPa^{-1})	R^2
293.15	2.58	0.80	0.9992	2.20	0.85	0.9991
303.15	2.29	0.83	0.9990	1.76	0.94	0.9995
313.15	2.07	0.90	0.9982	1.52	0.95	0.9985
323.15	1.93	0.91	0.9989	1.97	0.90	0.9985
333.15	1.80	0.94	0.9996	1.55	1.04	0.9992

2 The fitting parameters of the Freundlich equation for the adsorption capacity of Al-O and Si-O planes

Table 3
Freundlich parameters of CO at 2 planes of kaolinite

Temperature/ K	Al-O			Si-O		
	a/(mL·g^{-1})	*b*/(MPa^{-1})	R^2	*a*/(mL·g^{-1})	*b*/(MPa^{-1})	R^2
293.15	1.69	0.66	0.9771	1.67	0.55	0.9928
303.15	1.56	0.62	0.9222	1.23	0.80	0.9796
313.15	1.56	0.62	0.9222	1.23	0.80	0.9796
323.15	1.62	0.59	0.9423	1.02	1.09	0.9315
333.15	0.80	1.62	0.9601	1.38	0.46	0.9126

3 Free volume

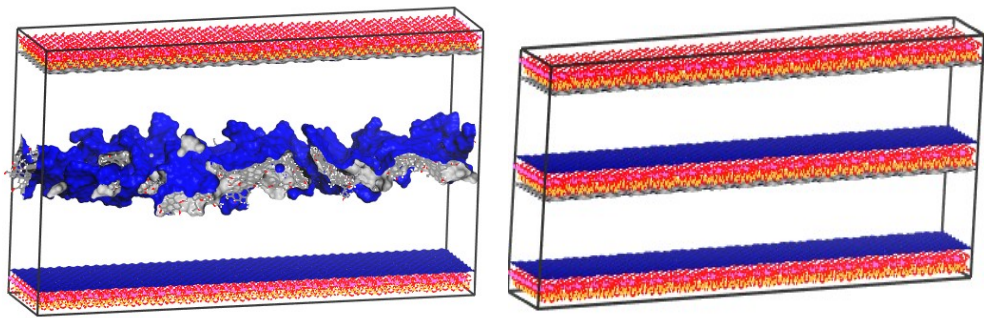

Fig.1 The Connelly surface of both models
KCK Free Volume: 384701.12 Å^3, TriK Free Volume: 246018.35 Å^3.(Connelly radius 0.184nm, Grid interval 0.04nm)

The extra free volume mainly comes from the pores of the layer carbon-containing structure.

4 Module introduction

Forcite: It is a classic molecular dynamics tool, developed from the discover module, which is used to perform a range of computational tasks, including fast energy calculations and geometric optimization for both monomolecular and periodic systems.

Qeq: Charge Equilibration is used in the calculation.

Amorphous Cell: It is a tool that uses the Monte Carlo method to set up the amorphous model. It can be used to build models with various components and different proportions.

Atom Based : It is a direct calculation method that directly calculates the non-bonding interactions between pairs of atoms. When the distance between pairs of atoms exceeds the set cutoff radius, the system considers the interaction between the pairs to be zero.

5 Distribution of CO adsorption density

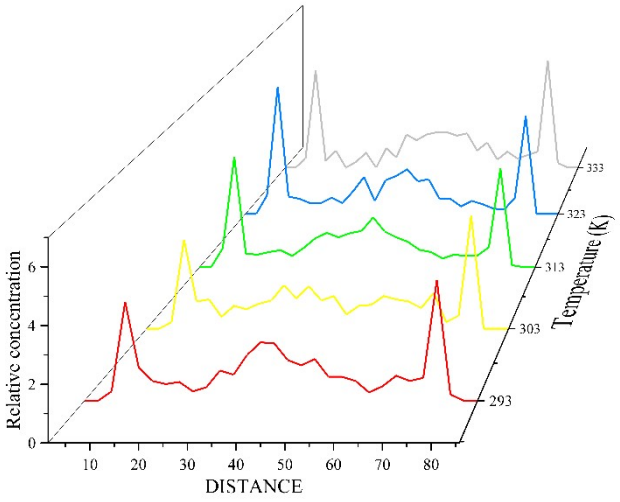

Fig.2 Relative density distribution curve of 1MPa

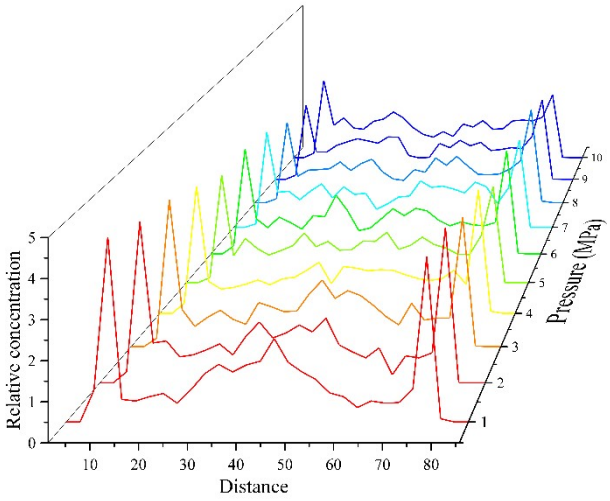

Fig.3 Relative density distribution curve of 313.15K

It can be seen that the shape of the density distribution curve is similar at the same temperature point and pressure point.

However, as more CO molecules are adsorbed, a large number of CO molecules fill the vacuum layer, causing the peak in the middle part to be less obvious.