Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

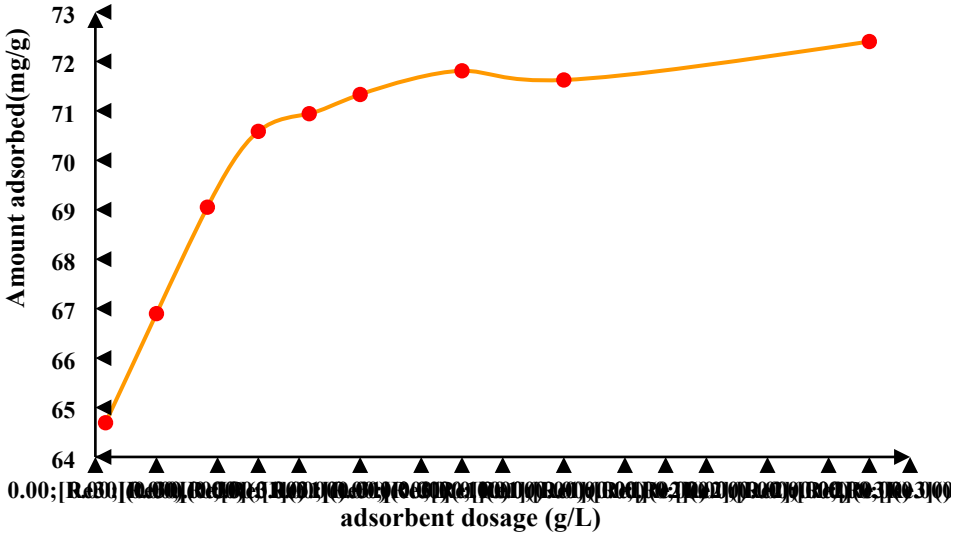


Figure S1 The effect of adsorbent dosage on the adsorption performance.

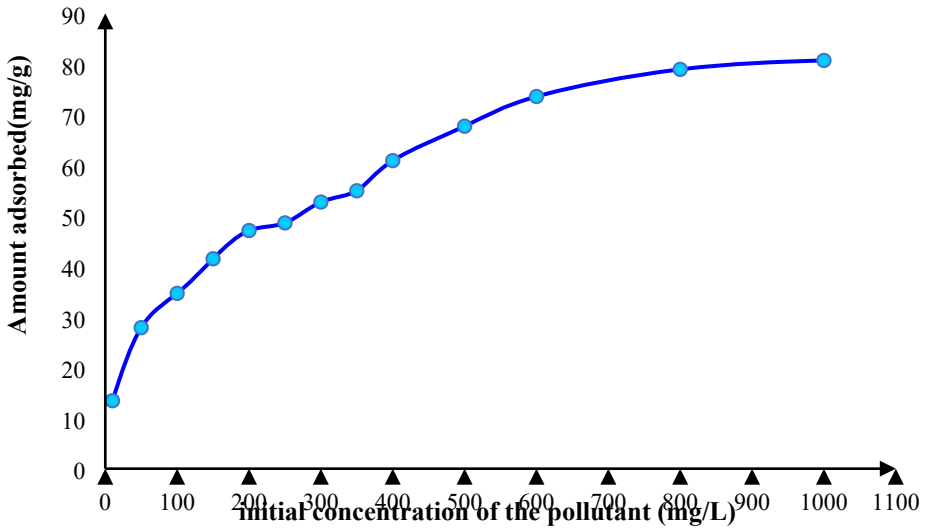
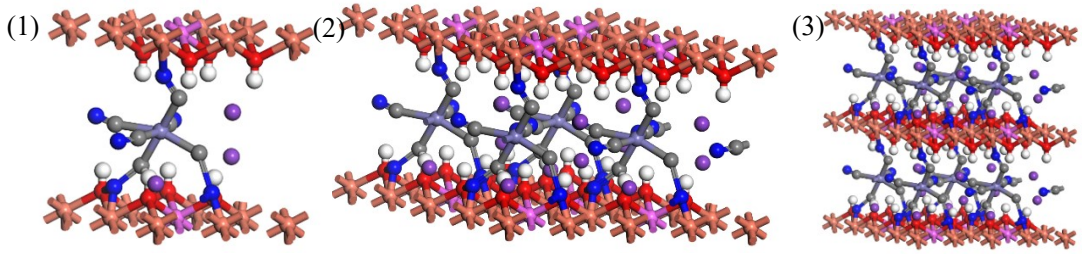


Figure S2 The effect of initial concentration of the pollutant on the adsorption performance.

After optimizing the structures of models with supercell parameters of 2×2×1, 4×4×1, and 4×4×2, CAL-441 was chosen as the base model for subsequent calculations due to its relatively efficient computational requirements while maintaining accuracy. The parameters of CAL-441 are shown in Supplementary Table S2.



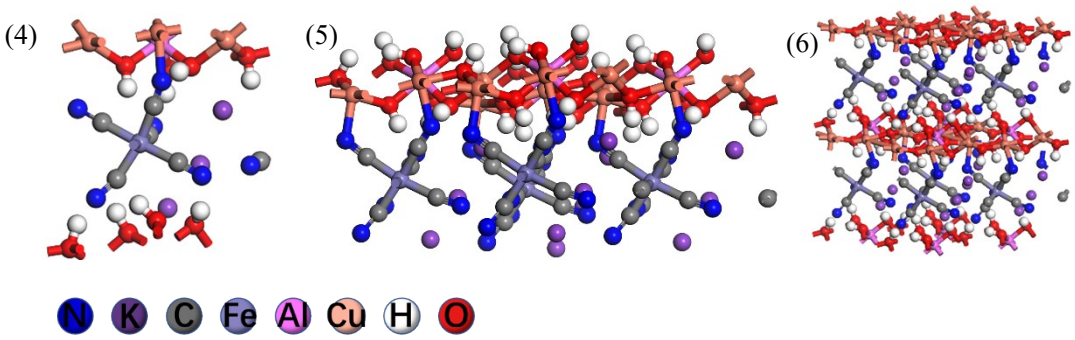


Figure S3 Initial guess models and optimized models for CAL-221 (1)(4), CAL-441 (2)(5), and CAL-442 (3)(6).

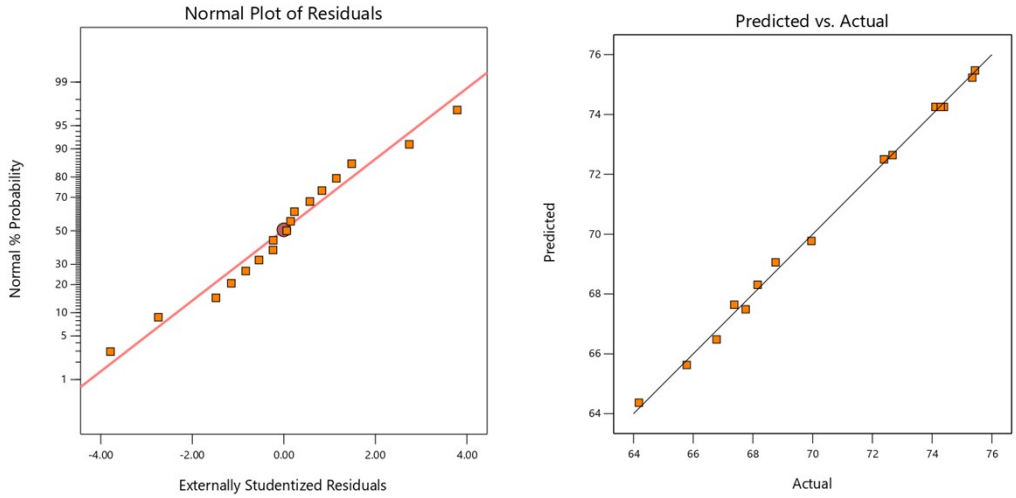


Figure S4 Normal % probability versus internally studentized residuals and experimental data versus predicted data.

Table S1 Response surface factors and levels

Factor	Name	Units	Minimum	Maximum	Mean
A	pH		9	11	10
B	temperature	℃	25	35	30
C	pollutant concentration	mg/L	200	500	350

Table S2 Parameters of CAL-441

Parameters	Value
Total Atoms	144
Formula Unit	$Cu_3H_8O_8AlFeC_6N_6K_3$
Full Formula Unit	$Cu_{12}H_{32}O_{32}Al_4Fe_4C_{24}N_{24}K_{12}$
Lattice Constants (Å)	12.662 12.662 8.700

Lattice Angles (°)	90.000	90.000	120.000
Volume (Å^3)		1207.946	

To verify the feasibility of interlayer anion exchange, the binding energies of nitrate and ferrocyanide insertion into hydrotalcite were calculated as follows:

$$E_b = E_{tot} - E_M - E_{none}$$

E_b represents the binding energy of the inserted anionic group in hydrotalcite, E_{tot} denotes the overall energy of the structure after the insertion of the anionic group, E_M refers to the energy associated with the anionic group, and E_{none} represents the total energy of the defective structure of hydrotalcite without the anionic group.

在优化好的结构基础上（CAL-441）进行铁氰基的结合能计算，得到的结果如 Table S3 所示。

Table S3 Calculation of binding energy of nitrate and ferricyanide inserted into hydrotalcite

Functional group	**E_{tot} (eV)**	**E_{none} (eV)**	**E_M (eV)**	**E_b (eV)**
NO_3	-141.1854	-114.524	-23.0032	-3.6582
$K_3Fe(CN)_6$	-834.3953	-392.084	-432.3346	-9.9767

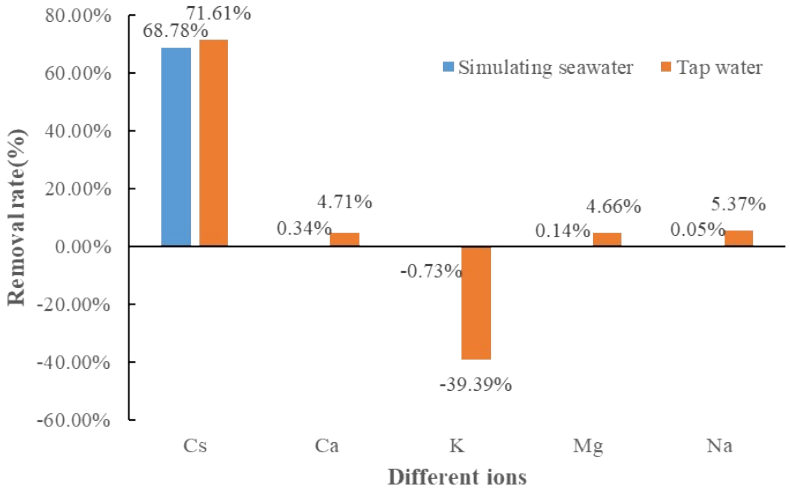
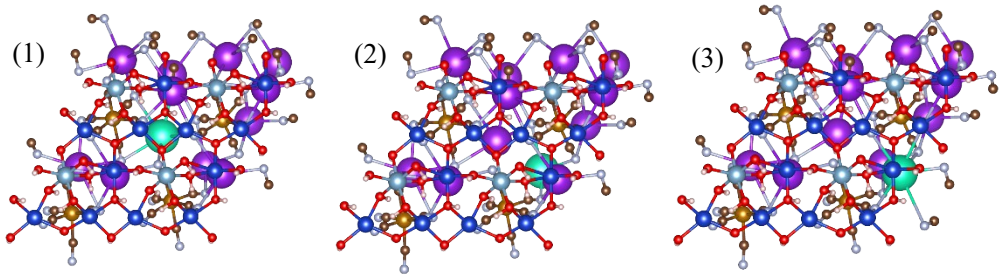


Figure S5 Removal rates of Cs from multicomponent simulating seawater and tap water.



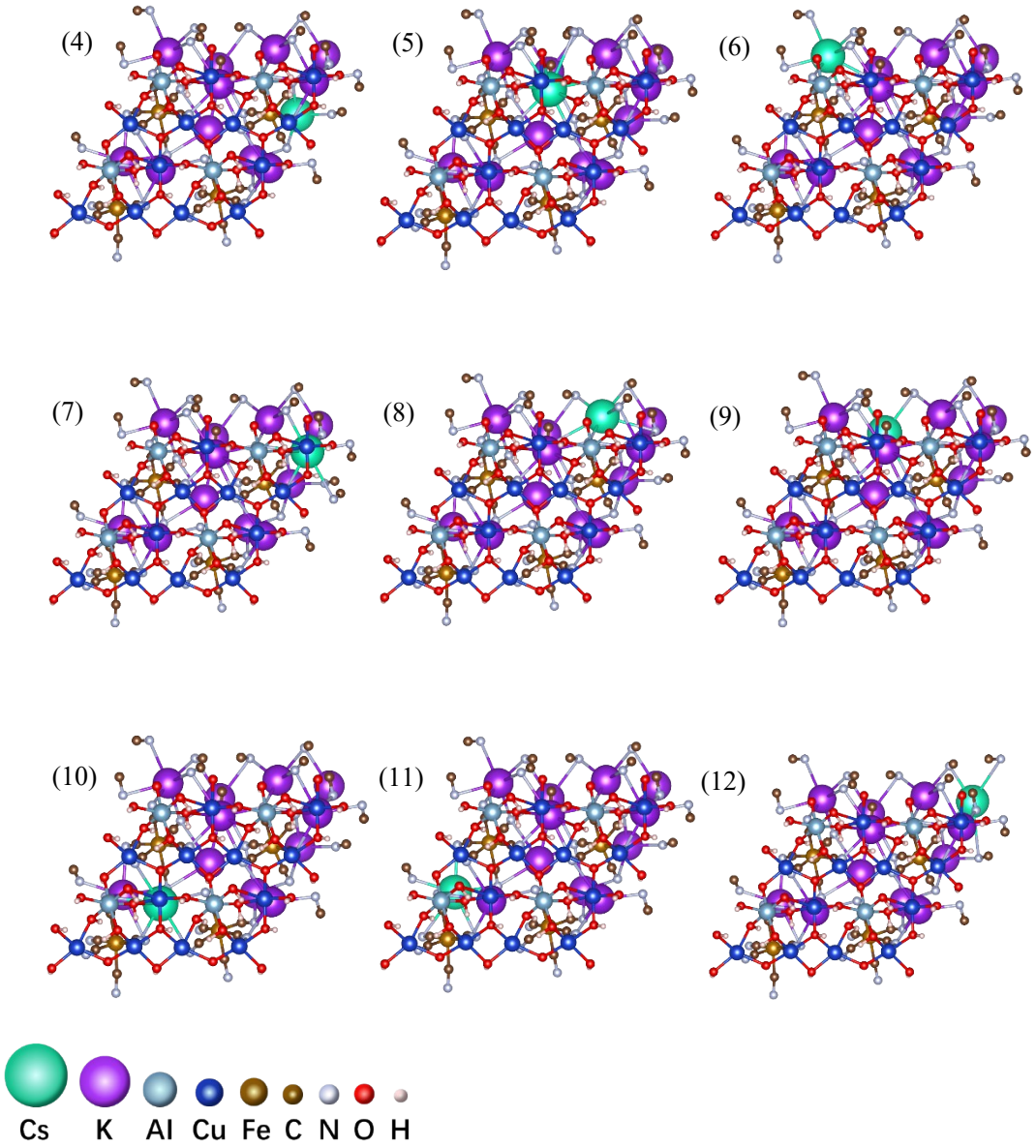


Figure S6 Computational models for the substitution of K atoms with Cs atoms in twelve possible Cs doping sites.

Table S4 process of combining energy calculations

unit：eV

	doping sites	E_{tot}	E_{none}	Binding energy of Cs_i	Binding energy of K_i	Exchange energy	E_{Cs}	E_K
CAL-441-slab	-	-863.75	-	-	-	-	-	-
CAL-441-Cs1	K1	-864.74	-860.49	-4.21	-3.22	-0.99	-0.0436	-0.0370
CAL-441-Cs2	K2	-865.09	-861.35	-3.70	-2.36	-1.33	-0.0435	-0.0371
CAL-441-Cs3	K3	-864.63	-860.89	-3.69	-2.82	-0.88	-0.0439	-0.0371
CAL-441-Cs4	K4	-873.05	-869.35	-3.66	5.64	-9.29	-0.0436	-0.0371

CAL-441-Cs5	K5	-873.33	-870.95	-2.33	7.24	-9.57	-0.0441	-0.0373
CAL-441-Cs6	K6	-872.85	-869.5	-3.31	5.79	-9.10	-0.0440	-0.0373
CAL-441-Cs7	K7	-873.50	-870.1	-3.36	6.39	-9.75	-0.0441	-0.0373
CAL-441-Cs8	K8	-873.46	-870.16	-3.26	6.45	-9.71	-0.0439	-0.0373
CAL-441-Cs9	K9	-874.02	-869.95	-4.02	6.24	-10.26	-0.0442	-0.0373
CAL-441-Cs10	K10	-873.49	-869.97	-3.47	6.26	-9.73	-0.0441	-0.0373
CAL-441-Cs11	K11	-874.22	-870.16	-4.02	6.45	-10.47	-0.0443	-0.0373
CAL-441-Cs12	K12	-873.79	-870.25	-3.50	6.54	-10.04	-0.0441	-0.0373

Table S5 Parameters of the kinetic model

Kinetic models	**parameters**	**LDH**	**PB-LDH**
pseudo_first_order	q_e(mg/g)	30.1518	111.081
	k1 (min-1)	0.02372	0.01670
	R^2	0.98809	0.94668
pseudo_second_order	q_e(mg/g)	32.6736	123.412
	k2 (g/(mg•min))	0.00099	0.00017
	R^2	0.98355	0.98891
Eovich	α ×10-3 (mg/(g•min))	3.60627	8.72238
	β (g/mg)	0.19289	0.05069
	R^2	0.87677	0.93524

Table S6 Parameters of the isothermal adsorption model

Isothermmodels	**parameters**	**LDH**			**PB@LDH**		
		288K	**298K**	**308K**	**288K**	**298K**	**308K**
Langmuir	q_{max}(mg/g)	30.3272	34.8353	37.5606	72.9721	89.4354	109.221
	K_L(L/mg)	0.00448	0.00509	0.00846	0.00975	0.00772	0.00582
	R^2	0.98914	0.93761	0.99247	0.82250	0.88508	0.89662
Freundlich	$K_F(L^n/g\cdot mg^{n-1})$	1.37995	3.16918	3.98149	10.1035	10.3304	9.10797
	1/n	0.43376	0.33129	0.32472	0.28286	0.30361	0.34446
	R^2	0.94469	0.80131	0.92526	0.98307	0.98064	0.99572
Tempkin	A	6.70030	8.04500	7.97518	12.4534	15.3512	18.9051
	$K_T(mM^{-1})$	0.04388	0.04829	0.08928	0.22867	0.17609	0.12572
	R^2	0.98604	0.91129	0.98511	0.93066	0.93607	0.92826

Dubinin-Radushkevich	qs(mg/g)	20.9672	27.6100	29.2187	58.6068	69.2992	79.0707
	$K_{DR}(mol^2/kJ^2)$	0.00159	0.00175	0.00069	0.00057	0.00076	0.00087
	R^2	0.83881	0.96832	0.82332	0.35994	0.49343	0.52492

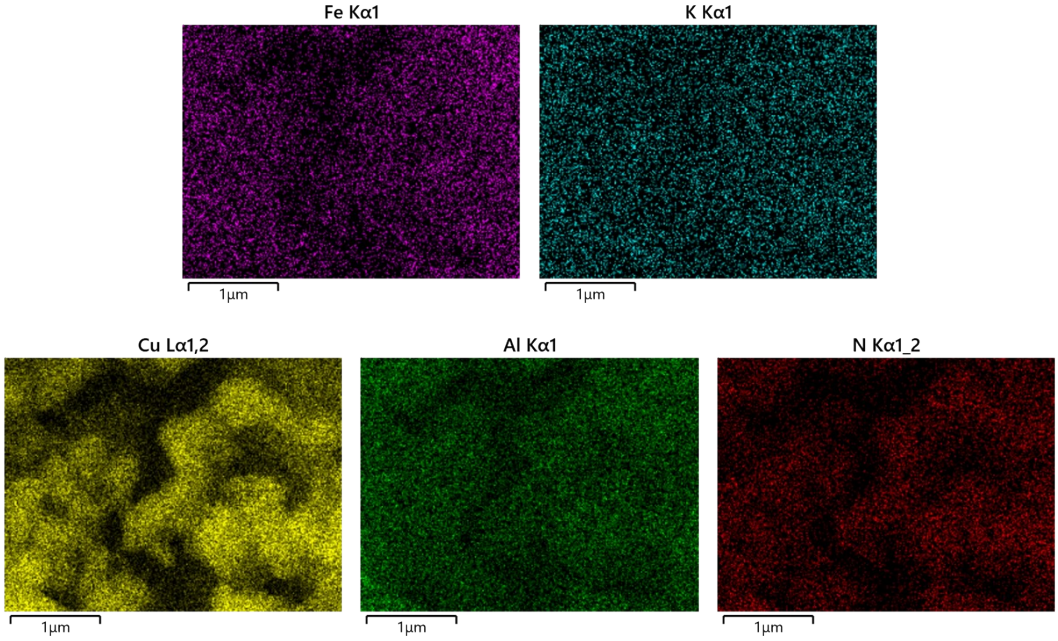


Figure S7 EDS mapping image before adsorption

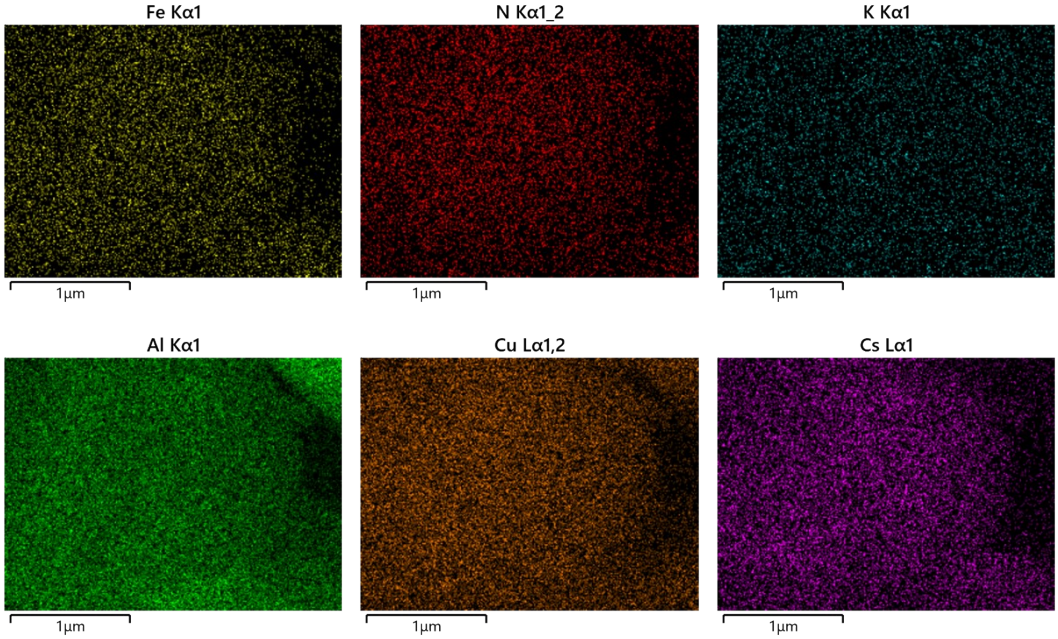


Figure S8 EDS mapping image after adsorption