Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary material

Design and development of 3D hierarchical ultra-microporous CO_2-sieving carbon architectures for potential flow-through CO_2 capture at typical practical flue gas temperatures

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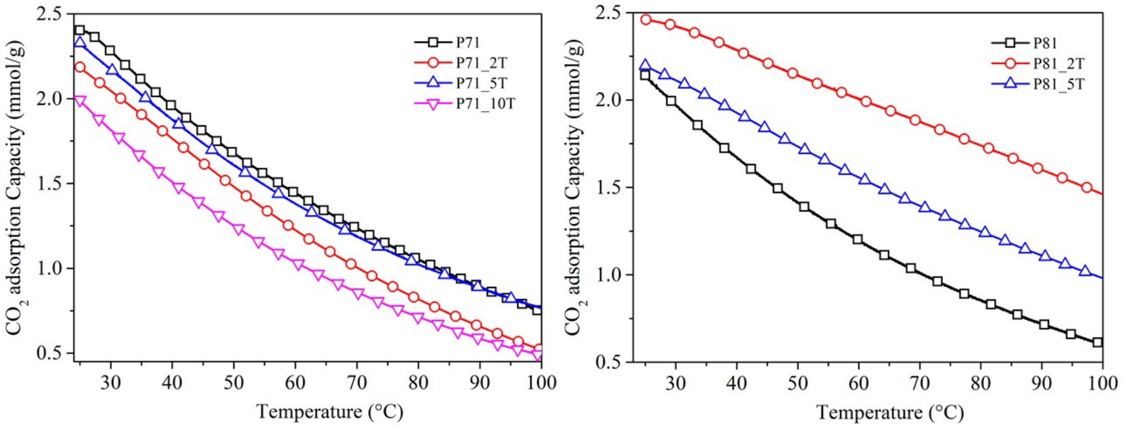
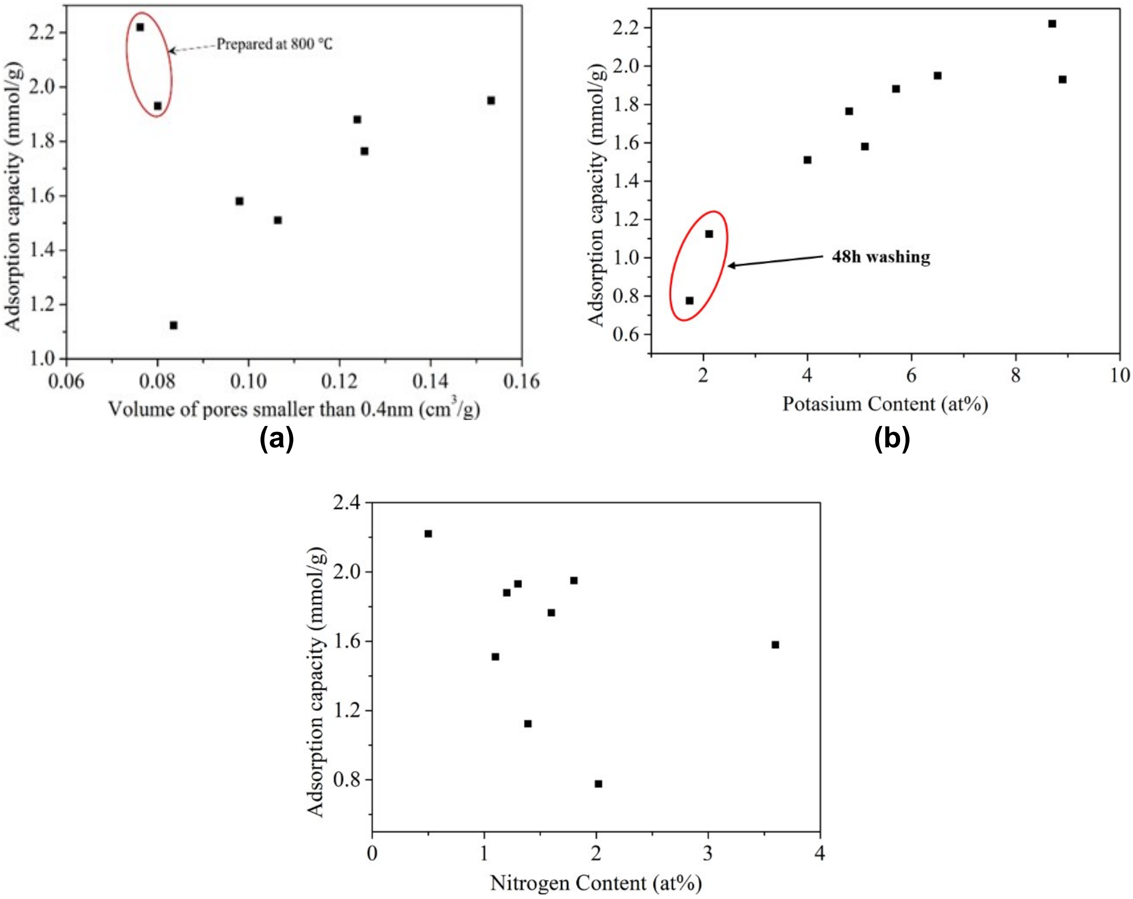


(a) (b)

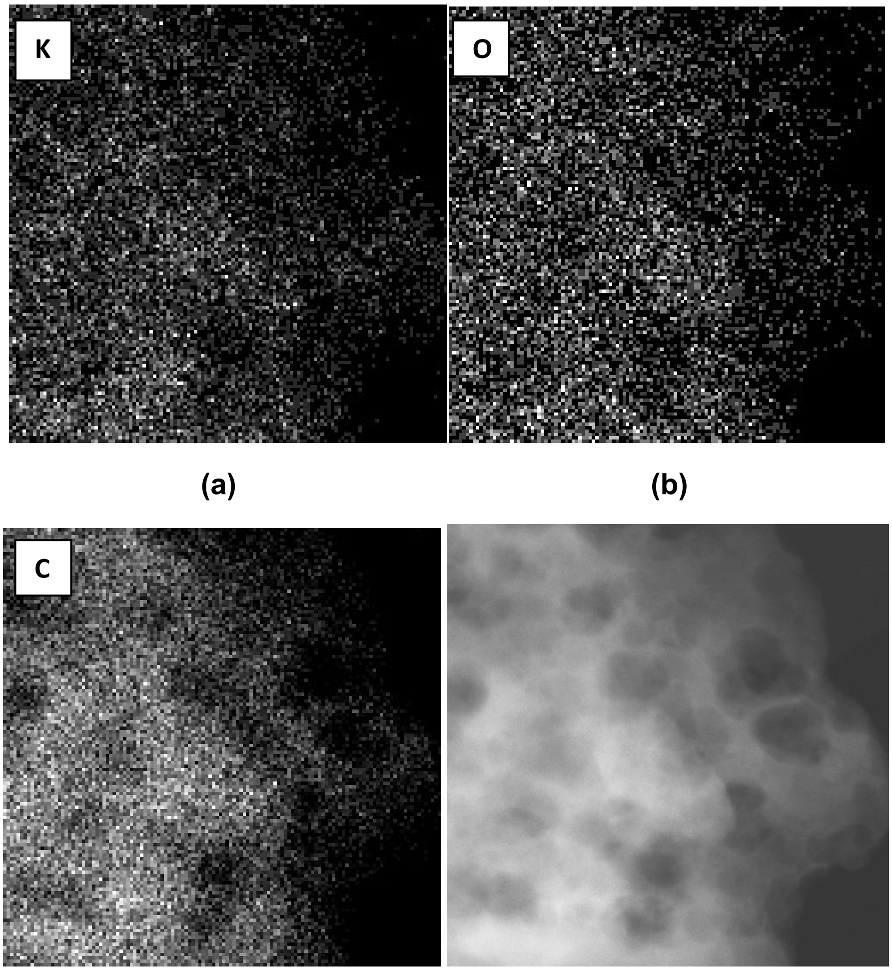
Figure S1 CO_2 adsorption profiles: the change of CO_2 adsorption capacity of PIR carbons with increasing adsorption temperature in simulated flue gas condition (15% CO_2 + 85% N_2): (a) activation temperature 700 °C; (b) activation temperature 800 °C



(a) **(b)**

(c)

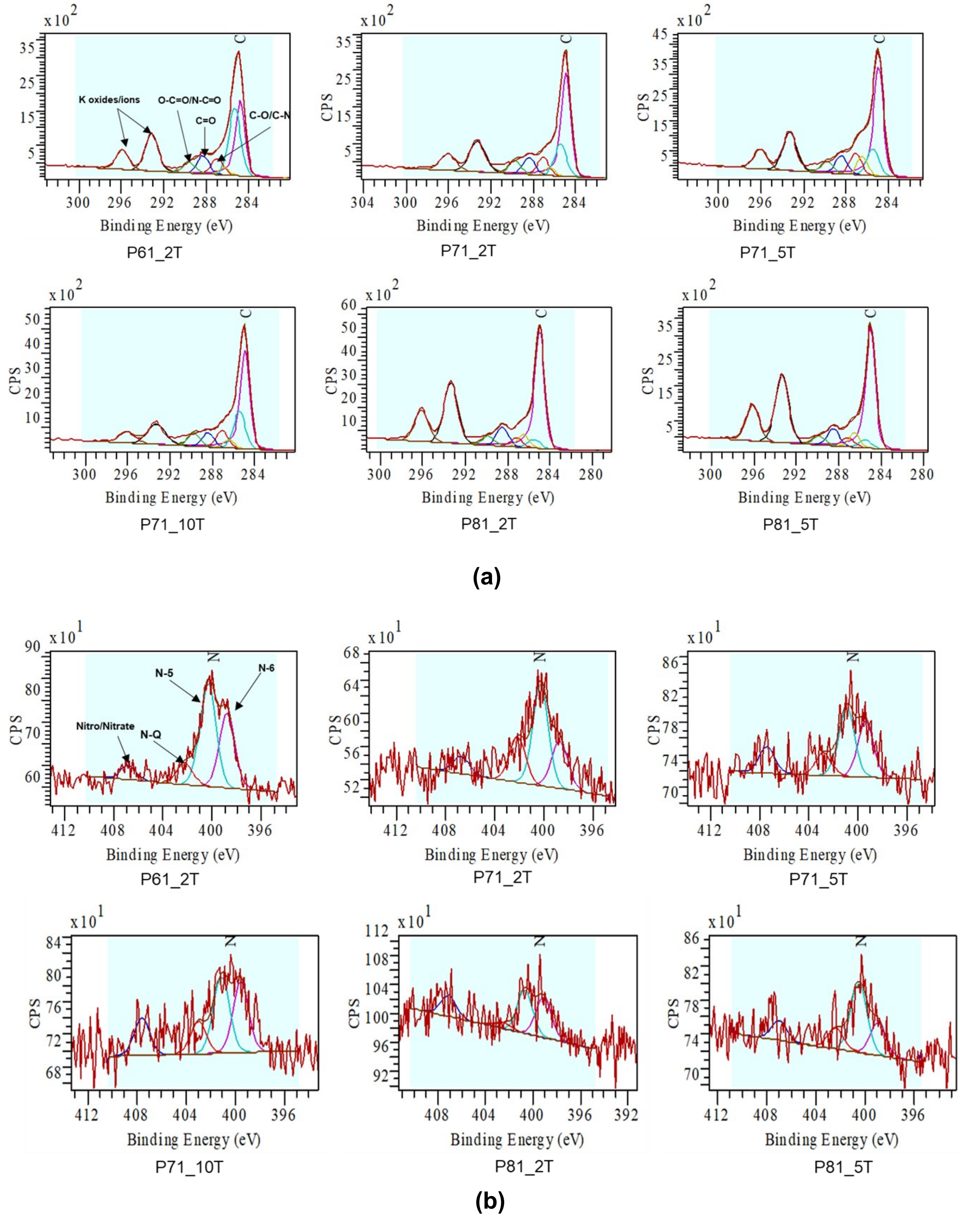
Figure S2 The underlying role of surface chemistry and porous structures of the PIR carbons in CO_2 adsorption: (a) relation between the ultra-micropore volumes and CO_2 capacity; (b) relation between CO_2 uptake and the content of intercalated potassium in the carbons;(c) relation between CO_2 uptake and the content of nitrogen in the carbons



(a) (b)

(c) (d)

Figure S3 Elemental mapping of sample P71_2T: (a) potassium; (b) oxygen; (c) carbon; (d) image of scanning area



(a)

(b)

Figure S4 XPS spectra of the PIR carbons (a) C1s (b) N1s

Table S1 CO_2 adsorption capacity of activated carbon materials at 0.15 bar CO_2

Precursors	N (wt%)	Adsorption capacity at 0.15 bar			Ref
		25-30 °C	40 °C	50 °C	
Phenolic resin	0	1.50	--	0.80	S1
Graphene	0	0.50	--	0.32	S2
Lignin	0	1.20	--	0.50	S3
Starbon	0	0.93	--	0.64	S4
Epoxy resin	0	0.66	--	0.50	S5
Polyvinylidene fluoride	0	1.25	--	0.68	S6
Sawdust derived carbon	0	1.20	--	0.50	S7
Lignocellulosic feedstock	0	1.20	--	0.75	S8
mango fruit(Mangifera indica L.) seed shells	0	1.13	--	0.7	S9
N-enriched carbon monoliths	3.38	1.51	--	1.00	S10
Chitosan	4.59	1.10	--	0.65	S11
Indole-3-butyricacid potassium	4.98	1.23	--	0.66	S12
urea-formaldehyde resin	5.00	1.20	--	0.50	S13
Polypyrrole	5.80	1.70	--	1.00	S14
IRMOF-3	7.00	0.95	--	0.45	S15
Polypyrrole	10.14	0.92	--	0.50	S16
p-diaminobenzene	12.91	1.43	--	0.75	S17
Benzimidazole	17.60	2.03	--	1.00	S18
om-ph-MR	18.16	0.80	--	0.45	S19
ZIF-8	25.52	1.45	--	0.80	S20
Urea formaldehyde resin	26.27	0.73	--	0.52	S21
urea formaldehyde resin	5.62	1.40	--	1.00	S22
Activated carbon	0	0.55	0.43	0.39	S23
Starch	0	0.55	0.34	--	S24
Phenolic resin	0	1.34	--	--	S25
PVDC-methyl acrylate	0	1.16	0.66	--	S26
mangosteen peel waste	0	2.00	1.00	--	S27
N-doped Pitch	5.28	1.10	0.70	--	S28
Benzimidazole-Linked Polymer	7.88	2.10	1.40	--	S29
Dicyandiamide and F127	13.10	0.98	0.66	--	S30
Melamine–formaldehyde resin	27.20	1.50	1.10	--	S31
Olive stones	0.20		0.65	--	S32
Graphene oxide	0	0.60			S33
Carbon-rGO	0	1.00			S34
Graphene	0	1.07			S35
Graphene	0	0.85			S36
Tar pitch and coal powder	0	1.27			S37
Petroleum pitch	0	0.90			S38
Waste Coca Cola R	0	1.36			S39
Sucrose	0	0.84			S40
Chestnut tannin	0	0.93			S41
Celtuce leaves	0	0.95			S42
Phenolic resin	0	0.80			S43

Precursors	N (wt%)	Adsorption capacity at 0.15 bar			Ref
		25-30 °C	40 °C	50 °C	
Phenolic resin	0	1.25			S44
Microporous organic polymers MOP8-MOP10	0	0.50			S45
Phenolic resin	0	0.43			S46
ion exchange resin	0	0.90			S47
Reduced graphene oxide/poly-thiophene	0	1.32			S48
Phenolic resin	0	0.70			S49
Phenolic resin/carbon nanotubes	0	1.18			S50
Glucose	0	0.82			S51
Dicyandiamide/glucose or melamine/glucose	0	1.60			S52
Coconut shell	0	1.34			S53
Potassium hydrogen phthalate derived carbon	0	1.60			S54
Jujun grass derived carbon	0	1.50			S55
Granular Bamboo-Derived Activated Carbon	0	1.30			S56
Phenolic resin spheres (CS-8)	0	1.30			S57
Waste coffee ground derived carbons	0	1.20			S58
Carboxylic phenolic resins	0	1.10			S59
cellulose fibers	0	0.90			S60
Cross-linked microporouscarbon beads	0	1.35			S61
polythiophene	0	0.94			S62
Sucrose	0	1.00			S63
lotus stem waste	0	1.05			S64
acrylic acid + glucose	0	1.33			S65
d carbon black	0	1.50			S66
Coffee	0	1.10			S67
coconut shell	0	0.99			S68
Benzidine	0	1.00			S69
Vine shoots	0	1.35			S70
Coconut shell derived carbon	0.20	1.40			S71
Bean dreg derived carbon	0.28	1.40			S72
Pine cone	0.50	1.64			S73
Coconut shell	0.91	1.45			S74
MOF-5	0.94	0.75			S75
Pitch-Based Carbon Spheres	1.10	1.86			S76
d-glucose and aniline	1.20	1.10			S77
Phenolic resin	1.51	1.36			S78
Nitrogen-containing carbon spheres	1.60	1.48			S79
Popcorn	1.62	1.20			S80
Urea and petroleum coke	1.64	1.27			S81
Imine-linked polymer	1.73	1.07			S82
Phenolic resin	1.92	1.30			S83

Precursors	N (wt%)	Adsorption capacity at 0.15 bar			Ref
		25-30 °C	40 °C	50 °C	
Nitrogen-Doped Porous Carbon Monolith	1.92	1.27			S84
Poly(ammonium-4-Styrenesulfonate)	2.08	0.84			S85
Carboxymethylcellulose	2.23	0.98			S86
melamine and 4,4′-Biphenyldicarbaldehyde	2.30	1.70			S87
Phenolic Resin	2.33	1.30			S88
Polycarbazole	2.99	1.55			S89
Biomass derived carbon	3.00	1.20			S90
Chitosan	3.23	1.58			S91
microalgae-NaAlg	3.34	1.25			S92
Polymer NUT-2	3.50	1.39			S93
Phenolic resin	3.55	1.36			S94
Meta-aminophenol–formaldehyde resin	3.80	1.67			S95
Poplar anthers derived carbon	3.81	1.40			S96
Banana peel	4.20	1.27			S97
Cetylpyridinium bromide	4.20	0.45			S98
N-doped porous carbons	4.32	1.80			S99
1,3,5-THB and nitrobenzene	4.61	1.35			S100
N-doped carbons	4.70	1.00			S101
Polypyrrole functionalized graphene sheets	4.80	1.50			S102
water chestnut and melamine	4.89	1.90			S103
Polyimine	5.05	0.84			S104
Polybenzoxazine derived carbon	5.25	1.77			S105
phenolic resin	5.36	1.35			S106
Polybenzoxazine resins	5.60	0.86			S107
N-doped carbon nanotube	5.90	1.00			S108
Polyacrylonitrile	6.10	1.20			S109
Procambarus Clarkii Shells	6.38	1.40			S110
Chitosan	6.80	1.86			S111
Polyindole	6.87	1.57			S112
Polyurethane foam	6.92	1.25			S113
Nanostructured templated carbon	7.00	1.48			S114
Lignin	7.10	1.55			S115
Ammonia modified biomass carbon	7.21	1.59			S116
N-doped Coal Tar Pitch	7.70	1.45			S117
Polycarbazol	8.06	0.84			S118
Sucrose/urea based carbon	8.90	1.55			S119
Polymer/ionic liquid	9.20	0.89			S120
Wheat flour	10.00	1.45			S121
pigskin collagen	10.40	1.27			S122
p-diaminobenzene derived carbon	10.50	1.80			S123
Corncob	11.52	1.23			S124
Melamine-doped phenolic resin	11.80	1.15			S125

Precursors	N (wt%)	Adsorption capacity at 0.15 bar			Ref
		25-30 °C	40 °C	50 °C	
D-glucose and urea	12.27	1.30			S126
Hexamethoxymethylmelamine resin	13.60	0.55			S127
argan fruit shells	13.90	1.50			S128
Hexamethoxymethylmelamine resin	14.11	0.68			S129
Pyrazole	15.30	2.05			S130
Polyacrylonitrile	16.48	1.15			S131
dopamine-melamine	20.90	1.60			S132
1,3-bis(cynomethyl imidazolium) chloride	22.30	1.70			S133
Nitrogen-rich porous polymer	28.00	1.36			S134
ZIF-8 derived carbon	--	1.40			S135

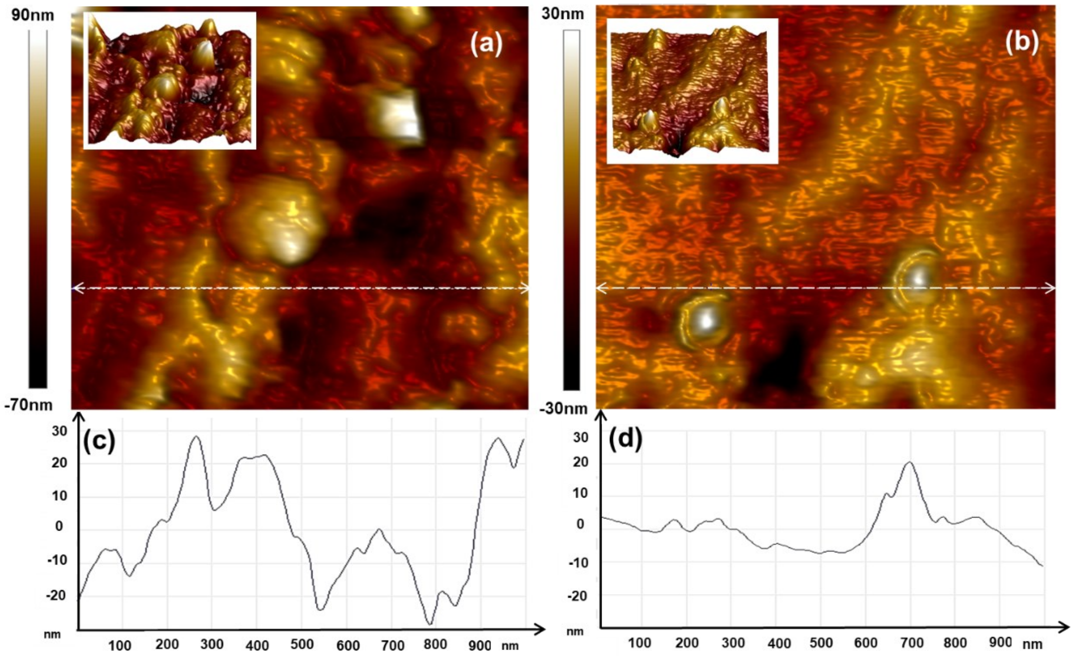


Figure S5 KPFM 3D topography image and the variation of height along the selected horizontal line: (a) (c) P81_2T; (b) (d) P81_2T_48H

Table S2 the average surface potential of the selected carbon samples

Sample	Test area	Scan size	Potential (mV)	Roughness (R_q) (mV)	Mean (mV)
P81_2T	1	1 µm	-611.3	8.0	-589.5±26.0
	2	1 µm	-559.6	20.4	
	3	1 µm	-597.6	11.2	
P81_2T_48H	1	1 µm	-784.8	8.7	-742.6±49.0
	2	1 µm	-688.9	10.0	
	3	1 µm	-754.2	8.1	

KPFM could provide the contact potential difference (CPD) induced by the difference in the surface potential between the tip and the carbon surface ($\Phi_{\text{sample}} - \Phi_{\text{tip}}$). The 3D surface topography and the surface potential of selected carbons within a scanning area of 1 µm x 1 µm were shown in Figure S4 and Table S2. It can be found that the local CPD distribution was uniform within the scanning area, which in average was -589.5±26 mV and -742.6±49 mV for P81_2T and P81_2T_48H, respectively. The line profile of topography (Figure S4) and CPD (Figure 7) showed that the CPD distribution is independent of topographic variations. For instance, the line profile of P81_2T_48H exhibited a topographic variation about 30 nm but the variation of CPD distribution was within 20 mV, accounting for about 3% of the measured CPD. It is noteworthy that a clear contrast of 153±20 mV was observed between the CPD of P81_2T and P81_2T_48H. Because same tip and experimental setup were used to test both samples, the observed CPD contrast is independent of the properties of the tip, we therefore concluded that it must stem from the actual difference in the electronic surface potential between two samples.

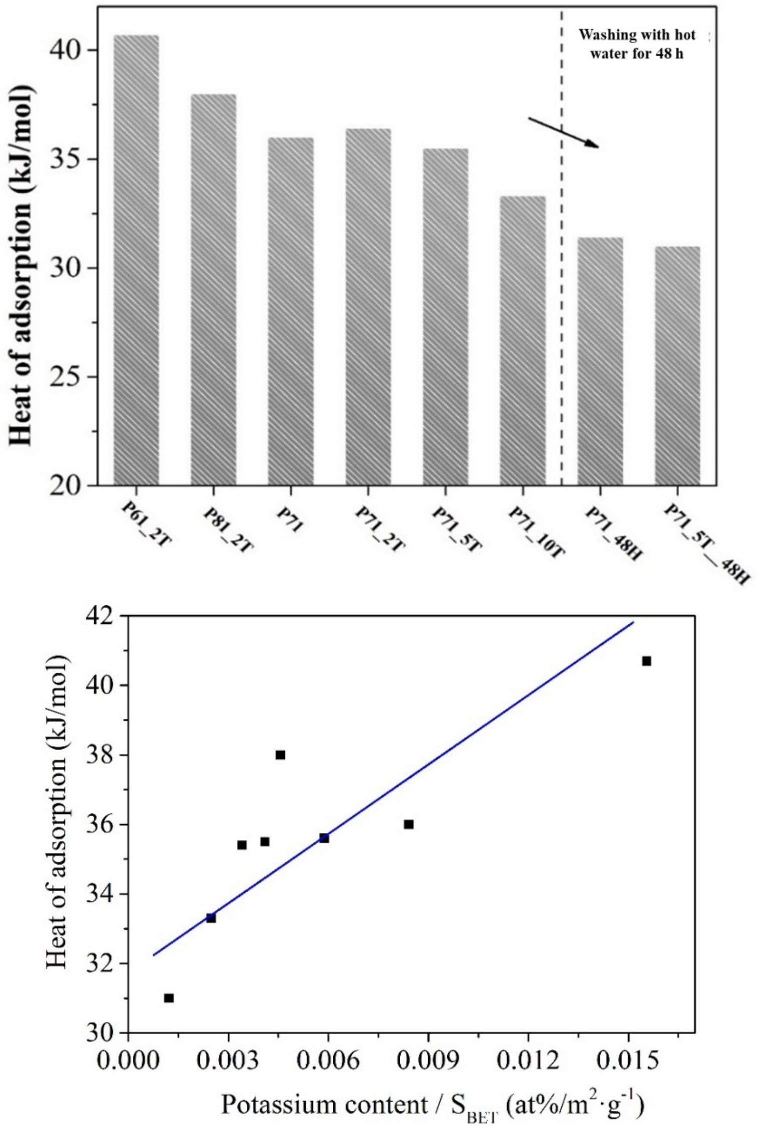


Figure S6 Heat of adsorption of PIR carbons at 40 °C in 15% CO_2/N_2 and its relationship with the density of intercalated potassium in the carbons

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