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

Preparation of Covalent Triazine Framework with the Imidazolium Cation Embedded in Basic Sites and Its application for CO_2 Capture

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Contents

List of Figures

List of Tables

1. Synthesis of bpim-CTF-400 with various $ZnCl_2$ equivalent and porosity measurements

Sample name	BET SA	$V_{pore, tot}$	Average pore diameter
monomer : $ZnCl_2$	[$m^2 g^{-1}$]	$cm^3 g^{-1}$	nm
1 :1	12	0.0057	-
1 : 5	787	0.3436	2.19
1 : 10	610	0.7541	2.28

Table S1. Porosity data of bpim-CTFs synthesized with different $ZnCl_2$ equivalent

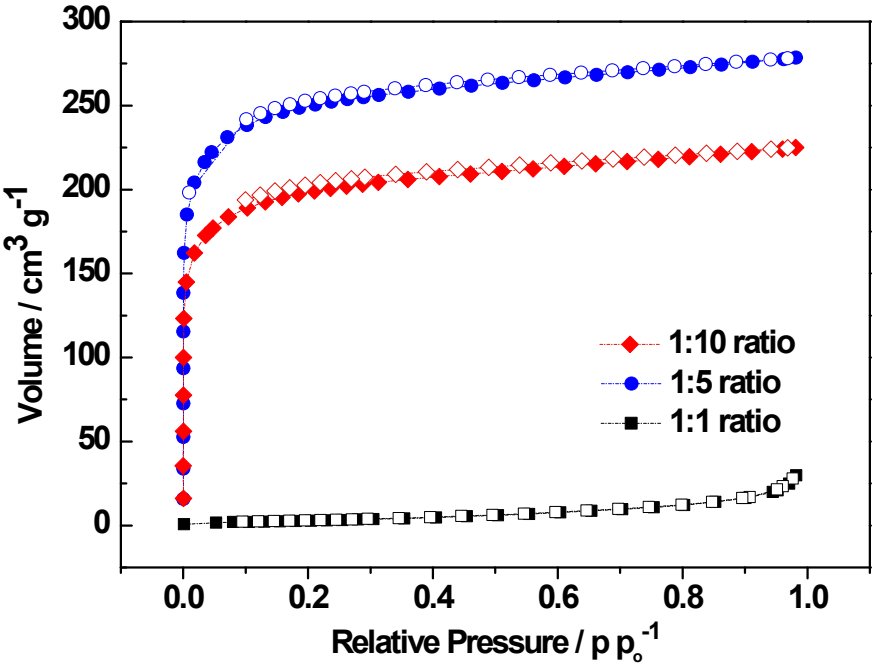


Figure S1. Nitrogen sorption isotherms of bpim-CTF synthesized with different $ZnCl_2$ equivalent at 77 K

2. SEM and EDS measurements.

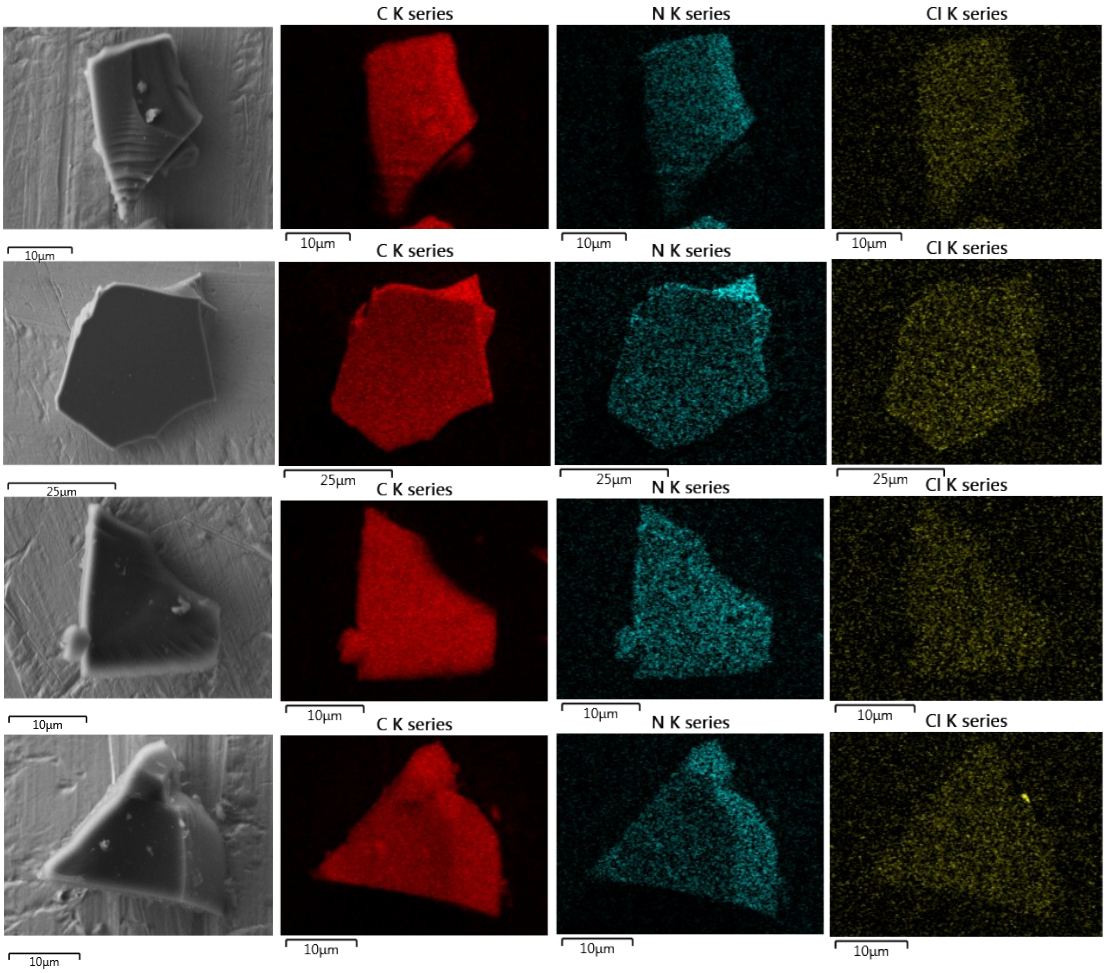


Figure S2. SEM & EDS mapping of bpim-CTFs

Sample	C[a]	N[a]	Cl[a]	Br[a]	Zn[a]
bpim-CTF-250	63.8	33.0	3.2	0.0	NA
bpim-CTF-300	68.2	27.2	4.4	0.2	NA
bpim-CTF-400	67.8	30.4	1.8	0.0	NA
bpim-CTF-500	75.1	21.9	3.0	0.0	NA

Table S2. Atomic composition of bpim-CTFs by EDS

[a]Weight %; NA refers to "Not Available.".

3. Powder X-ray diffraction

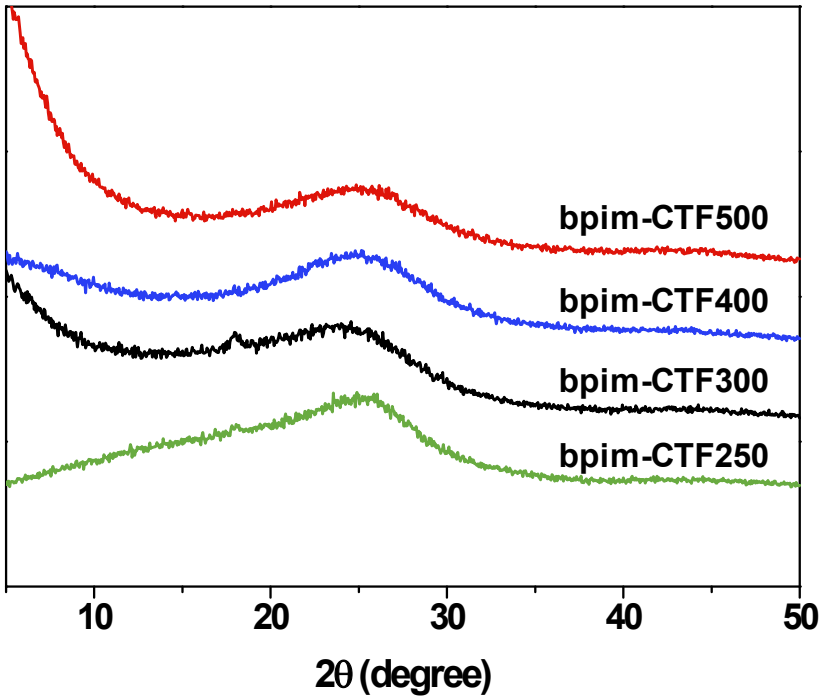


Figure S3. XPS measurement of bpim-CTFs

4. Thermal stability analysis

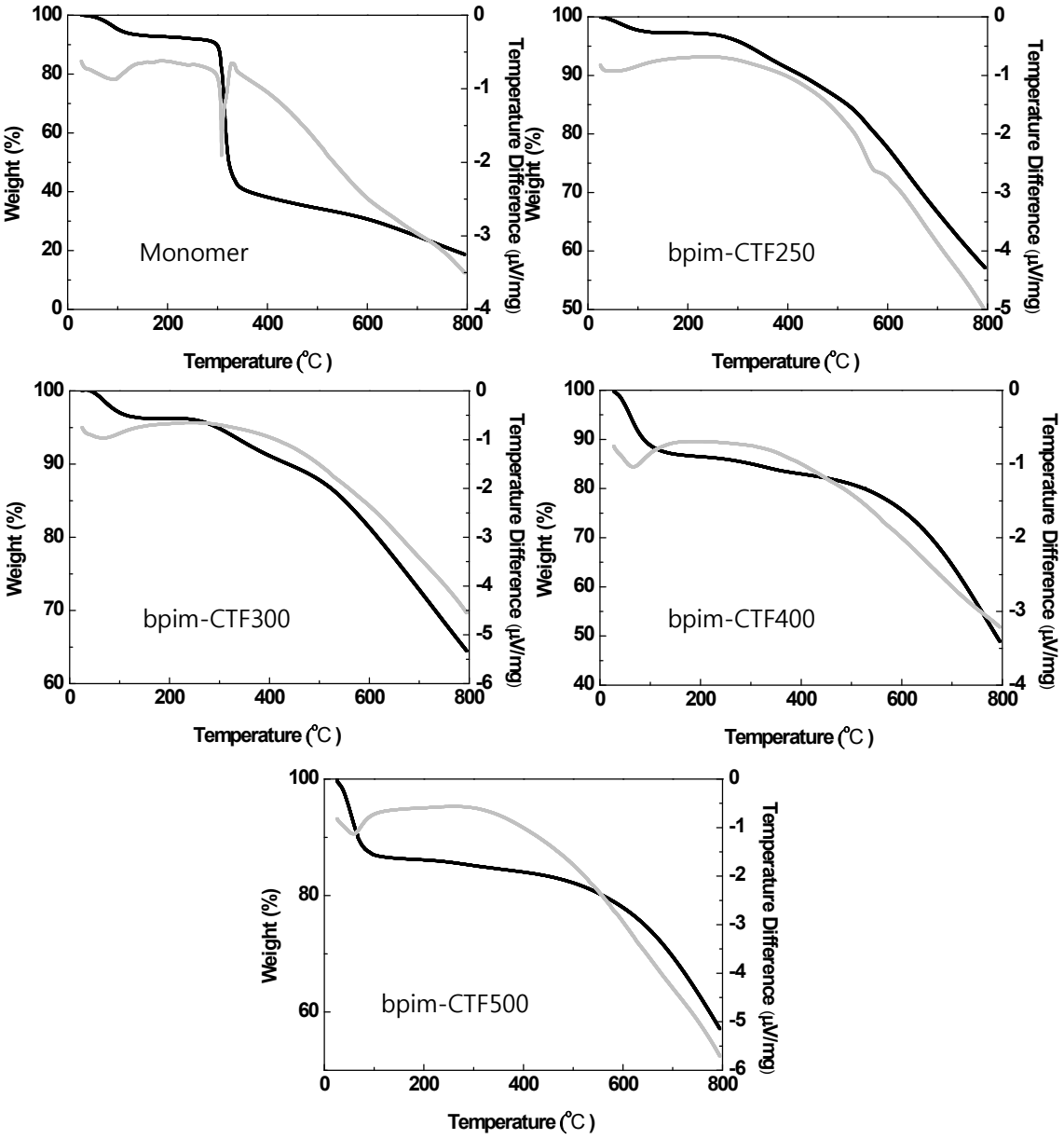


Figure S4. TGA measurements of bpim-CTFs and monomer

5. Carbon dioxide and Nitrogen physisorption measurements.

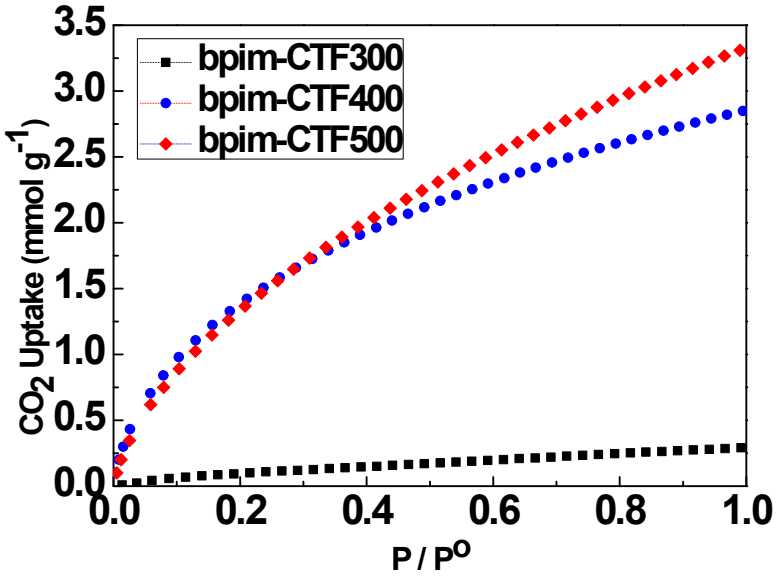


Figure S5. Carbon dioxide adsorption isotherms of bpim-CTFs at 288 K

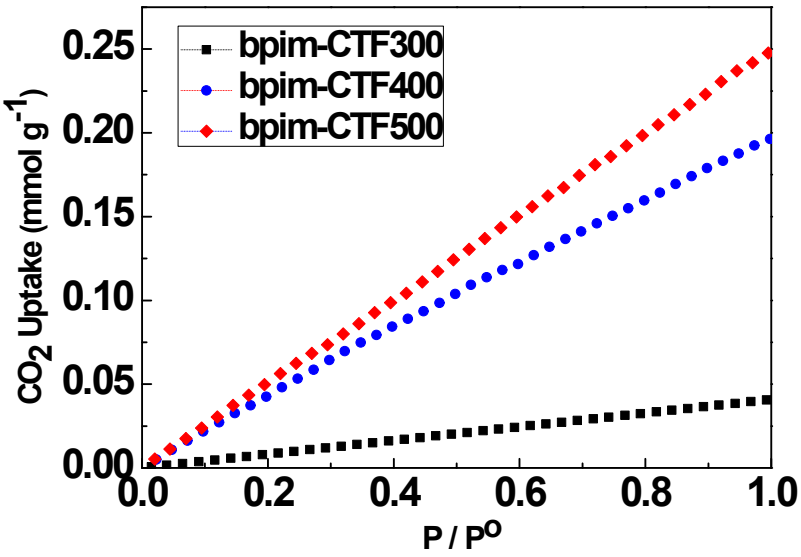


Figure S6. Nitrogen adsorption isotherms of bpim-CTFs at 298 K

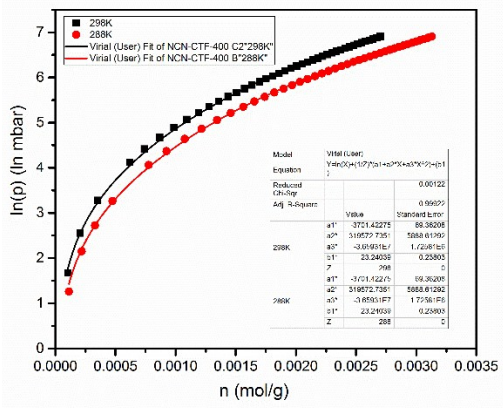
6. Heat of CO_2 adsorption

The CO_2 adsorption data measured at 288 K and 298 K were fitted by the virial equation (1) to estimate the enthalpy of adsorption.

$$\ln(p) = \ln(n) + (1/T)\sum_{i=0}^{m} a_i n^i + \sum_{i=0}^{m} b_i n^i \qquad (1)$$

where p is pressure, n is amount adsorbed, T is temperature, and a_i and b_i are temperature independent empirical parameters. The isosteric heat of adsorption was estimated from the following equation (2) as a function of CO_2 uptake.

$$Q_{st} = -R\sum_{i=0}^{m} a_i n^i \qquad (2)$$



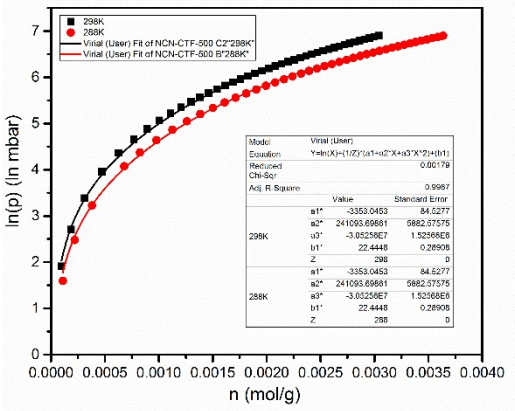


Figure S7. Virial analysis of CO_2 adsorption data for bpim-CTF400 (top) and 500 (bottom)

7. CO_2/N_2 selectivity studies by IAST calculation.

Ideal adsorbed solution theory (IAST) data was calculated based on a single-site Langmuir model for N_2 and dual-site Langmuir model for CO_2 to fit the adsorption data. [2]

The dual-site Langmuir model is defined as

$$q = q_A + q_B = \frac{q_{sat,A} b_A p}{1 + b_A p} + \frac{q_{sat,B} b_B p}{1 + b_B p}$$

(A,B = distinct adsorption sites)

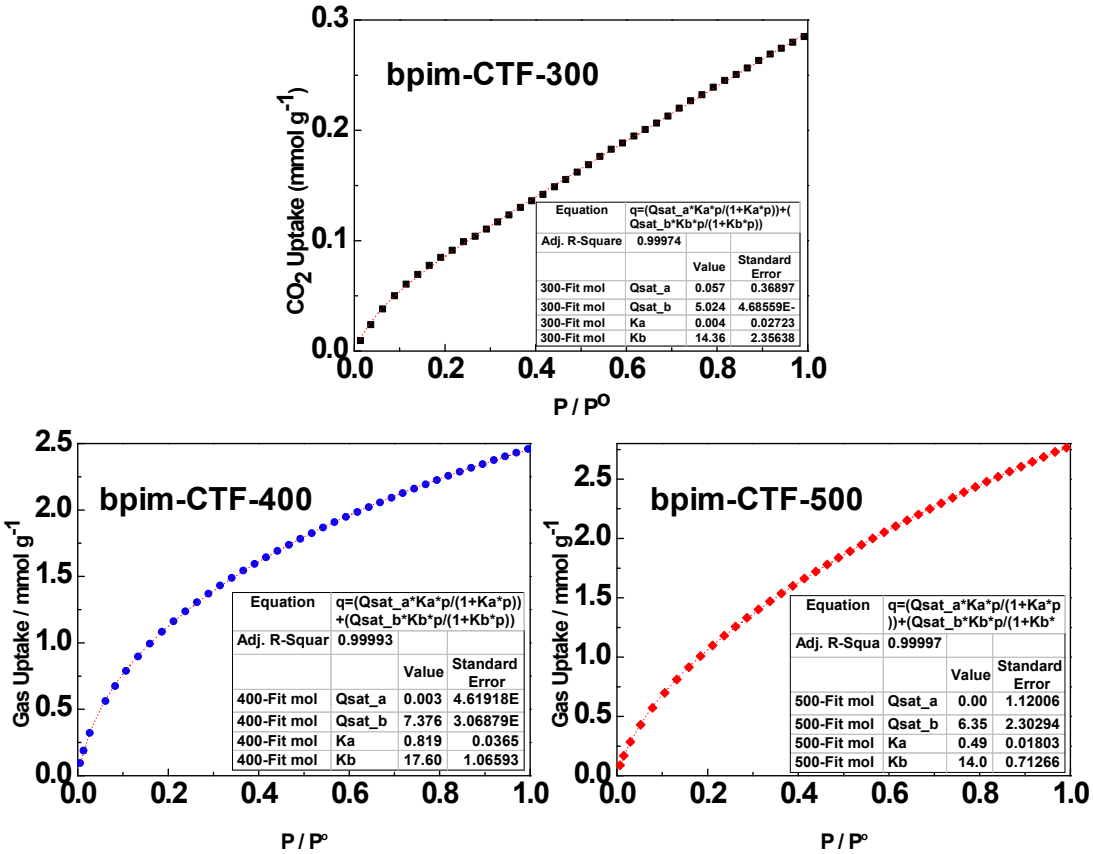


Figure S8. IAST plots of bpim-CTF-300 (top), 400 (bottom left), and 500 (bottom right) calculated from the carbon dioxide isotherms measured at 298 K.

The single-site Langmuir model is defined as,

$$q = \frac{q_{sat} bp}{1 + bp}$$

(q = molar loading of adsorbate, q_{sat} = saturation loading, b = Langmuir constant)

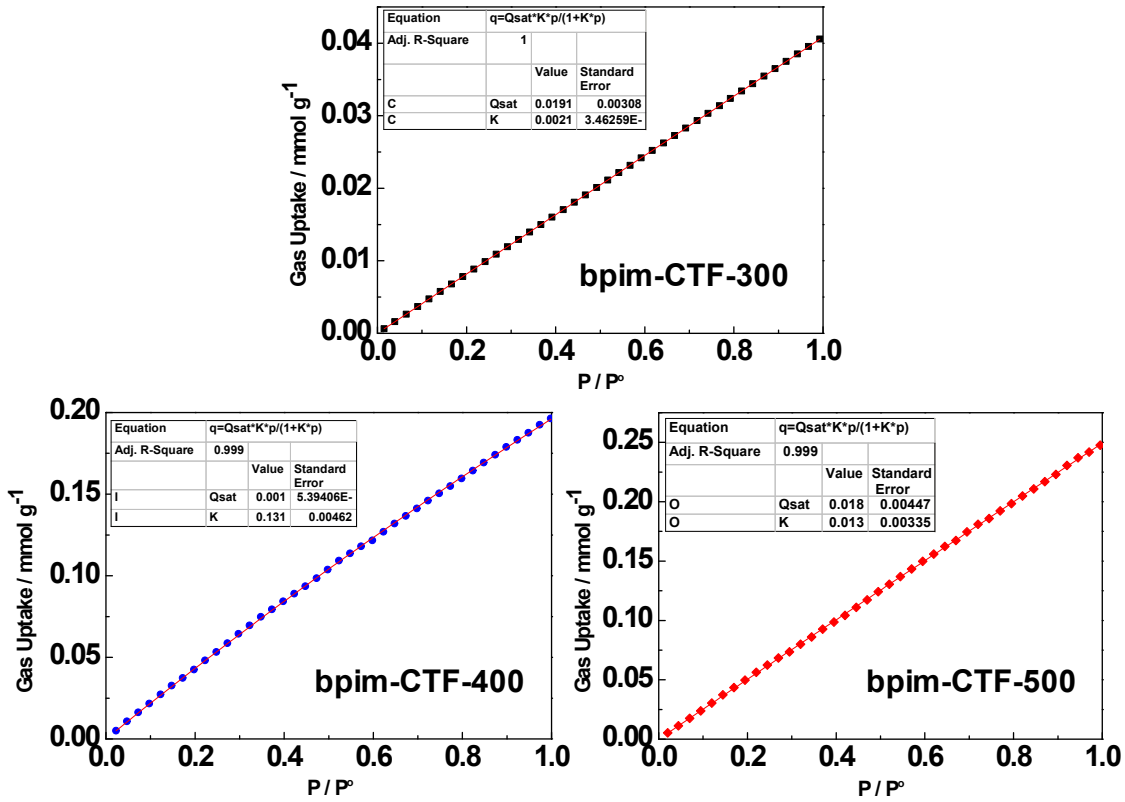
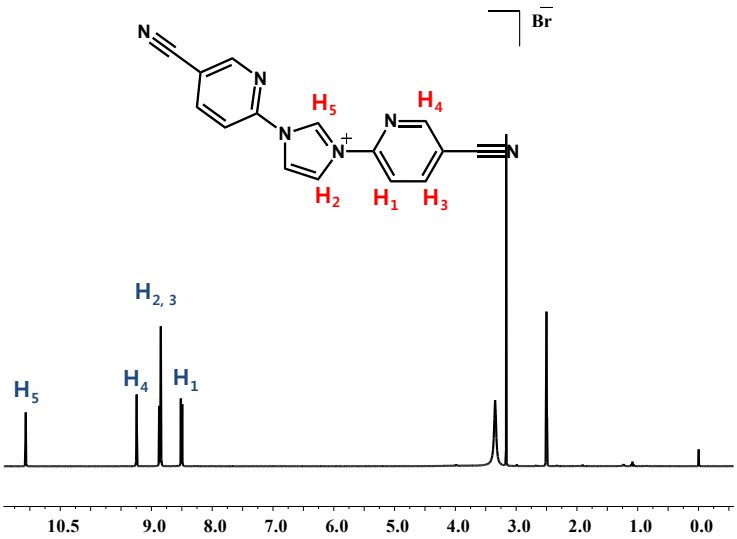


Figure S9. IAST plots of bpim-CTF-300 (top), 400 (bottom left), and 500 (bottom right) calculated from the nitrogen isotherms measured at 298 K.

Finally, the selectivity of bpim-CTFs were obtained with the fitted values using following equation,

$$S = \frac{q_1/q_2}{p_1/p_2}$$

the ratio of CO_2:N_2 were defined as 15:85 for the calculation.



8. Liquid-state NMR spectra.

Figure S10. ^{1}H-NMR spectrum of 1,3-bis(5-cyanopyridyl)-imidazolium bromide in DMSO-d_6.

^{1}H NMR (400 MHz, DMSO-d_6) δ [ppm] = 11.07 (t, $^{4}J_{HH}$ = 1.66 Hz, 1H, NCHN), 9.24 (dd, $^{5}J_{HH}$ = 0.77 Hz, $^{4}J_{HH}$ = 2.23 Hz, 2H, CH-$_{6py}$), 8.86 (dd, $^{4}J_{HH}$ = 2.23 Hz, $^{3}J_{HH}$ = 8.70 Hz, 2H, CH-$_{4py}$), 8.84 (d, $^{4}J_{HH}$ = 1.66 Hz, 2H, NCHCHN), 8.50 (dd, $^{5}J_{HH}$ = 0.77 Hz, $^{3}J_{HH}$ = 8.70 Hz, 2H, CH-$_{3py}$)

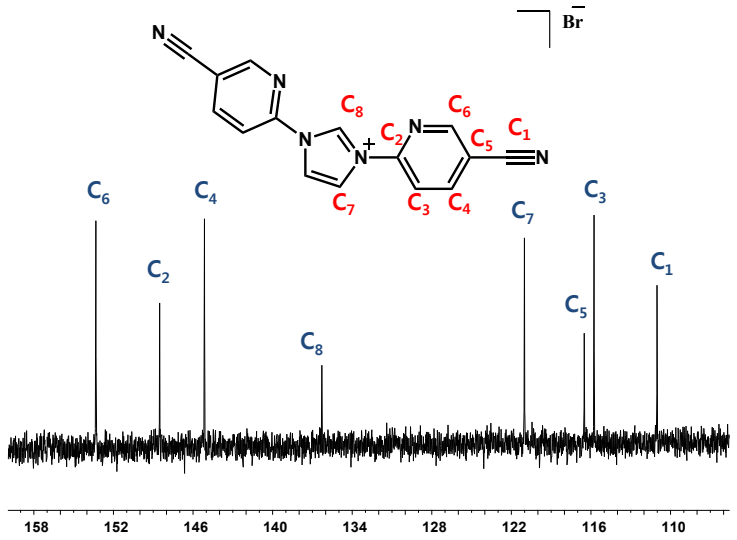


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Figure S11. ^{13}C-NMR spectrum of 1,3-bis(5-cyanopyridyl)-imidazolium bromide in DMSO-d_6.

^{13}C NMR (400 MHz, DMSO-d_6) δ [ppm] = 153.3 (s, CH-$_{6py}$), 148.5 (s, CH-$_{2py}$), 145.1 (s, CH-$_{4py}$), 136.3 (s, NCHN), 121.0 (s, NCHCHN), 116.5 (s, C-$_{5py}$), 115.7 (s, C-$_{3py}$), 111.0 (s, CN).

9. Mass spectrum

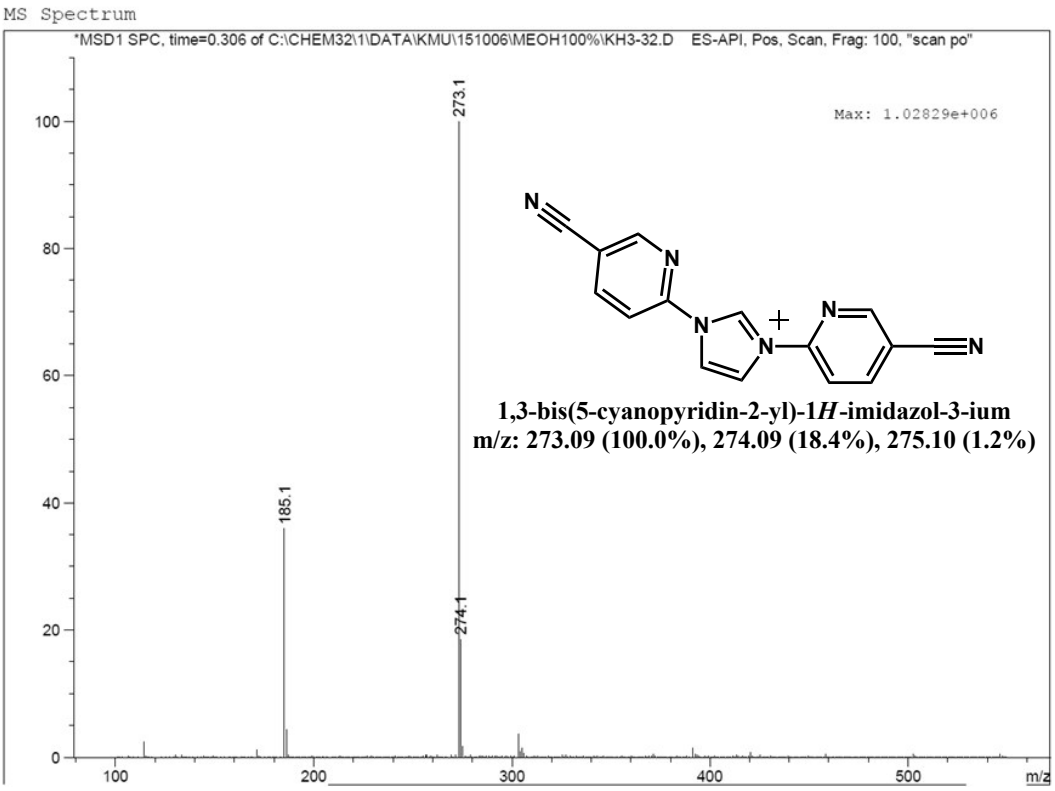

Figure S12. LC-MS spectrum of 1,3-bis(5-cyanopyridyl)-imidazolium bromide

10. Proposed structural change of bpim-CTFs in varying synthetic temperature

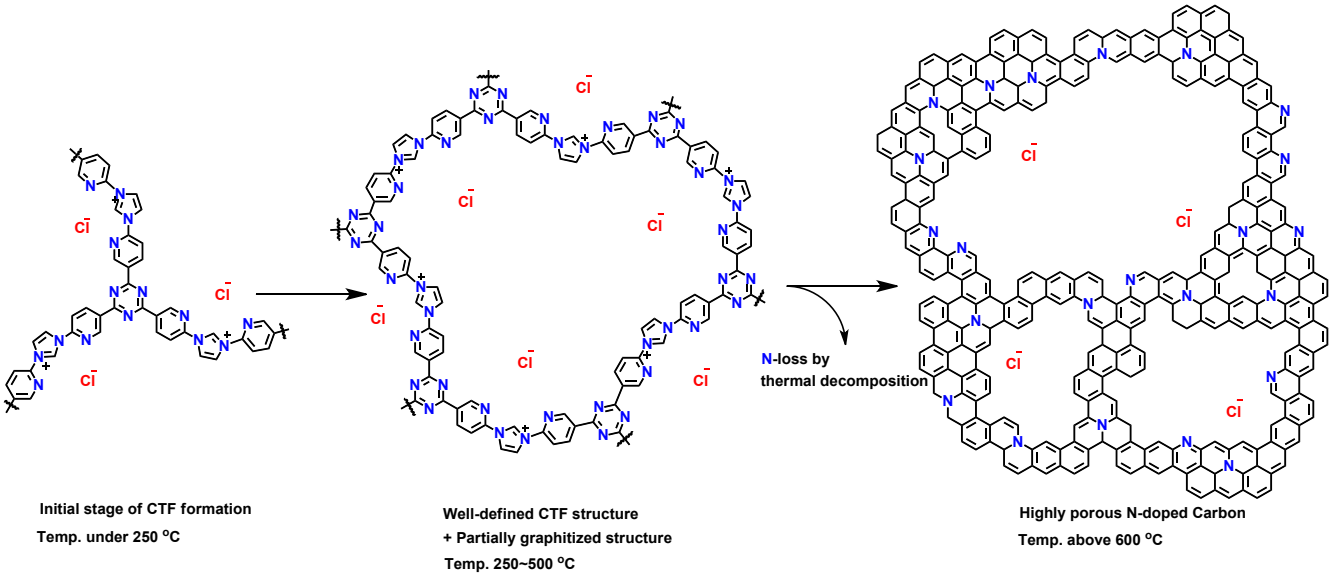

Figure S13. Proposed schematic representation for the gradual transformation of bpim-CTFs to N-doped porous carbons derived by thermal decomposition*(concept for N-doped porous carbon was referred to ref 122)

11. Representative characteristics of other CTFs and POPs

Table S3. Characteristics of reported CTFs

CTFs	BET surface area ($m^2 g^{-1}$)	CO_2 (mmol g^{-1})[a]	CO_2/N_2[d]	Ref.
fl-CTF300	15	0.71	37	1
fl-CTF350	1235	2.29	23	
fl-CTF400	2862	1.97	16	
fl-CTF500	2322	1.65	12	
fl-CTF600	2113	1.80	12	
MCTF@300	640	1.41	-	2
MCTF@400	1060	1.58	-	
MCTF@500	1510	2.26	-	
PCTF-1	2235	1.87[b]	14	3
PCTF-2	784	1.01[b]	14	
PCTF-3	641	1.35[b]	25	4
PCTF-4	1090	1.51[b]	26	
PCTF-5	1183	1.51[b]	32	
PCTF-6	79	-	-	
PCTF-7	613	1.34[b]	41	
FCTF-1	662	3.21	31	5
FCTF-1-600	1535	3.41	19	
CTF-FUM-350	230	2.31	102.4	6
CTF-FUM-400	480	1.98	96.3	
CTF-FUM-500	603	1.50	67.3	
CTF-DCN-400	690	1.18	33.7	
CTF-DCN-500	735	1.55	37.0	
COP-3	413	1.14	24.4	7
PCTF-1	853	2.05	24	8
PCTF-2	811	1.70	17	
PCTF-3	391	0.95	14	
PCTF-4	1404	2.86	56	
CTF-BI-3	677	1.97[c]	52.4[e]	9
CTF-BI-4	1025	2.43[c]	102.7[e]	
CTF-BI-5	836	2.39[c]	40.2[e]	
CTF-BI-6	759	1.75[c]	41.0[e]	
CTF-BI-7	642	1.48[c]	33.6[e]	
CTF-BI-8	55	-	-	
CTF-BI-9	885	2.10[c]	29.0[e]	
CTF-BI-10	1099	2.29[c]	39.1[e]	
CTF-BI-11	1549	2.34[c]	34.9[e]	
CTF-TB-1	294	1.33[c]	50.0[e]	10
CTF-TB-2	446	1.73[c]	52.2[e]	
CTF-TB-3	612	2.19[c]	48.1[e]	
CTF-TB-4	581	1.49[c]	39.0[e]	
CTF-TB-5	495	1.33[c]	39.5[e]	
CTF-TB-7	732	2.01[c]	35.4[e]	

CTF-TB-8	689	1.59[c]	32.5[e]	
CTF-TB-9	626	1.63[c]	41.0[e]	
bipy -CTF-300	360	0.98	41	11
bipy -CTF-400	753	1.78	40	
bipy -CTF-500	1548	3.07	42	
bipy-CTF-600	2479	2.95	24	
CTF1-400	610	1.52	45	
CTF1-500	1830	2.23	29	
CTF1-600	2557	2.21	17	
pym-CTF500	208	1.77	502	
pym-CTF600	689	2.15	124	
lut-CTF300	486	2.14	57	
lut-CTF350	635	2.41	66	
lut-CTF400	968	2.72	53	
lut-CTF500	1680	2.58	27	
lut-CTF600	2815	2.52	23	
HAT-CTF-450/600	1090	4.8	110	12
TPI-1	809	1.25	30.9	13
TPI-2	796	1.23	33.5	
TPI-3	40	0.43	35.1	
TPI-4	245	1.11	46.2	
TPI-5	201	0.96	46.2	
TPI-6	510	1.10	33.8	
TPI-7	< 10	1.10	55.5	
TPI-1@IC	1053	2.11	80	14
TPI-2@IC	814	1.43	151	
TPI-3@IC	963	1.44	77	
NOP-1	749	1.08	32.8	15
NOP-2	803	1.42	34.1	
NOP-3	894	1.41	33.8	
NOP-4	428	0.84	31.9	16
NOP-5	613	0.73	30.5	
NOP-6	720	0.50	29.2	
NOP-19	982	1.57	53[f]	
NOP-20	952	1.64	81[f]	
NOP-21	565	1.57	68[f]	
TFM-1	791	0.91	-	17
2	-	1.53	49	18
3	646	1.65	34	
4	1266	2.06	20	
2C	427	2.95	33	
3C	1173	3.03	31	
4C	1316	3.55	22	
TSP-1	562.5	1.90	32[f]	19
TSP-2	913.0	2.60	38[f]	
TCMP-0	963	1.34	9.6	20

TNCMP-2	995	1.45	7.6	
TCMP-3	691	1.26	25.2	
TCMP-5	494	0.68	17.0	
MCTP-1	1452	2.70	15.4[e]	21
MCTP-2	859	2.46	68.6[e]	
PCTP-1	1200	3.25	46.1[e]	22
PCTP-2	523	2.34	31.6[e]	
APOP-1	1298	2.69	20.3	23
APOP-1-OH	875	1.86	26.0	
APOP-1-ONa	760	1.71	29.2	
APOP-1-F	724	2.02	31.8	
APOP-2	906	1.30	20.2	
APOP-3	1402	2.59	26.0	
APOP-4	833	1.64	23.3	
bpim-CTF400	786	2.46	32	This work
bpim-CTF500	1556	2.77	23.5	

[a]CO_2 adsorption at 1bar and in 298 K. [b]CO_2 adsorption at 1 bar in 293 K.· [c]CO_2 adsorption at 1 bar in 303 K. [d]CO_2/N_2 selectivity by using IAST method at 298 K [e]CO_2/N_2 selectivity by using Henry method at 303K. [f]CO_2/N_2 selectivity by using IAST method at 273 K

Table S4. Characteristics of reported POPs*

*original information from ESI data of ref. 7.

COFs	BET surface area ($m^2 g^{-1}$)	CO_2[a] ($mmol\ g^{-1}$)	CO_2/N_2[b]	Ref.
CMPs	522-1043	0.93 - 1.18	-	25
JUC-Z2	2034	1.56	-	26
polyamine particles	246	2.20	-	27
HMPs	437-726	1.41 - 1.70	-	28
PAFs (41~44)	515-1119	1.24 - 2.26	-	29
POF1B ~ POF3B	608-917	1.48 - 2.16	-	30
SMPs-1 ~ SMPs-14	757-1421	1.73 - 2.61	-	31
HCPs	3-1684	0.25 - 1.68	-	32
ACMPs	46-629	0.57 - 1.08	-	33
Networks	1102-3160	0.91 - 1.49	-	34
CEs	540-960	0.86 - 0.91	-	35
BLPs	1360-2244	0.25 - 1.68	-	36
PAFs (32)	1230-1679	0.99 - 1.25	-	37
PI-ADPM	868	1.59	-	38
PSNs	376-1045	1.11 - 2.23	-	39
ILCOF-1	2273	0.84	-	40
CMPs	772-965	1.61 - 1.80	-	41
TDCOF-5	2497	1.23	-	42
GPOPs	680-1010	1.20 – 1.89	-	43
PIMs	531-771	1.41 - 1.65	-	44
F-MOPs	832-1031	1.32 - 1.68	-	45
SNU-C1-va	595	2.31	-	46
SNU-C1-sca	830	3.14	-	
PAF-16-2	979	1.18	-	47
PAFs	2246-5460	1.09 - 1.82	-	48
MOPs (A-$B1^{II}$ ~$B3^{III}$)	142-614	1.70 - 2.01	-	49
Networks	618-1980	1.12 - 1.99	-	50
NPTNs	1055-1558	1.34 - 1.84	-	51
PBIs	62-85	0.22 - 1.56	-	52
STPIs	4-541	1.09 - 2.30	-	53
JUC-12	750	1.70	-	54
TEPOs	485-592	0.83 - 1.21	-	55
TB-MOP	694	2.57	-	56
SMPIs	23-574	1.43 - 1.87	-	57
TzTz-POPs	299-488	1.30 - 1.50	-	58
CP-CMPs	847-2241	1.53 - 2.44	-	59
PTPAs	544-1557	0.82 - 1.56	-	60
TBCs	540-917	1.09 - 1.79	-	61

Cz-POFs	671-2065	1.32 - 3.05	-	62
HP_E-CMP	662	1.70	-	63
PPFs (1~4)	419-1740	1.43 - 3.57	-	64
Networks	1147-1236	1.66 - 1.71	-	65
ACOF-1	1176	2.05	-	66
PIs	26-744	1.23 - 2.02	-	67
MPIs	586-1454	1.65 - 2.14	-	68
BILP-10(C1)	924	1.41	15.8	69
HCPs	742-847	1.14 - 1.70	19-27	70
Networks	653-4077	1.08 - 2.20	8.7-19.5	71
CC-6	99	0.89	11	72
PPN-6s	555-1740	1.23 - 3.59	13-442	73
ZCs	-	0.10 - 0.36	38-138	74
Cs (C5~C7)	-	0.18 - 0.20	38.0-73.0	75
Fs (F1~F3)	-	0.10 - 0.23	42-213	76
NPAF	1790	2.33	89	77
PAF-18s	981-1121	1.50 - 2.02	20-65	78
azo-COPs	11.1-729.6	1.12 - 1.53	175-325	79, 80
MOP Networks	333-1015	1.24 - 2.27	16-26	81
COPs (1~2)	158-167	0.93 - 1.36	7.9-25	82
IBN9-NCs	890-1181	1.81 - 4.50	-32	83
PIs	506-568	1.00 - 1.41	12-27	84
MCs (0~100)	7-1289	0.35 - 1.61	15.9-49.2	85
CE-Ps	195-630	0.67 - 1.07	14.2-37.8	86
PDVBs	19-825	0.35 - 1.49	-373	87
BILP-101	107.2	1.30	100	88
Oz-COP	553.4	1.47	40	89
PAF-56P	747-1082	1.70 - 1.90	39.5-40	90
MBMOPs	962-1044	0.43 - 0.48	3-15	91
H_2P-COFs	0.094-1340	0.50 - 3.16	-140	92
TB-COPs	439-712	0.12 - 0.34	46-54	93
Azo-POFs	622-755	0.15 - 0.16	31-34	94
Fne-POFs	370-953	0.75 - 1.25	19.4-79.8	
PAFs (33~35)	1096	0.23 - 1.14	-	95
PPN-101	859-1452	2.46 - 2.70	15.4-68.6	96
BILPs (10~13)	862-1235	0.23 - 0.30	27-35	97
ALPs (1~4)	791-983	1.12 - 1.13	3.4-4.8	98
POPs (1,2)	243-1022	1.10 - 2.20	41-80	99

CBZ, DBT, DBF, IN, BT, BF	523-1200	0.03 - 0.30	31.6-46.1	100
POPs (1~2)		1.62 - 1.9.	600-3000	101
PPNs (80~81)	1261-1306	2.75 - 3.30	31-39	102
BILPs (2~7)	599-1135	1.98 - 3.59	32-71	103, 104
POP-diimides	560-960	1.50 - 2.07	12.5-30	105
DBMOP-6	750	-	-	106
OMPM-1	-	2.01	-	107
NPOFs (4, NO_2, NH_2)	337-1249	1.40 - 1.88	16-66	108
PPNs (1~3)	1249-2840	-	-	109
PAF-30	540	1.55	-	110
COPs (1~4)	827-3041	0.90 - 1.40	2-30	111
BILP-1	1172	2.98	7	112
P-1, P-2	611-1222	1.27-1.64	8-29	113
SNW-1	-	2.10	25	114
NPIs (1~3)	291-721	1.09 - 1.80	15.7-45.2	115
PSN-3	865	2.00	69	116
PPN-6-SO_3NH_4	593	3.50	196	117
Ni-Pors (1~4)	778-1711	2.26 - 3.13	-	118
PECONFs (1~4)	-851	1.34 - 2.47	41-51.1	119
PAF-26s	430-717	1.47 - 1.65	27-113	120
TTPPs	593-606	1.04 - 1.56	20.5-22.0	121
PINs	28-458	0.70 - 1.22	40-41	122

12. References

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