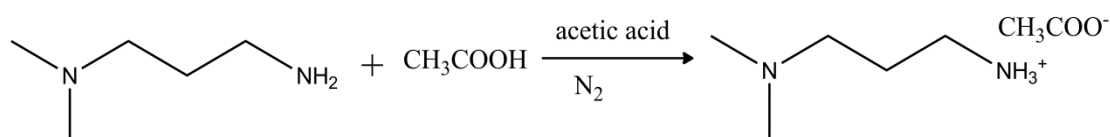# Supplementary Information

Ionothermal Synthesis and Crystal Structures of Novel Aluminum Phosphates with in Situ Generated Templates

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Scheme S1 synthetic route of (HDMAPA)(Ac)

Table S1 Synthesis details and conditions for the preparation of materials

Entry	Mass of reagents added (g) (molar ratio of reagents)				Tem.(°C)	Time (h)	Product
	Al(OH)$_3$	H$_3$PO$_4$	PILs	HF			
1	0.078(1.0)	0.057 (0.5)	1.62(10.0)	0	180	72	amorphous
2	0.078(1.0)	0.113 (1.0)	1.62(10.0)	0	180	72	amorphous
3	0.078(1.0)	0.226 (2.0)	1.62(10.0)	0	180	72	AlPO-PIL1
4	0.078(1.0)	0.339 (3.0)	1.62(10.0)	0	180	72	AlPO-PIL1
5	0.078(1.0)	0.452 (4.0)	1.62(10.0)	0	180	72	AlPO-PIL1
6	0.078(1.0)	0.576 (5.0)	1.62(10.0)	0	180	72	AlPO-PIL1
7	0.078(1.0)	0.692 (6.0)	1.62(10.0)	0	180	72	AlPO-PIL1
8	0.078(1.0)	0.791 (7.0)	1.62(10.0)	0	180	72	AlPO-PIL1
9	0.078(1.0)	0.057 (0.5)	1.62(10.0)	0.104 (0.5)	180	72	amorphous
10	0.078(1.0)	0.113 (1.0)	1.62(10.0)	0.104 (0.5)	180	72	AlPO-PIL2
11	0.078(1.0)	0.226 (2.0)	1.62(10.0)	0.104 (0.5)	180	72	AlPO-PIL2
12	0.078(1.0)	0.339 (3.0)	1.62(10.0)	0.104 (0.5)	180	72	AlPO-PIL2
13	0.078(1.0)	0.452 (4.0)	1.62(10.0)	0.104 (0.5)	180	72	amorphous

Figure S1 — ¹H-NMR

Peaks: 8.205, 8.136, 7.314, 2.983, 2.961, 2.939, 2.513, 2.497, 2.476, 2.454, 2.259, 1.938, 1.845, 1.824, 1.802, 1.780, 1.759, -0.001

Structure labels: 1, 2, 4, 5 on $N(CH_3)_2$–CH_2–CH_2–CH_2–NH_3^+ [CH_3COO^-], 3

Active Hydrogen

Integrals: 3.48 | 1.96 | 2.01 | 6.00 | 3.28 | 2.03
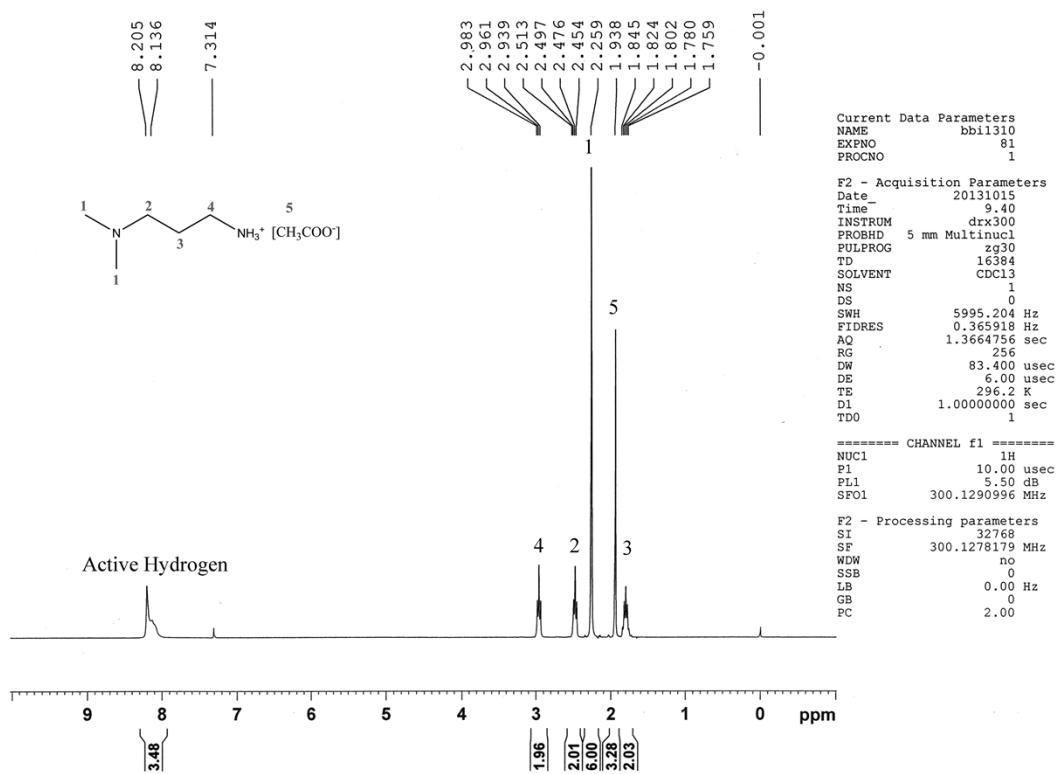
Figure S1 ¹H-NMR spectrum of (HDMAPA)(Ac)

Figure S2 — ¹³C-NMR

Peaks: 178.685, 78.191, 77.767, 77.342, 58.189, 45.569, 39.580, 25.656, 25.067

Structure labels: 1, 2, 4, 5, 6 on $N(CH_3)_2$–CH_2–CH_2–CH_2–NH_3^+ [CH_3COO^-], 3
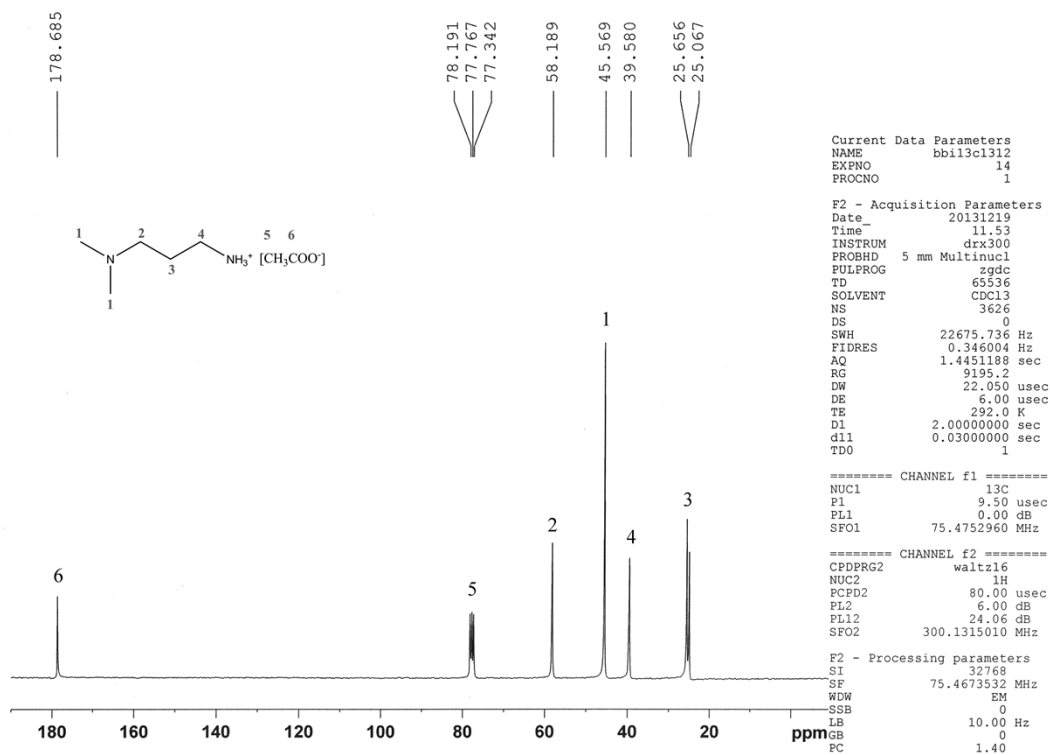
Figure S2 ¹³C-NMR spectrum of (HDMAPA)(Ac)
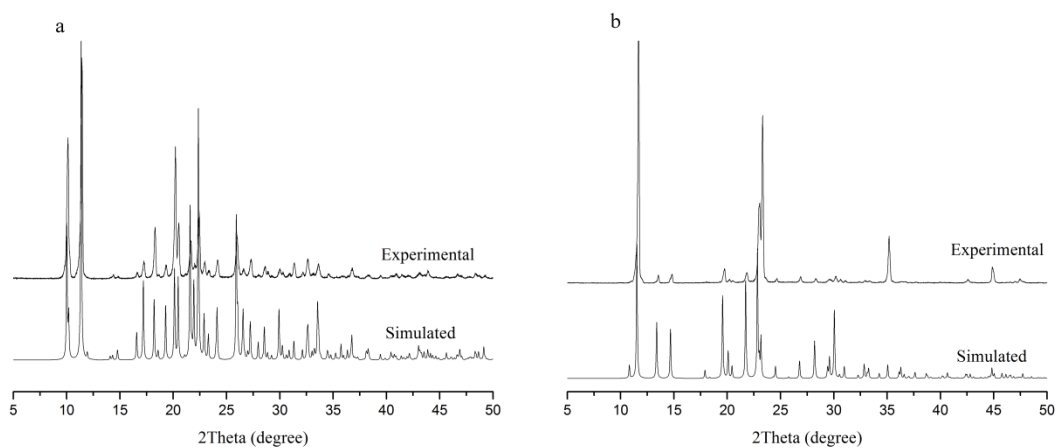
Figure S3 Simulated and experimental powder XRD patterns of AlPO–PIL1 (a) and AlPO–PIL2 (b)
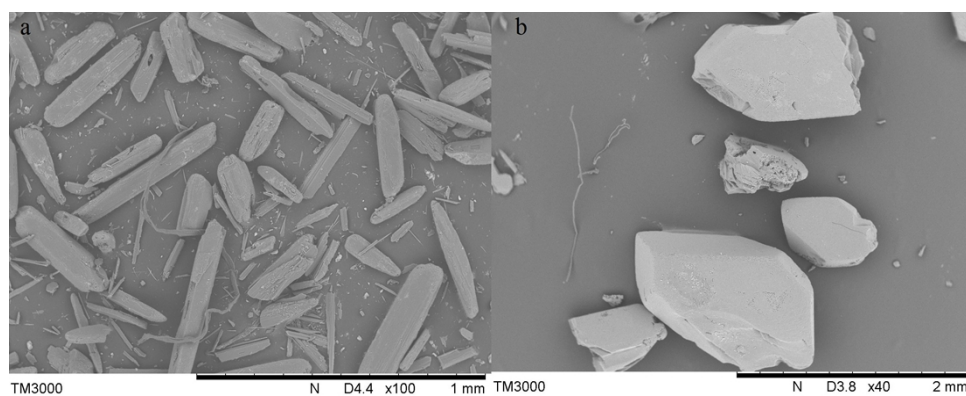

Figure S4 SEM images of AlPO–PIL1 (a) and AlPO–PIL2 (b)
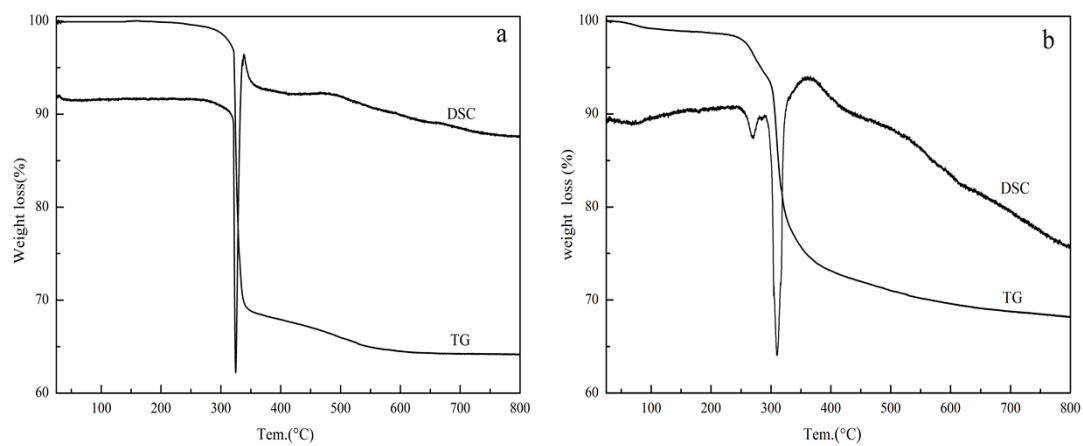

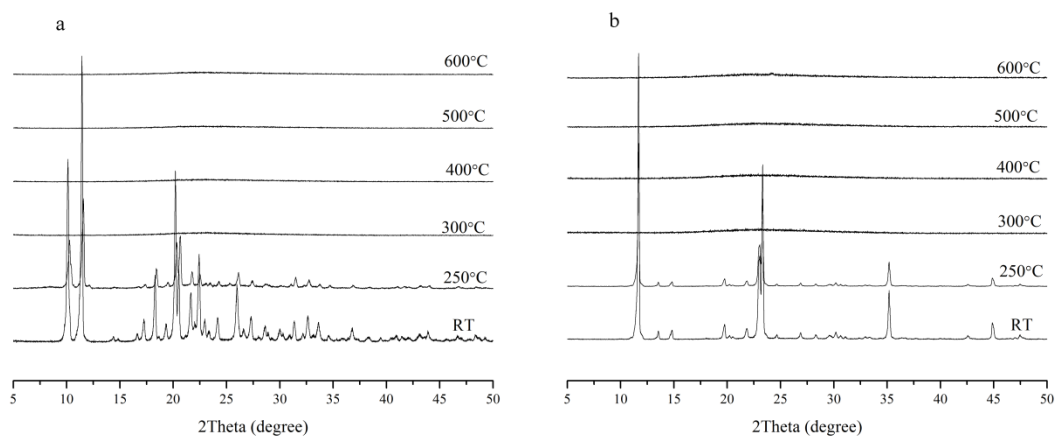**Figure S5** TG-DSC curves of AlPO-PIL1 (a) and AlPO-PIL2 (b)

Figure S6 Powder XRD patterns of the two materials after heat treatment in air at different temperatures: AlPO–PIL1 (a) and AlPO–PIL2 (b).
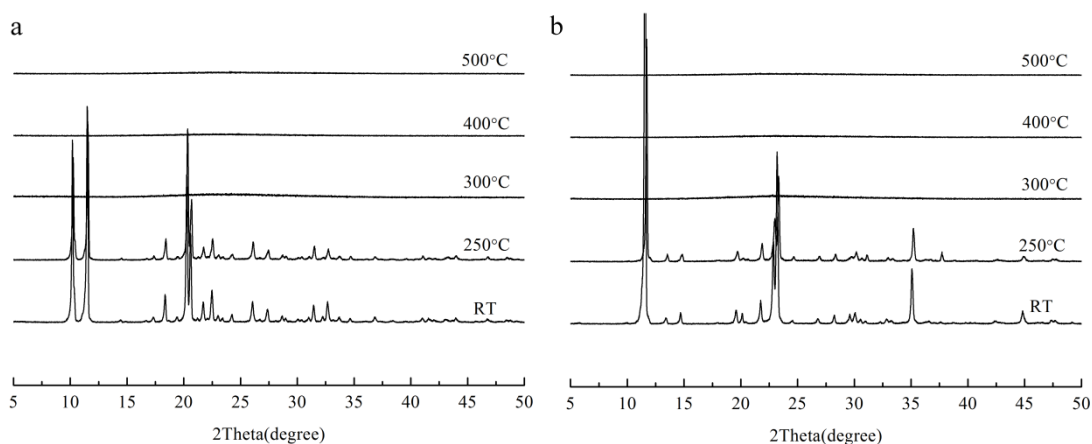

Figure S7 Powder XRD patterns of the two materials after heat treatment in N_2 atmosphere at different temperatures: AlPO–PIL1 (a) and AlPO–PIL2 (b).
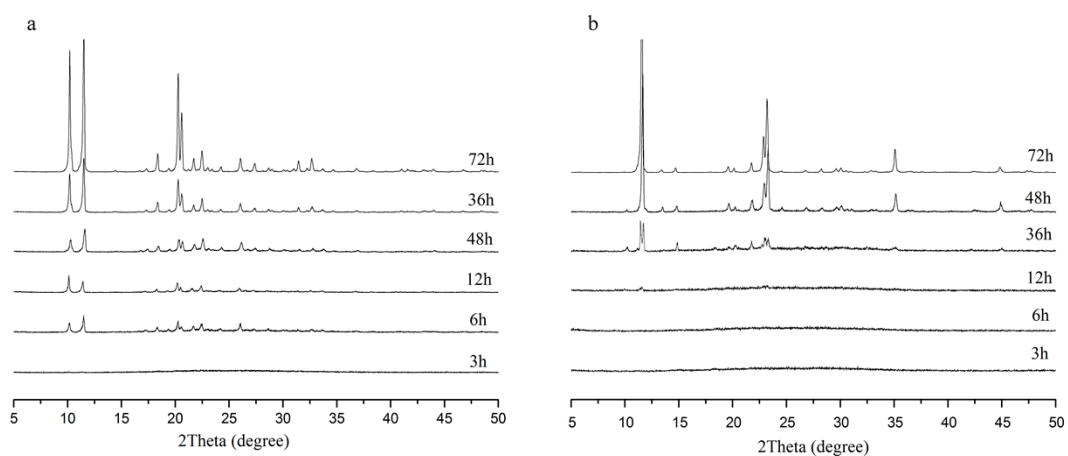

Figure S8 Powder XRD patterns illustrating the structure evolution of the two aluminophosphates AlPO-PIL1 (a) and AlPO-PIL2 (b)