

Supplementary Information

Table S1. Synthesis conditions and Al content of AlC_x materials.

Name	Nominal Al Content (at%)	Al Precursor	Synthesis Temperature (°C)	Actual Al (at%)
AlC ₅₀₀	0.2	AlCl ₃	1100	0.0
AlC ₃₃₃	0.3	AlCl ₃	1100	0.1
AlC ₂₀₀	0.5	AlCl ₃	1100	0.1
AlC ₁₀	9	AlCl ₃	1100	0.3
AlC ₆	14	AlCl ₃	1100	0.4
AlC ₃	25	AlCl ₃	1100	0.3
TMA-AlC ₃	25	TMA	800	0.0

Table S2. Raman spectral features of AlCl₃- and TMA-derived AlC_x samples post-HF wash.

Material	Pos(D) (cm ⁻¹)	FWHM(D) (cm ⁻¹)	Pos(G) (cm ⁻¹)	FWHM(G) (cm ⁻¹)	I(D)	I(G)	I(D)/I(G)	Pos(2D) (cm ⁻¹)
Ordered								
AlC ₂₀₀	1333	22	1571	47	19562	16286	1.20	2673
AlC ₃₃₃	1342	25	1580	52	18952	15391	1.23	2687
AlC ₅₀₀	1334	38	1567	60	6934	7974	0.87	2673
AlC ₁₀	1339	27	1575	53	12582	13541	0.93	2674
AlC ₆	1327	24	1563	47	10446	11416	0.92	2666
AlC ₃	1337	32	1573	58	10935	12003	0.92	2674
TMA-AlC ₃	--	--	--	--	--	--	--	--
Disordered								
AlC ₂₀₀	1344	52	1584	71	10075	7780	1.30	--
AlC ₃₃₃	1348	52	1590	73	9962	7284	1.37	--
AlC ₅₀₀	1343	53	1585	79	12513	9237	1.35	--
AlC ₁₀	1342	66	1579	86	13367	11619	1.16	--
AlC ₆	1345	58	1584	85	8390	6514	1.28	--
AlC ₃	1335	57	1575	74	6830	5474	1.25	--
TMA-AlC ₃	1338	91	1584	84	3517	3180	1.11	--

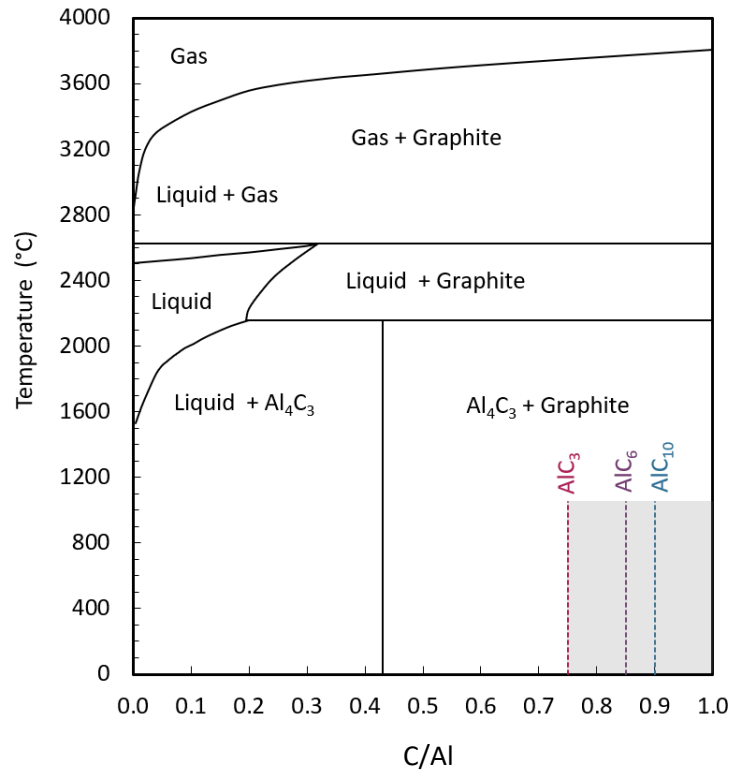


Figure S1. Aluminum-carbon phase diagram, adapted from Gokcen and Oden.[19]

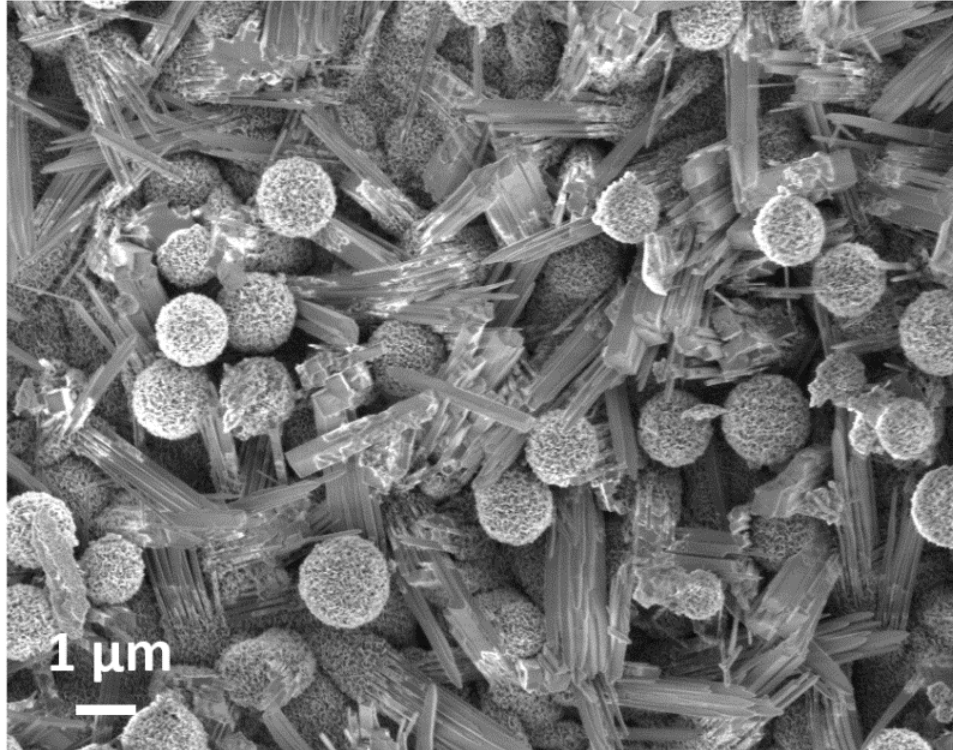


Figure S2. Scanning electron micrograph of the needle-like crystals in pre-HF washed AlC_x materials, exhibiting the morphology and composition of mullite.[23]

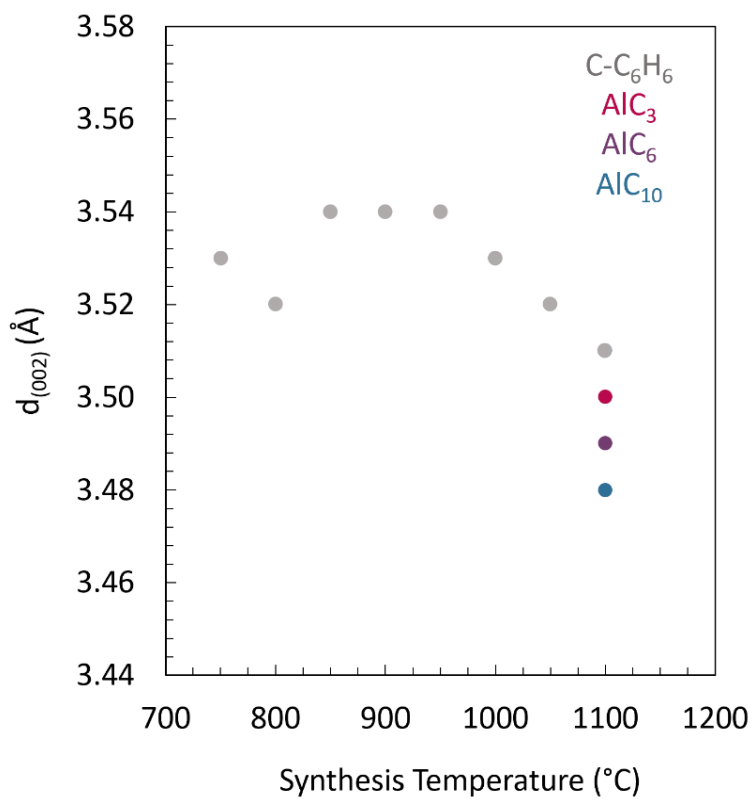


Figure S3. X-ray diffraction d-spacing of AlC_x-AlCl_3 and $C-C_6H_6$ materials.

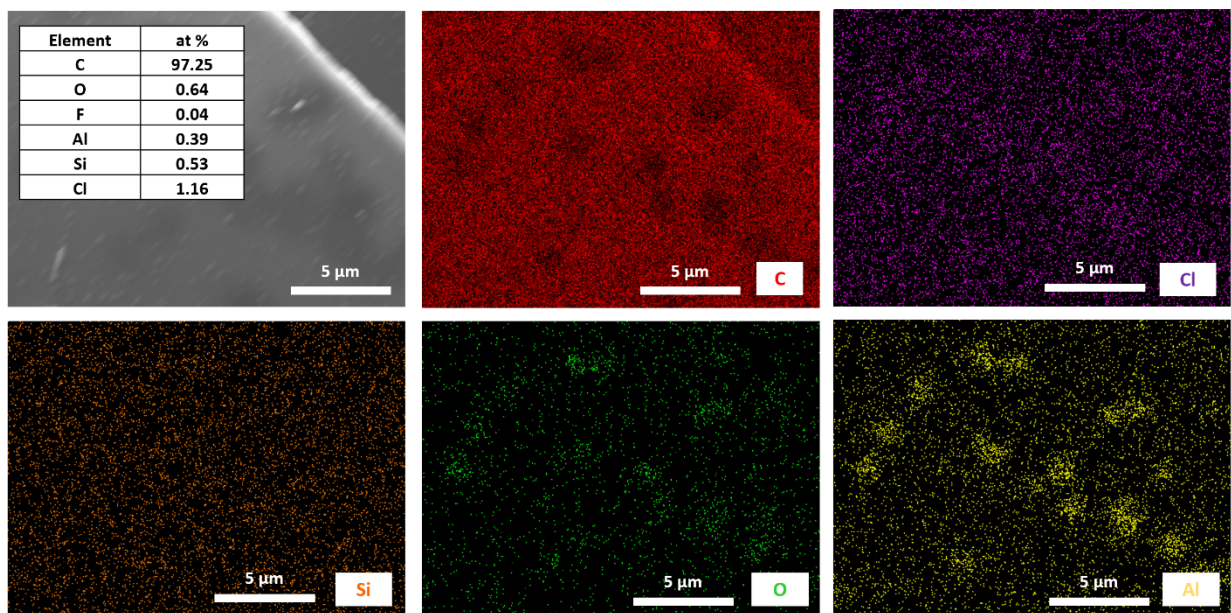


Figure S4. EDX chemical composition mapping of $\text{AlCl}_3\text{-AlCl}_3$ synthesized at 1100 °C.