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Microwave exfoliation of organicintercalated fluorographites

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Experimental section

Sample preparation. Organic-intercalated fluorographites were prepared from $C_2F \cdot 0.13CIF_3$ following exchange reactions, as described in our earlier report¹ (manipulation of strong fluorinating agents to obtain $C_2F \cdot 0.13CIF_3$ requires special safety precautions). To obtain C_2F with intercalated acetone molecules, acetone was added to $C_2F \cdot 0.13CIF_3$ and left for ~ 30 min. Within 3 – 4 days, repeated decantations were carried out, and fresh portions of acetone were added. As a result, after 4 – 5 of such operations, a complete replacement of the inorganic intercalant (CIF₃) was achieved, and di-carbon fluoride intercalated with acetone $C_2F \cdot x(CH_3)_2CO$ was obtained. Other organic intercalants were introduced in a similar manner, by treating acetone-intercalated fluorographite $C_2F \cdot x(CH_3)_2CO$ with a desired reagent (acetonitrile, benzene or carbon tetrachloride). As a result, intercalated compounds of di-carbon fluoride $C_2F \cdot xCH_3CN$, $C_2F \cdot xC_6H_6$, and $C_2F \cdot xCCl_4$ were obtained, respectively (*x* was varied in the range from 0.07 to 0.13).

Microwave heating. MW heating of the samples was carried out using a MW module "Discover-S-Class" ("CEM Corporation", USA), operating at 2.45 GHz. This device ensures continuous recording of the sample temperature and the overpressure in the reactor. The MW power is regulated in the interval from 0 to 300 W with a discreteness of 1 W. Initial intercalated sample mass was 0.13 g, and the yield of the MW exfoliation product was around 35 wt.%, owing to the escape of large volume of gaseous products from the evaporation of the intercalated phase and the decomposition of the C_2F host matrix.

Pollutant uptake measurements. To measure the uptake of liquid pollutants (kerosene, phenol, tetraethyllead and anhydrous nitric acid), MLG was weighted and covered with liquid in a beaker. After 1 h of impregnation, the contents of the beaker were emptied onto a pre-weighed polyethylene mesh, allowing the complete removal of the residual liquid. MLG was weighted again, and the uptake ("sorption") capacity was evaluated as the difference between the initial and final masses.² For oil, also an oil-on-water system was modelled. An oil film was created by adding 4 – 5 ml of oil to water surface, and then MLG (0.025 g) was added. The film was quickly (within a couple of minutes) disintegrated through sorption, and the water surface cleared.

Characterization. TEM images were obtained on the JEOL operating at an accelerating voltage of 200 kV. Samples for TEM were prepared by dispersing MLGs by ultrasound in ethanol and depositing it onto a carbon-coated Cu grid (200 mesh). Raman scattering spectra were recorded

on a LabRAM HR Evolution (Horiba) spectrometer (514 nm excitation). Elemental analysis method is described in.³

Table S1. Chemical analysis of the starting intercalates (white lines) and final graphitic products
(grey lines). Data for the starting intercalated compounds $C_2F \cdot xR$ (R = CH ₃ CN, CH ₃ COCH ₃ ,
C_6H_6 , CCl_4 , ClF_3) is taken from earlier works. ^{3,4}

Sample	Content, wt.% ¹			
	С	F	Cl	R ²
Intercalated compound $C_2F \cdot x_1ClF_3$	46.41	35.27	4.80	13.52
MLG from $C_2F \cdot x_1ClF_3$ (convective)	94.06	2.95	1.86	-
MLG from $C_2F \cdot x_1ClF_3$ (MW)	97.11	1.83	1.21	-
Intercalated compound C ₂ F·x ₂ (CH ₃) ₂ CO	47.90	35.58	4.77	13.75
MLG from $C_2F \cdot x_2(CH_3)_2CO$ (convective)	92.60	4.98	1.98	-
MLG from $C_2F \cdot x_2(CH_3)_2CO$ (MW)	96.27	1.88	1.29	-
Intercalated compound C ₂ F·x ₃ CH ₃ CN	50.71	39.34	0.37	9.58
MLG from $C_2F \cdot x_3CH_3CN$ (convective)	91.16	5.35	2.10	-
MLG from $C_2F \cdot x_3CH_3CN$ (MW)	94.68	2.15	1.41	-
Intercalated compound $C_2F \cdot x_4CCl_4$	42.00	30.57	4.70	22.73
MLG from $C_2F \cdot x_4CCl_4$ (convective)	92.47	5.53	1.99	-
MLG from $C_2F \cdot x_4CCl_4$ (MW)	95.88	2.23	1.39	-
Intercalated compound $C_2F \cdot x_5C_6H_6$	43.80	32.12	4.88	19.2
MLG from $C_2F \cdot x_5C_6H_6$ (convective)	93.20	4.81	1.82	-
MLG from $C_2 F \cdot x_5 C_6 H_6$ (MW)	95.33	2.21	1.35	-

¹MLG samples also contain < 1wt% of H.

 ${}^{2}R$ = calculated content of the intercalated molecules. Analytical method is described in more details in previous work.³



Fig. S1. Microscopy images of MLGs obtained by exfoliation of $C_2F \cdot CH_3COCH_3$: (a) SEM image, rapid convective heating; (b) SEM image, MW heating; (c) TEM image, rapid convective heating; (d) TEM image, MW heating.



Fig. S2. Raman spectra of the starting intercalated fluorographite $C_2F \cdot x CH_3CN$ (lower red line) and MLG derived from it by MW treatment (upper black line). Disorder-induced D peak at ~1357 cm⁻¹, G peak at 1584 cm⁻¹ (sp² carbons; positioned at 1580 cm⁻¹ in pure graphite), $I_D/I_G =$ 1.3.Inset shows 2D peak region for the MLG. 2D peak at 2707 cm⁻¹. Other precursors show similar spectral characteristics.



Fig. S3. (a) Sorption capacities of MLGs obtained by MW-assisted exfoliation of different precursors $C_2F \cdot xR$ (R = CH₃CN, CH₃COCH₃, C_6H_6 , CCl₄, ClF₃) plotted against sample's surface areas (S_{BET,} m²/g); (b) photographs of oil on the water surface before (upper photo) and after (lower photo) the addition of the MLG sample.

Supporting references

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