EXPERIMENTAL METHODS

MAX phase synthesis into MXene: Synthesis of MAX phase powders into MXene was carried out following the methods described in the literature.[1,2] Ti_2AlC powder precursor was obtained commercially (KANTHAL, Maxthal 211 Ti_2AlC). Etching of the aluminium from Ti_2AlC was conducted in a 20% aqueous hydrofluoric acid, HF, solution (Sigma Aldrich) for 24 hr at room temperature. The resulting suspension was filtered and washed with deionised water (DI) to reach a pH > 6. The etched MXene powder was then freeze dried for ~ 24 hr (HETO PowerDry PL3000) (obtained yield *ca.* 35%). *p*-Phosphonic acid calix[*n*]arenes (*n* = 4, 5, 6 and 8) were prepared according to literature procedure.[3]

Ultrasonication of MXene: Solutions of aluminium etched MXene Ti_2C (M) with varying *p*-phosphonic acid calix[*n*]arene (PCX*n*, where *n* = 4, 5, 6 and 8) compounds were prepared at set concentration of 2 mg ml^{-1} MXene and 5 mg ml^{-1} PCX*n* in DI water purged with argon to prevent oxidation. These solutions were each ultrasonicated using a MSE SONIPREP 150 Ultrasonic Disintegrator under a 0.5 bar Argon flow (BOC, PureShield Argon). The dispersed sonicated solutions where centrifuged at 1500 rpm for 30 min and decanted to obtain successfully exfoliated MXenes from the supernatant. The resulting colloidal supernatant was then ultracentrifuged (Thermo Electron Corporation, SORVALL RC6 plus), and washed with Ar purged DI water at 17,000 rpm for four 20 min cycles. The ultrasonicated MXene powders were freeze-dried (HETO PowerDry PL3000) for ~ 16 hrs (obtained yield ranging from *ca.* 20-36% for the control and the MPCXn samples).

Characterisation:

Scanning Electron Microscopy imaging (SEM) of all MXene samples were acquired using a FEI Helios Nanolab 600i microscope. Powdered samples were prepared by sputter coating approximately 10 nm of chromium prior to characterisation to ensure conduction and to produce a smooth layer. Each MXene sample was imaged at both 20 and 3 kV. The images displayed in Fig 2(b-f) were recorded at 3kV.

Transmission Electron Microscopy imaging (TEM) for particle size and morphology analysis of as-prepared MXene samples were investigated using a JEOL 2100FCs TEM, equipped with a Schottky Field Emission Gun set to 200 kV accelerating voltage. Samples were collected on carbon-coated copper grids (Holey Carbon Film, 300 mesh Cu, Agar Scientific, UK) following brief ultrasonication in water for dispersal. Particle size analysis, where applicable, was performed using ImageJ particle size analysis software. The lattice spacing was calculated by measuring the distance from the center of the upper most lattice all the way to the lowest lattice, which was then divided by the number of lattices to obtain an average value. The MXene samples were re-dispersed in DI water following powder form.

X-ray photoelectron spectroscopy (XPS) measurements were performed using a Kratos Axis ultra DLD photoelectron spectrometer utilizing monochromatic Alka source operating at 144 W. MXene samples were mounted using conductive carbon tape. Survey and narrow scans were performed at constant pass energies of 160 and 40 eV, respectively. The base pressure of the system is *ca.* 1×10^{-9} Torr, rising to *ca.* 4×10^{-9} Torr under analysis of these samples.

Atomic Force Microscopy: A JPK Nanowizard II atomic force microscopy (AFM) instrument was used in intermittent contact mode with NCHV Bruker probes to image the surface of the materials. The image data was processed with levelling and polynomial subtraction to reveal the surface topography and profile.

Raman Spectroscopy: Raman spectra were collected using a Renishaw Raman system using the 488 nm line of an Ar+ ion laser at a power of ~10 mW.

Powder X-Ray Diffraction (PXRD) analysis samples was performed on a Bruker D8 Advance diffractometer equipped with Goebel mirrors for Cu-Kα primary parallel radiation and a Vantec position sensitive detector, operating at 40 mA and 40 kV. The samples were printed as films and were measured without any further preparation. Sample M-PCX6 was measured as a powder on a silicon low background sample holder.

Figure SI1: AFM images of MXene samples.

Pawley refinements

Pawley refinements were performed with software Topas V 4.1.[4] A shifted Chebyshev function with ten parameters was used to fit the background. LaB6 660b NIST standard was used to model the instrumental contribution to peak broadening. A Lorentzian size related isotropic function was used for the sample contribution. The samples were measured on the filters as film without any further preparation. For this reason, a correction for specimen displacement was used to account for the peak shift due to sample height which was found to be in the order of 1°. Early refinement trials using the unit cell and symmetry from the literature showed the *c* parameter corresponding to the layer stacking periodicity to be correct but could not fit all peaks in the pattern (see Figure SI2). The experimental pattern could be fitted by doubling the *a* parameter while keeping the same space group, $P6_3/mmc$ (see Figure SI3, SI4, SI5, SI6).

Figure SI7 shows a plot of the diffraction pattern of sample M-PCX6 that was not included in the data analyses because of its amorphous nature.
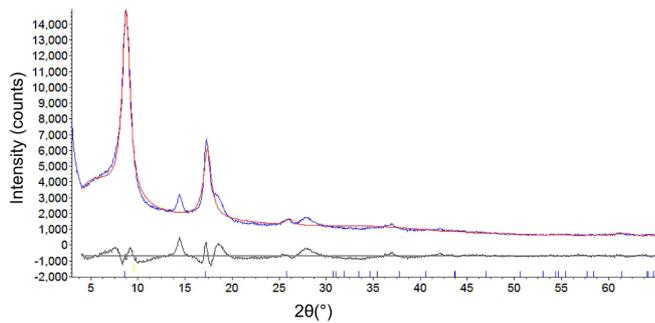
Figure SI2: Experimental (blue curve), calculated (red curve) and difference (grey curve) patterns for the Pawley refinement of control sample using the unit cell and space group reported in the literature.
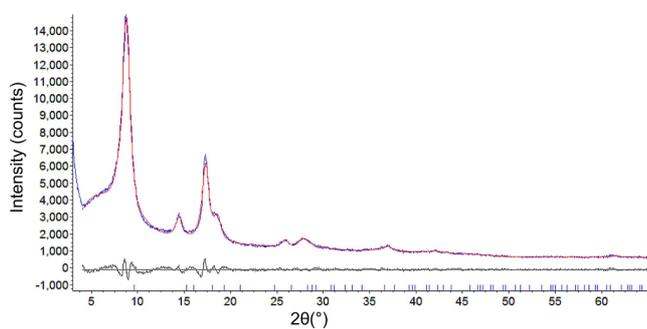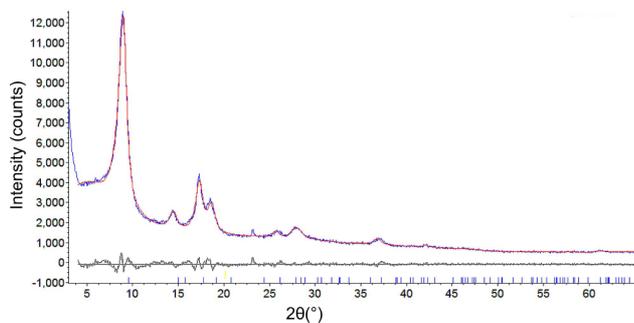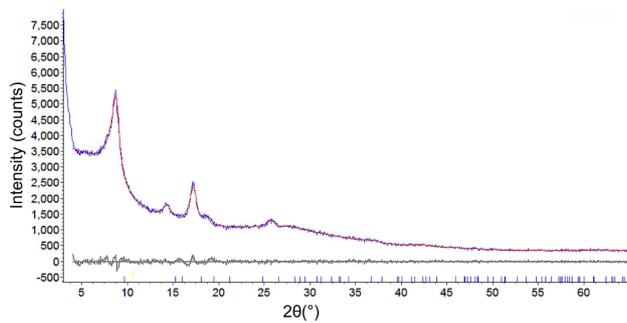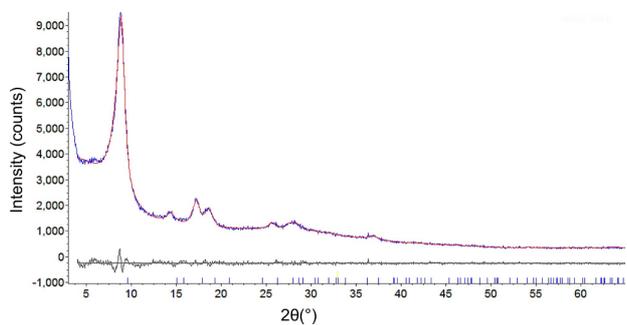

Figure SI3: Experimental (blue curve), calculated (red curve) and difference (grey curve) patterns for the Pawley refinement of control sample.

Figure SI4: Experimental (blue curve), calculated (red curve) and difference (grey curve) patterns for the Pawley refinement of M-PCX4 sample.

Figure SI5: Experimental (blue curve), calculated (red curve) and difference (grey curve) patterns for the Pawley refinement of M-PCX5 sample.

Figure SI6: Experimental (blue curve), calculated (red curve) and difference (grey curve) patterns for the Pawley refinement of M-PCX8 sample**.**
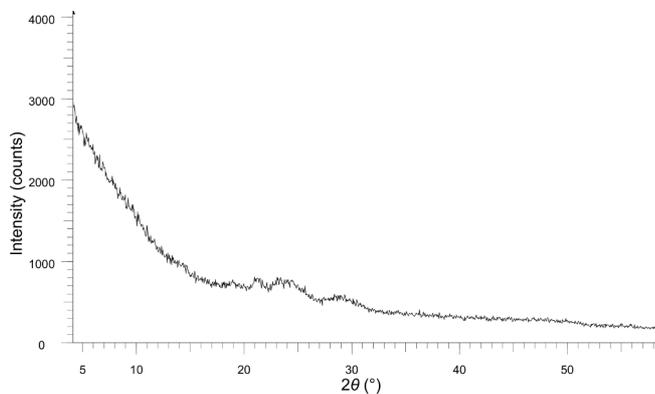

Figure SI7: PXRD pattern of M-PCX6 sample.

Similarity analysis of M-PCXn samples powder diffraction data

The analysis was performed using R studio.[5]

Data Treatment

Diffractograms were background subracted using the R library "baseline".[6] Peaks were aligned using the R library "MALDIquant".[7] Principal component analysis results were plotted using "ggbiplot",[8] while the correlation plot was created using "corrplot".[9] The background subtracted diffractograms were then normalised according to the height of their most intense first peak (see Figure SI8); finally the average pattern was subtracted from each resulting pattern to bring out the differences between them (see Figure SI8).

Figure SI8: Normalised diffractograms for principal component analysis.
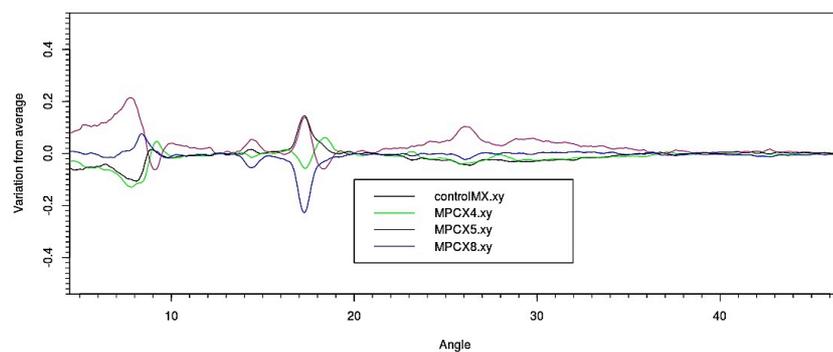

Figure SI9: Average subtracted patterns for principal component analysis.

Principal Component Analysis (PCA)

Principal component analysis was performed on control, M-PCX4, M-PCX5, M-PCX8 samples. The method provides as few common pattern components as possible to explain variations in the dataset given. In this case two principal components were identified, PC1 and PC2 which explain 70% and 23% of the entire variation respectively (see Figure SI10).
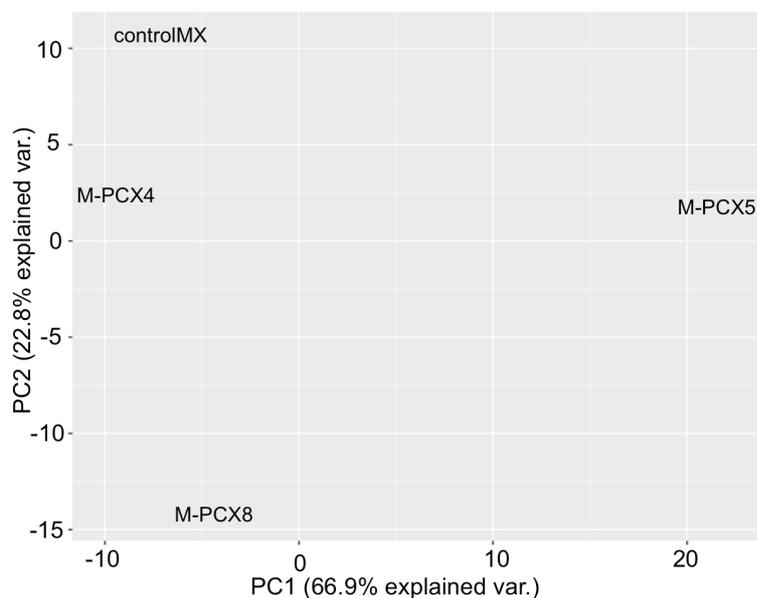

Figure SI10: Pattern positions in the principal component plane.

Correlation analysis

Similar results are produced using correlation analysis (see Figure SI11). M-PCX4 correlates positively with control sample. The correlation with control sample is instead negative in M-PCX5 and M-PCX8. M-PCX4 and M-PCX5 also anti-correlates between each other. M-PCX4 and M-PCX5 seems not to have any correlation with M-PCX8.
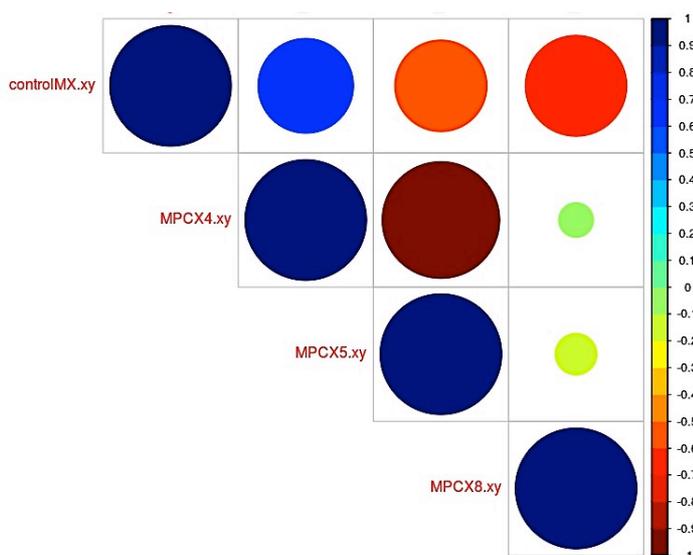

Figure SI11: Correlation plot for control, M-PCX4, M-PCX5, M-PCX8 samples. The correlation value is indicated by the colour bar.
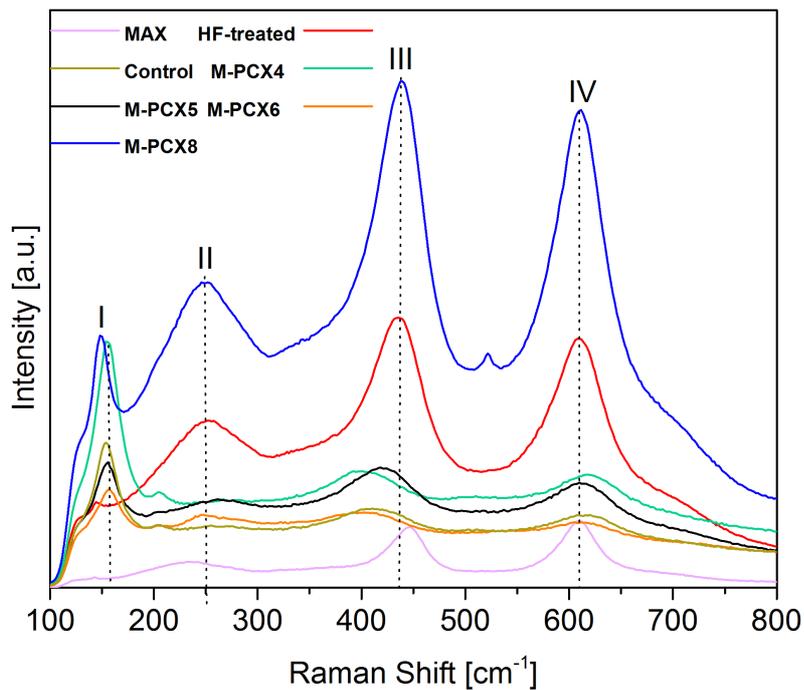
Figure SI12: Raman spectra for MXene samples.

Extended portofolio of electron microcopy images of MXene samples
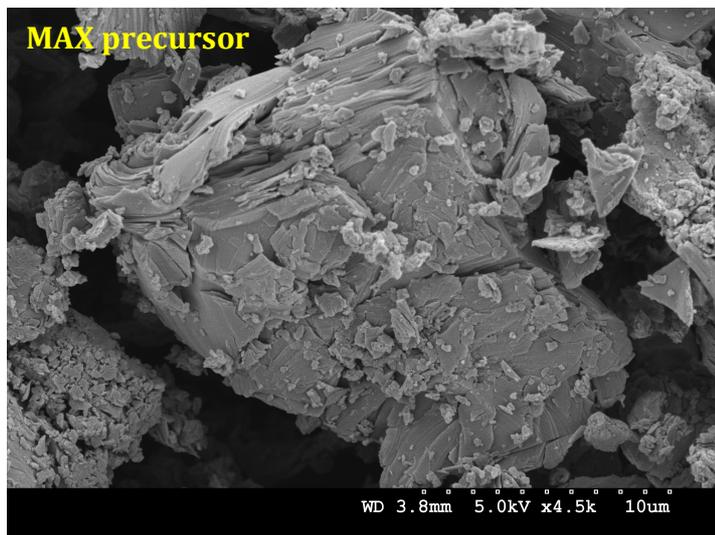

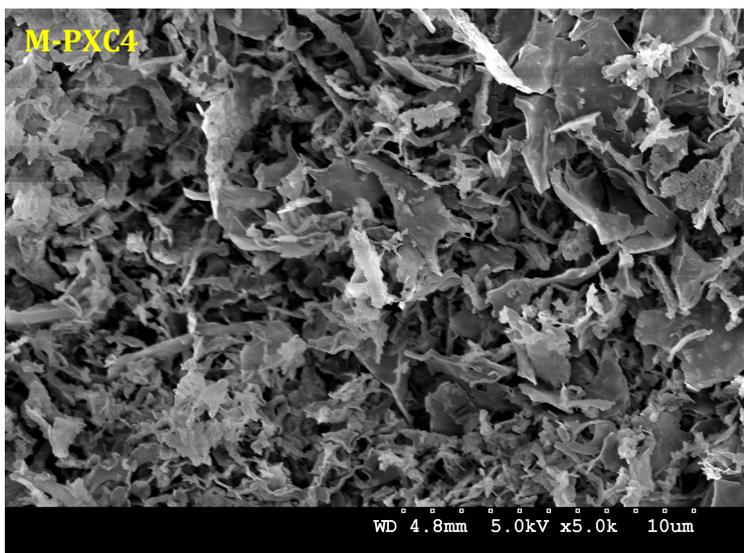**Figure SI13:** SEM images of MAX precursor.

Figure SI14: SEM images of M-PCX4 assisted sonication of MXene displaying thin sheet like morphology.
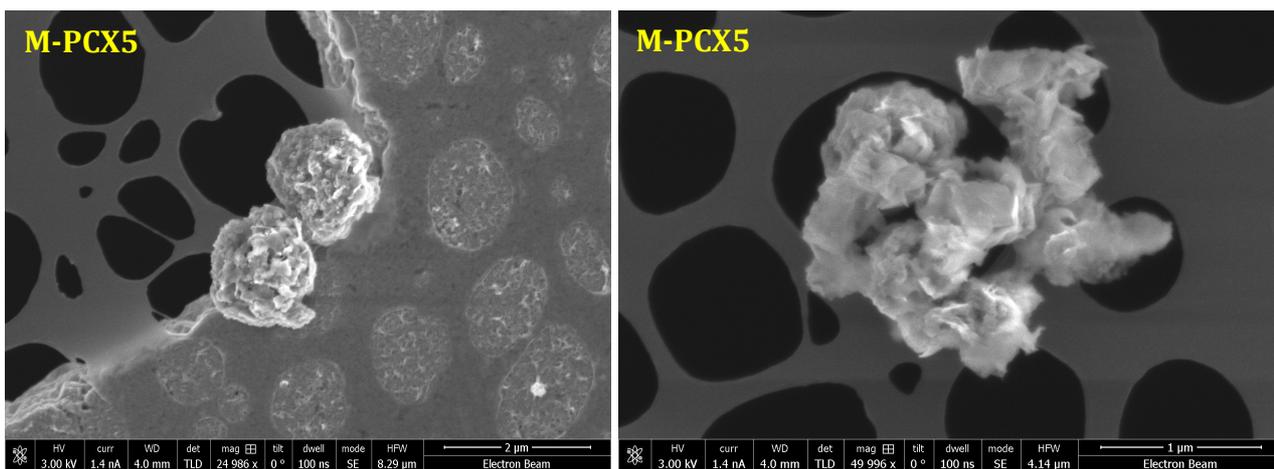

Figure SI15: SEM images of M-PCX5 assisted sonication displaying crumpled MXene sheets morphology.
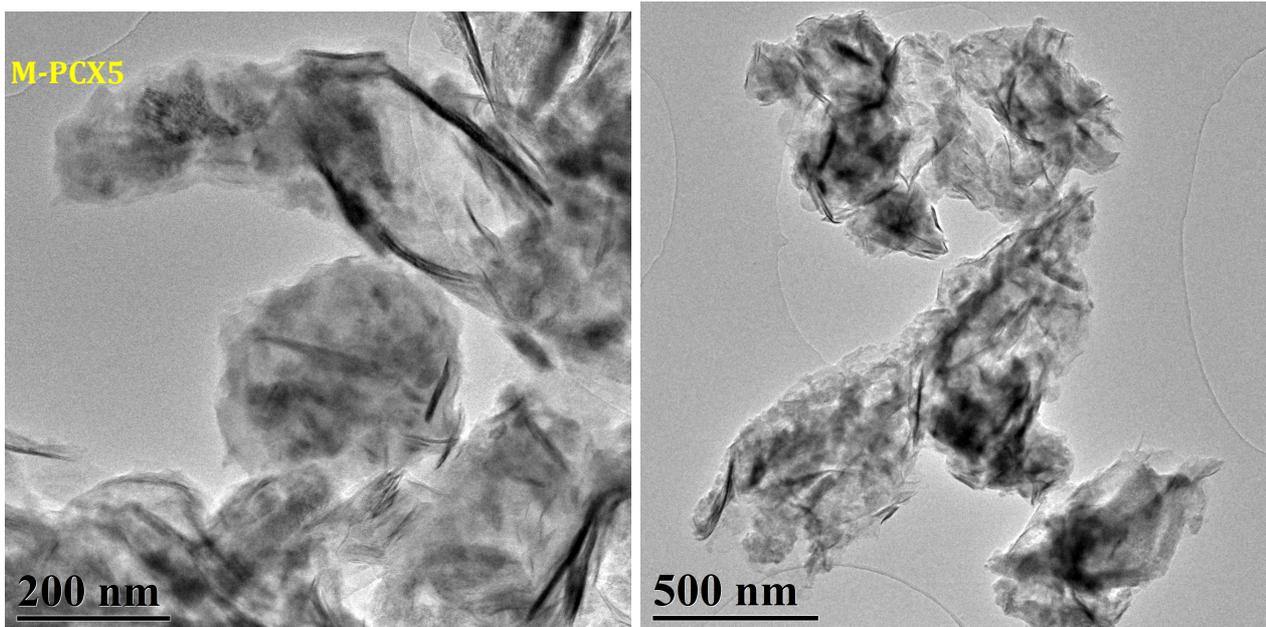
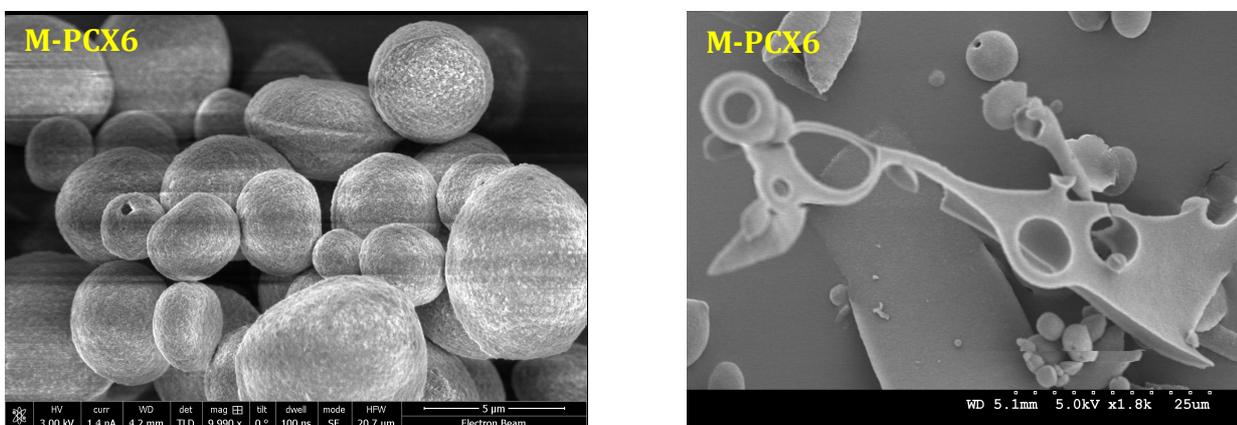**Figure SI16:** TEM images of M-PCX5 assisted sonication displaying crumpled MXene sheets morphology.

Figure SI17: SEM images of M-PCX6 assisted sonication of MXene displaying spherical morphology.
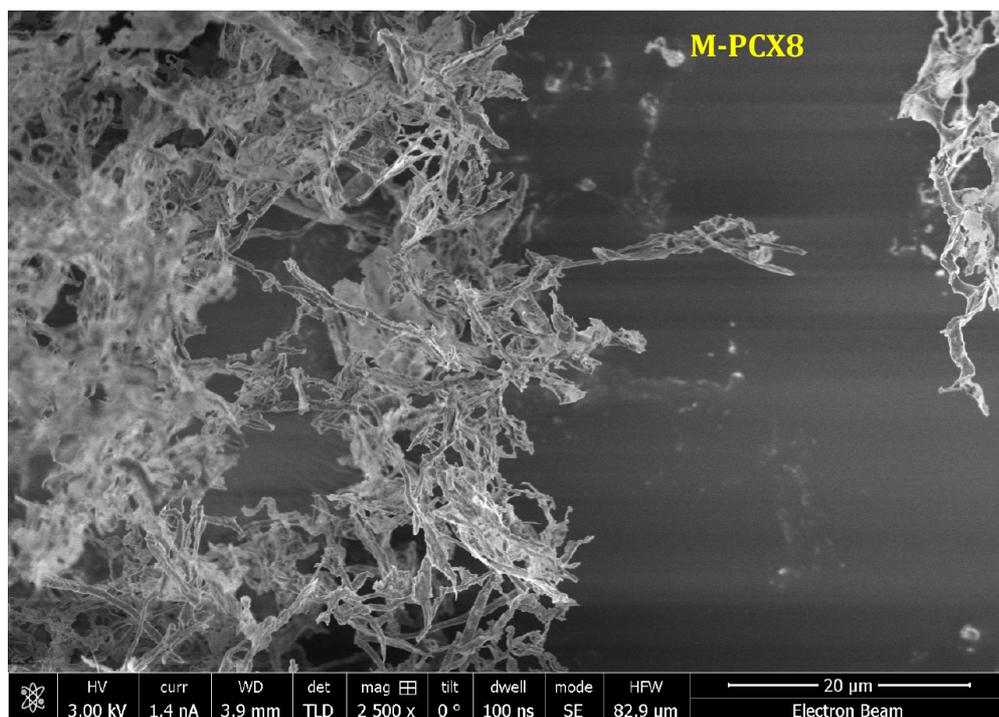
Figure SI18: SEM images of M-PCX8 assisted sonication displaying the formation of MXene scrolls.

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