SUPPLEMENTARY INFORMATION

Spontaneous MXenes monolayer assembly at liquid-air interface

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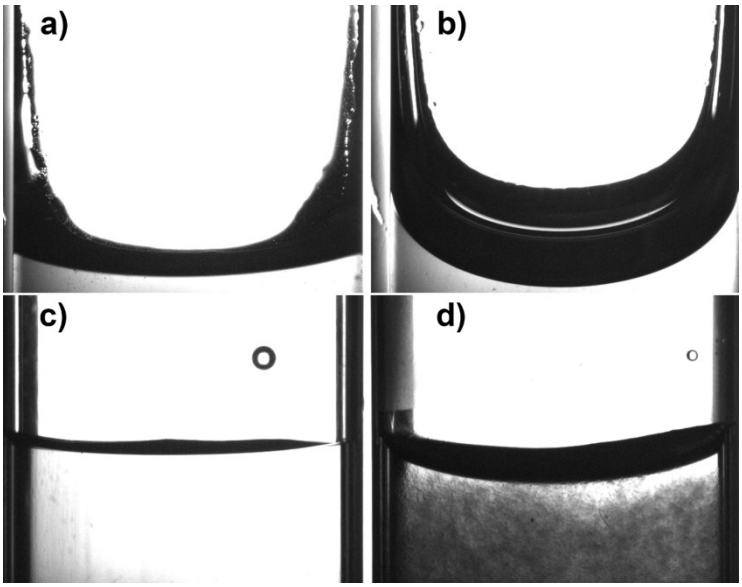
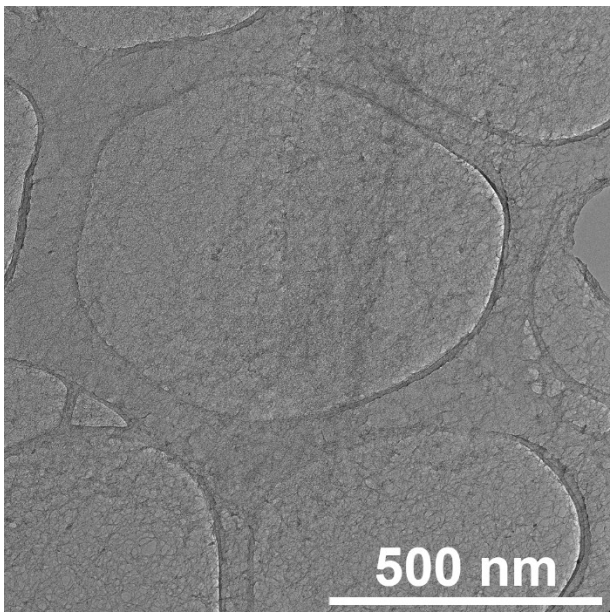
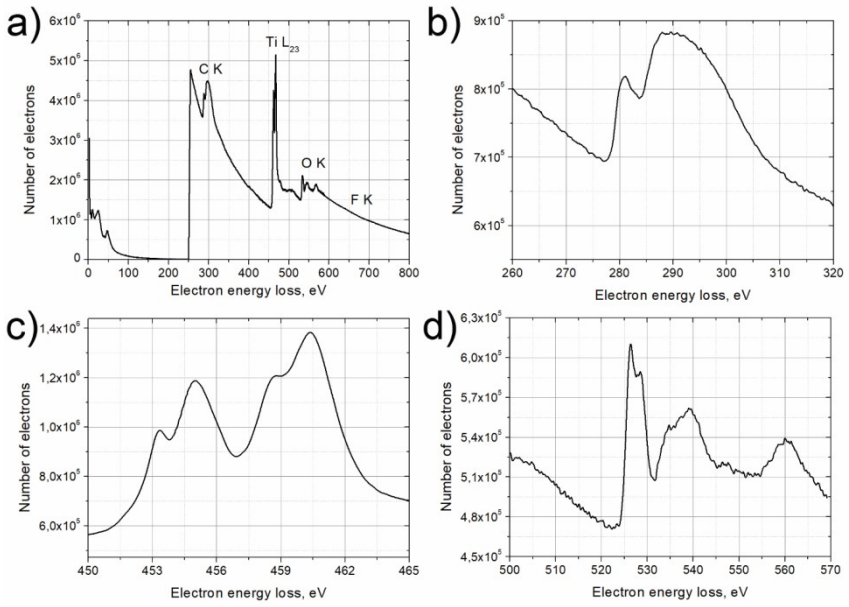


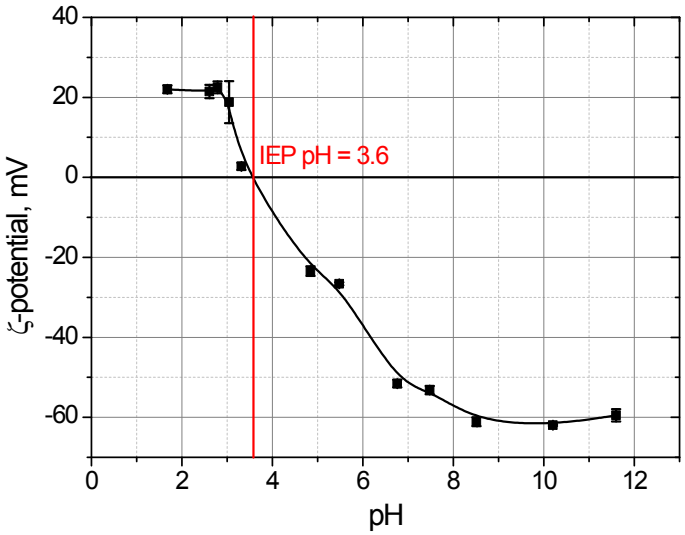
SI 1. Photographs of MXene suspensions with pH = 8 (a, c) and pH < 1 (b, d) in quartz (a, b) and PMMA (c, d) cuvettes.



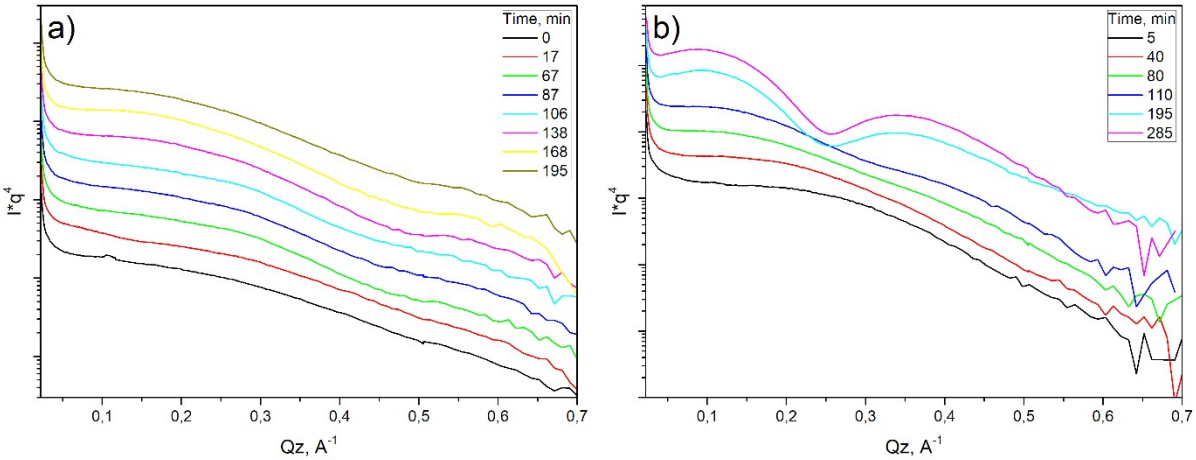
SI 2. Low resolution TEM image of continuous MXene film formed on lacey carbon microscopy grid.



SI 3. EELS survey spectrum and high-resolution spectra for Ti-L, C-K, and O-K edge regions. Data illustrates preferred coordination of titanium to oxygen atoms, while revealing only residues of fluorine and poor Ti-C binding (~10% of total titanium in the sample). Note some carbon signal can be originated from lacey carbon TEM grid.



SI. 4. Zeta potential dependence of MXene particles vs. pH of the suspension.

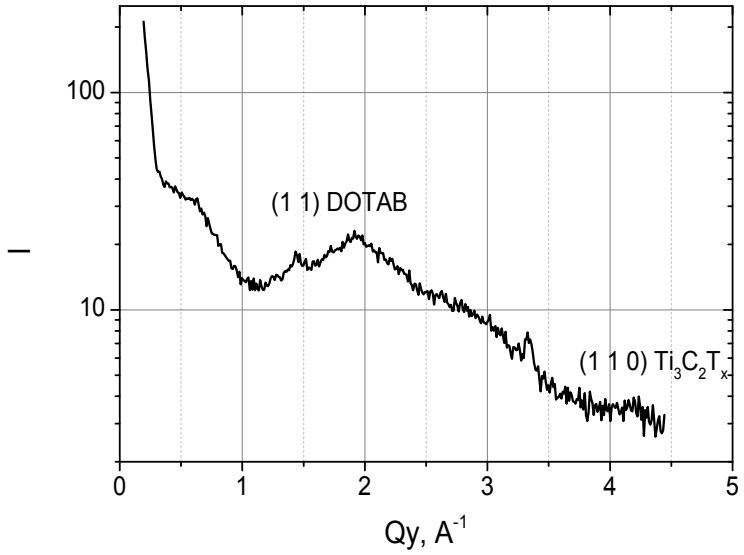


SI 5. XRR spectra time series of liquid-air interface for basic (pH = 8, a) and acidic (pH < 1, b) suspensions. Curves are shifted by multiplying a factor of 2 for clarity.

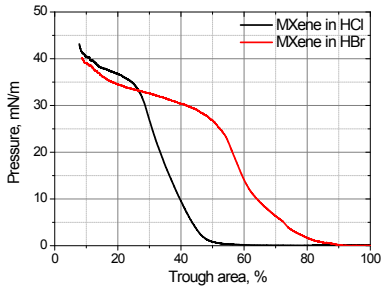
SI 6. Derived XRR fit parameters for modeled surface layers: thickness (d), roughness (σ), maximum scattering length density (SLD) of surface layers and calculated surface coverage for XRR spectra series acquired at liquid-air interfaces on basic (pH = 8) and acidic (pH < 1) suspensions of MXenes as well as DOTAB/$Ti_3C_2T_x$/subphase interface. Theoretical SLDs for MXenes monolayer and experimental best-fit parameters for pure water and DOTAB/water are given for reference.

Sample	Time (min)	Layer	D (A)	Sigma	SLD	Surface coverage (%)
Water	-	-	inf	2.98	0.410	-
DOTAB	-	tails	12.23	4.12	0.367	-
		heads	3.14	5.26	0.451	-
Mxenes (theory)	-	-	-	-	0.966	100.0
MXenes pH = 8 + DOTAB	-	tails	16.36	6.2	0.285	-
		heads	5.48	2.6	0.577	-
		MXene	15.28	11.1	0.383	49.1
MXenes pH = 8	0	MXene	15.16	3.07	0.423	54.2

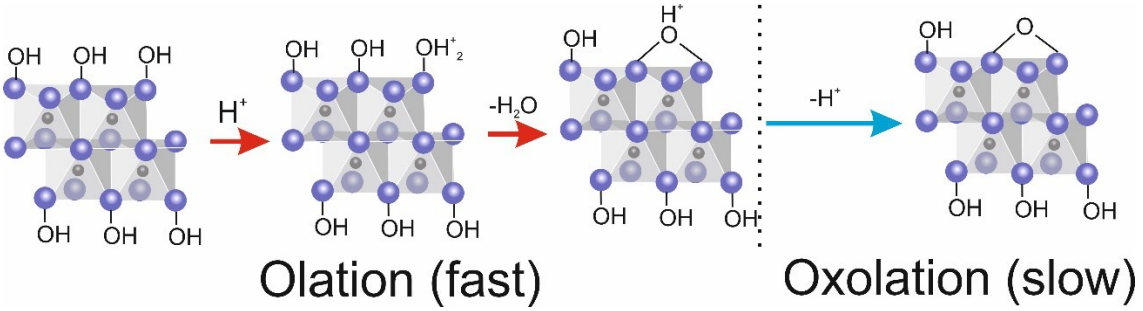
	17		15.31	3.11	0.421	53.9
	67		15	3.23	0.431	55.2
	87		15.48	3.24	0.431	55.2
	106		15.42	3.24	0.432	55.3
	138		15.16	3.36	0.437	56.0
	168		15.8	3.41	0.442	56.6
	195		15.45	3.3	0.433	55.5
Mxenes pH < 1	5	MXene	8.8	3.1	0.381	48.7
		anion	9.1	6.06	0.602	-
	40	MXene	11.33	3.16	0.422	54.0
		anion	8.77	5.68	0.618	-
	80	MXene	13.71	3.08	0.446	57.1
		anion	8.61	6.07	0.648	-
	110	MXene	13.04	2.89	0.483	61.9
		anion	7.21	5.68	0.633	-
	195	MXene	13.52	3.23	0.695	89.0
		anion	4.95	7.91	1.103	-
	285	MXene	13.28	3.94	0.750	96.1
		anion	3.02	7.37	1.354	-



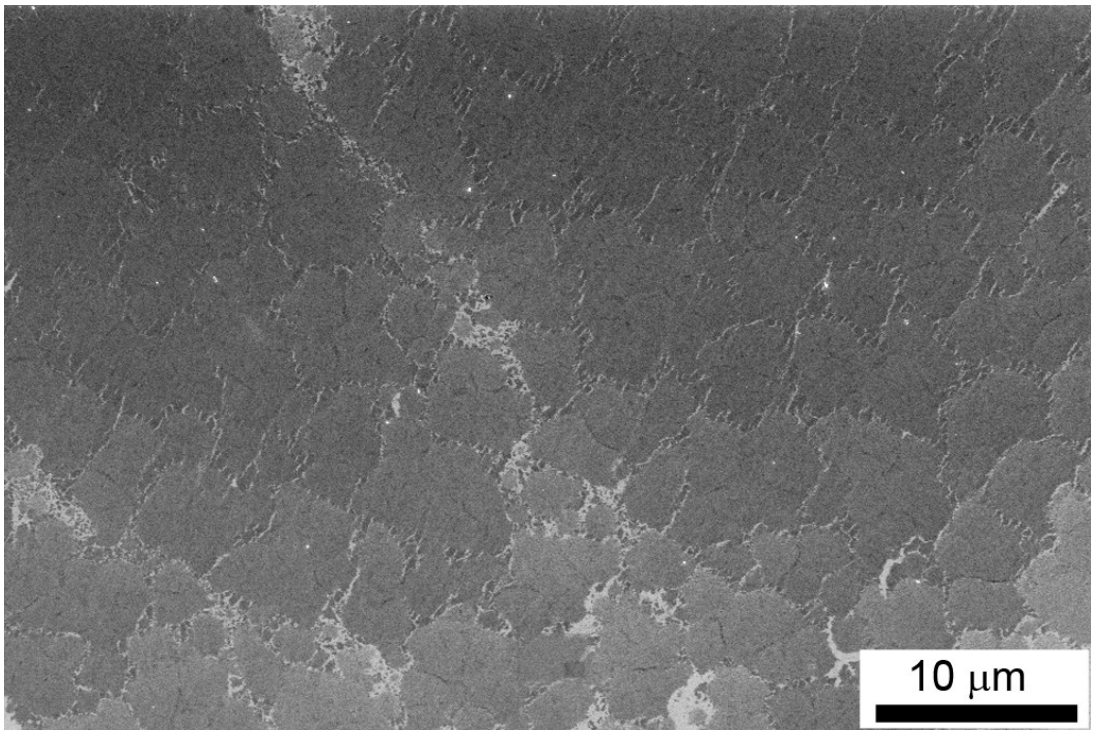
SI 7. Grazing incidence (μ = 0.05°) X-ray diffraction (GIXRD) pattern for DOTAB/$Ti_3C_2T_x$/subphase interface at out-of-plane reflection angle θ = 0..0.1° and variable in-plane (δ) reflection angle. GIXRD indicates both in-plane assembly of DOTAB molecules and $Ti_3C_2T_x$ diffraction maximum at ~4,2 $Å^{-1}$ (~1,5 Å), corresponding to (110) $Ti_3C_2T_x$ lattice reflection. Large width of MXene reflection is indicative for poor periodicity. As the reflection appears in q_y direction corresponding diffracting planes are strictly perpendicular to the plane of the film corresponding to in-plane texturing. Note the appearance of the diffraction maximum in the GIXRD geometry with an angle of incidence below critical angle, correspond to diffracting planes laying strictly in the surface layer.



SI. 8. Surface layer compression isotherms of MXenes suspensions at pH < 1 on HCl and HBr solutions.

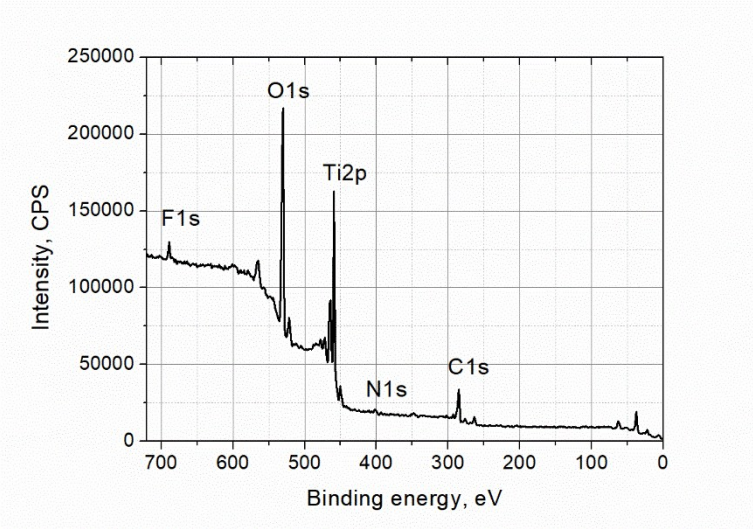


SI. 9 Scheme of olation and oxolation process on MXenes flakes.



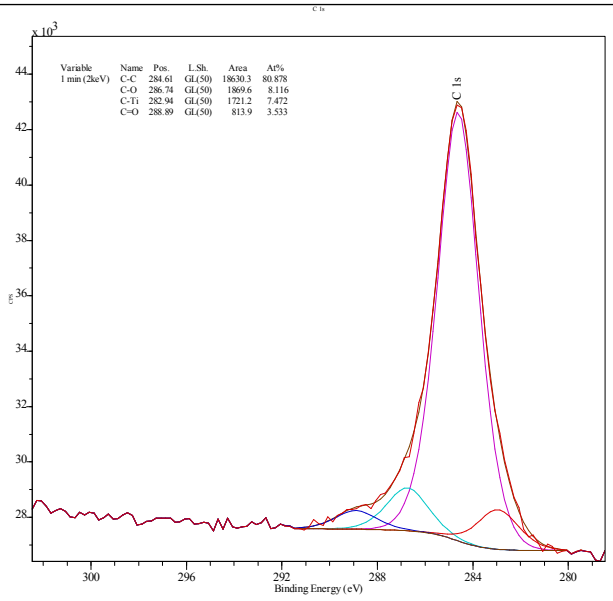
SI. 10 Low magnification SEM image for MXene films formed at liquid-air interface from acidic suspension at the surface tension σ=20 mN/m and compression degree of ~2.5 transferred onto silicon substrates by Langmuir-Blodgett method demonstrating uniform coverage on a large area.

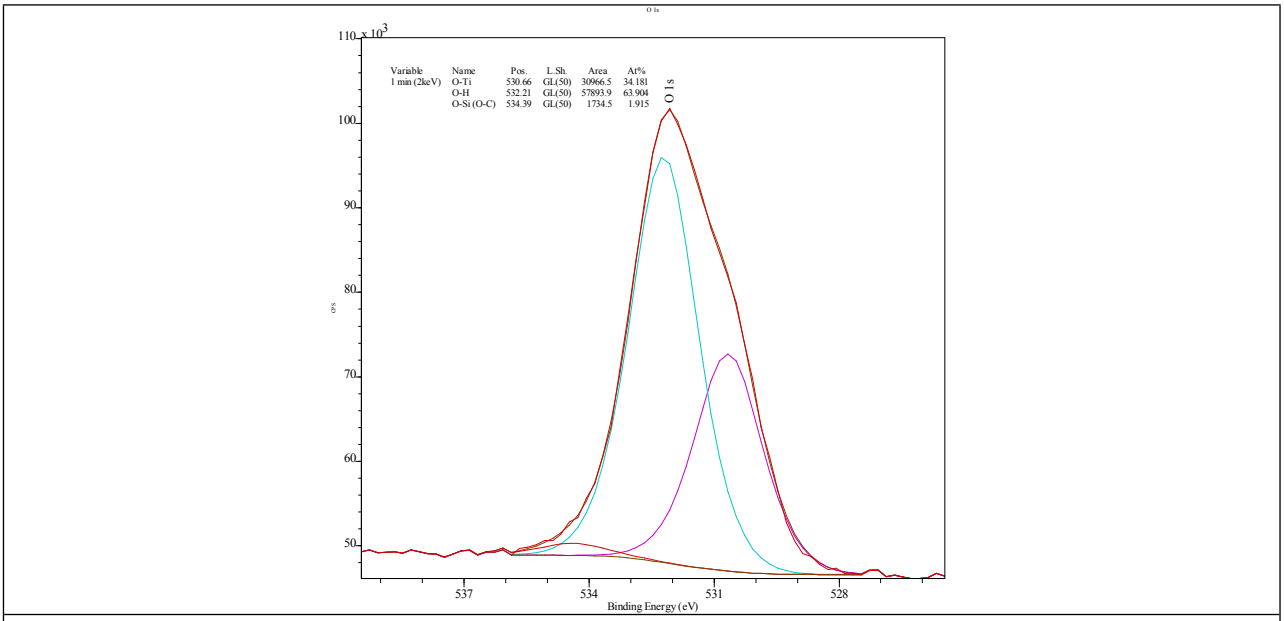
SI. 11 Panoramic XPS spectrum and Spectra for Ti2p, C1s and O1s regions for MXene film transferred on silicon substrate



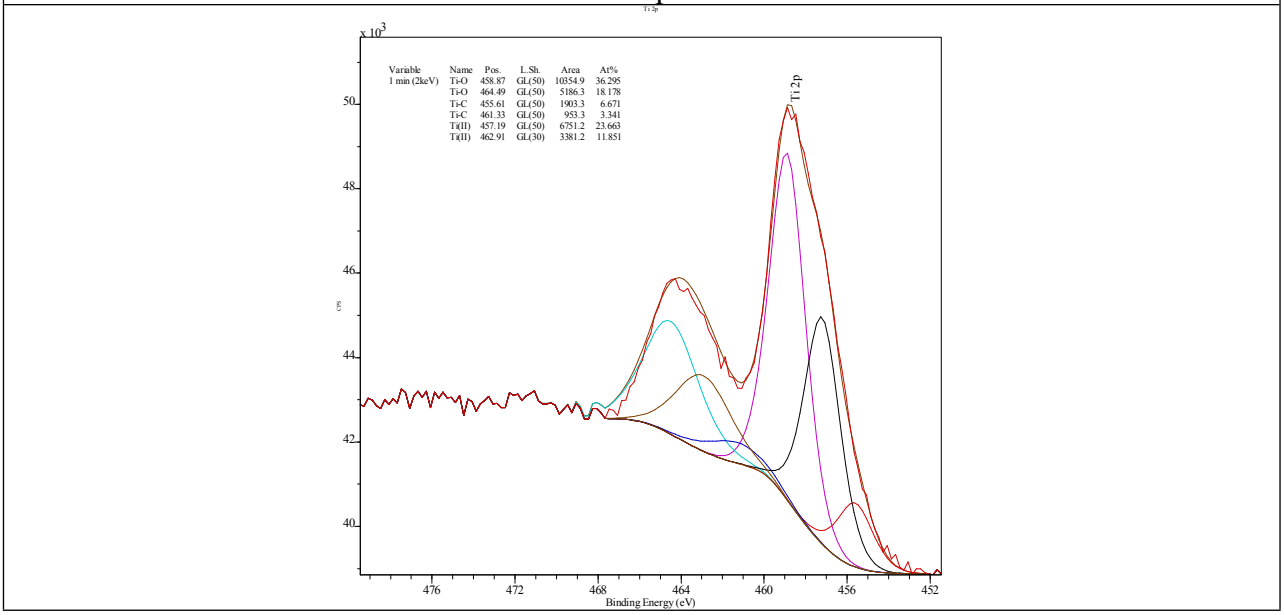
Overall formal chemical composition:

$Ti_3C_{1.5}O_{2.85}(OH)_{1.35}F_{0.15}$

C1s

O1s



Ti2p



Deconvolution of C1s spectra

	C-C	C-O	C=O	C-Ti
E, eV	284.6	286.7	288.9	282.9
c, %	80.89	8.12	3.53	7.47

Deconvolution of O1s spectra

	Ti-O	O-H	H_2O
E, eV	530.75	531.95	533.13
c, %	29.31	61.23	9.46

Deconvolution of Ti2p spectra

	Ti-O (4+)	Ti-O (2+)	Ti-C	Ti-O (4+)	Ti-O (3+)	Ti-C
E, eV	458.9	457.2	455.6	464.5	462.9	461.3
c, %	54.5	35.5	10.0			