

## Supplementary for High capture capacity of CO<sub>2</sub> in nickel intercalated Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene-Fluorohectorite clay heterostructure

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### 1. Details of experimental methods

#### 1.1 AFM

For the AFM measurements, 0.1 wt% water solution of delaminated clay nanosheets was drop-casted on plasma functionalized Si@SiO<sub>2</sub>(100 nm) substrate. The sample was scanned using MultiMode AFM operating with the Nanoscope III controller. Images were captured in tapping mode, using TAP190Al-G AFM probes purchased from Budget Sensors. Images were processed using Gwyddion software<sup>1</sup> by aligning rows, flattening and scar corrections.

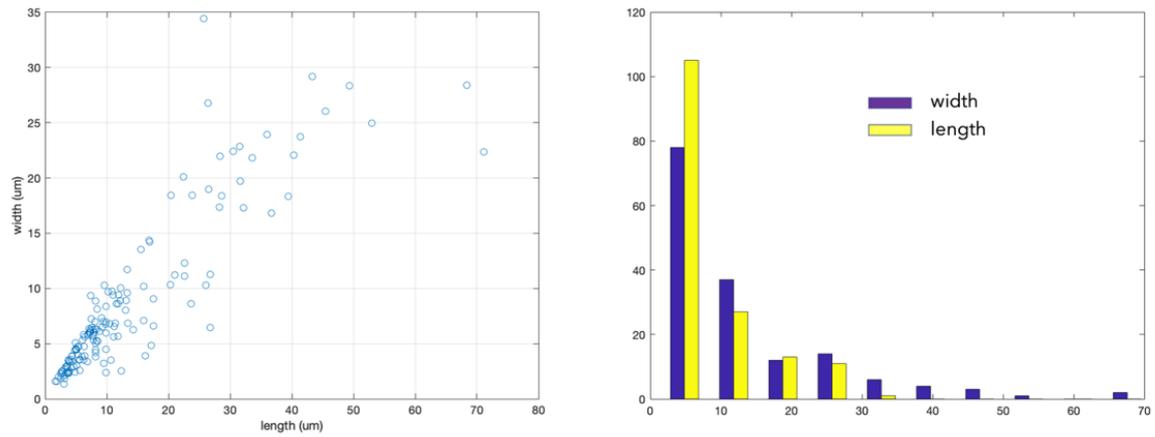
#### 1.2 Gravimetry

Before measurement, the sample was outgassed at 120 °C for 3 hours under secondary vacuum. The pressure is measured with Tecsis-Series P3382. The temperature was recorded with Pt-100 sensor placed close to the sample crucible. The complete system is placed into a heating chamber to maintain constant temperature within  $23 \pm 0.5$  °C. The suspension balance has a resolution of 10 µG. The sample mass variation  $m_{meas}$  (g) is measured as well as pressure and temperature when equilibrium is reached (with our criterion, when four of the five last mass measurements (noticed each 5 min) are included in an interval of 50 µg). The measured quantity is the excess adsorbed amount, which is obtained by correcting for the buoyancy of the skeletal volume of the sample material and the suspended metal parts. This volume is evaluated by a direct helium buoyancy effect measurement, according to helium not adsorbed at high pressure (10 bar up to 100 bar). The gas phase densities were determined using the equation of state for CO<sub>2</sub><sup>2</sup> and Helium<sup>3</sup>.

### 2. Additional data

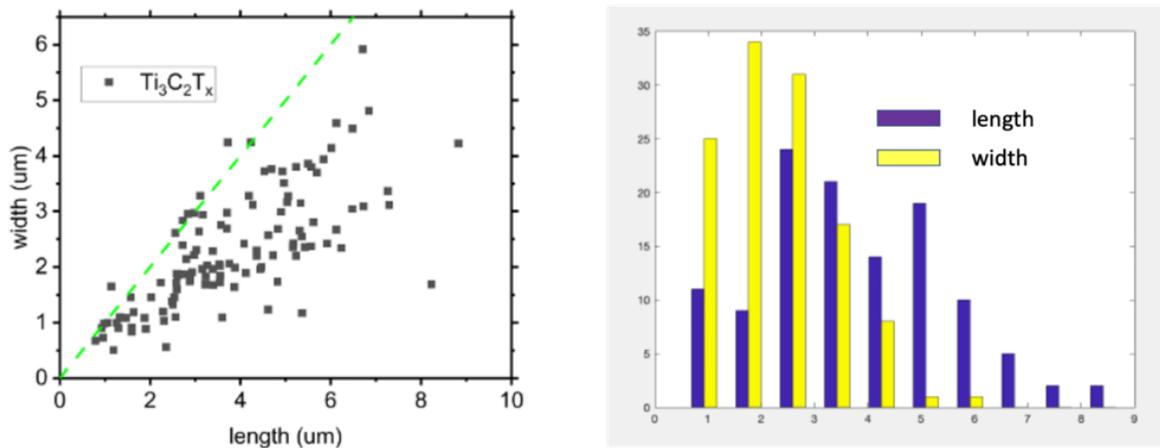
## 2.1 Atomic force microscopy

### Synthetic fluorohectorite clay



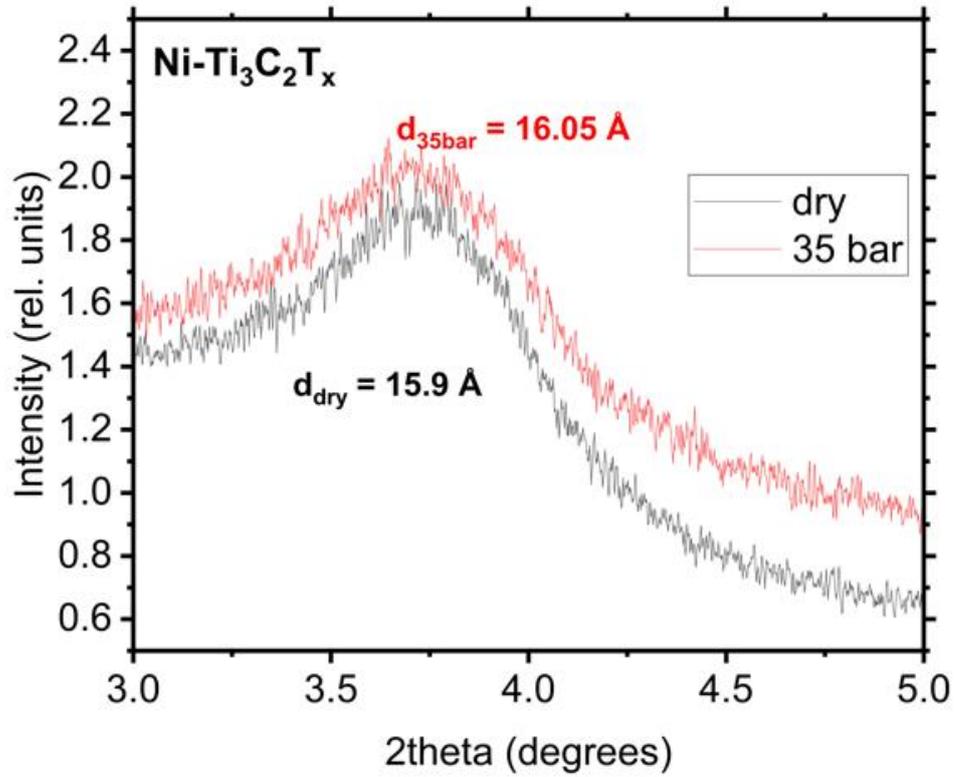
**Figure S1.** Left: width and length of the individual single Fht nanosheets. Right: histogram of the widths and lengths of all flakes.

### Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene



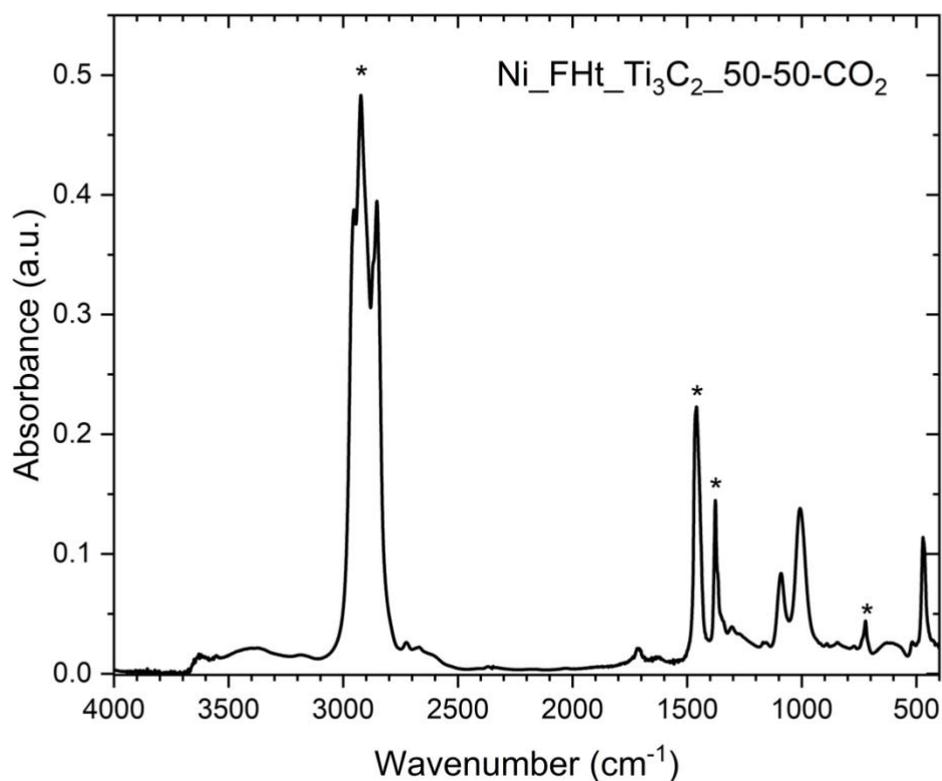
**Figure S2.** Left: width and length of the individual single MXene flakes. Right: histogram of the widths and lengths of all flakes.

## 2.2 CO<sub>2</sub> capture in Ni-Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> reference sample



**Figure S3.** Synchrotron X-ray diffraction pattern of Ni-Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene sample in dry state and exposed to 35 bars.

## 2.3 Infrared spectroscopy



**Figure S4.** FTIR spectra of the Ni-MXene-clay heterostructure.

The best FTIR spectra were measured by the transmission technique. The bands marked by an asterisk are assigned to the vibrational bands of nujol (paraffinum liquidum). The bands at 1090, 1008, and 471 cm<sup>-1</sup> in the **Ni-FHt-Ti<sub>3</sub>C<sub>2</sub>-50/50 CO<sub>2</sub>** FTIR spectrum are associated with the vibrational behaviour of layered silicate. The broad bands in the region 3700-3200 cm<sup>-1</sup> can be assigned to the stretching vibration of O-H bonds of  $\alpha$ -Ni(OH)<sub>2</sub> involved in hydrogen interactions in the FTIR spectrum.

Unfortunately, the presence of CO<sub>2</sub> in the **Ni-FHt-Ti<sub>3</sub>C<sub>2</sub>-50/50 CO<sub>2</sub>** sample cannot be verified by the method FTIR.

### 3. Summary of CO<sub>2</sub> adsorption capacity for different materials

Adsorbent	CO <sub>2</sub> adsorption capacity (mmol g <sup>-1</sup> )	Temperature (°C)	Pressure (bar)	References
<b>Zeolites</b>				4
Sodium type X zeolite (NaX)	5.39	25	1	5
Zeolite 13X (13x-c)	6.2	298 K	1	6,7
Zeolite 13X (13X-B)	4.8		1	7
Zeolite 4A	6.217	298 K	1	8
Zeolite 5A	3.68	298 K	1	9
Zeolite molecular sieve type 5A (5A)	3.88	30	10	10
	5.02	32	5	11
Lithium type X zeolite (LiX)	5.51	20	9	12
Zeolite molecular sieve type 13X (13X)	4.51	25	29	13
	5.87	32	2	14
	7.21	25	20	15
	0.0052	25	15	16
Zeolite Socony Mobil-5 (ZSM-5)	2.50	30	5.7	17
	0.90	40	1	18
Mobil Composition of Matter No. 41 (MCM-41)	0.53	30	1	19
	0.63	25	1	20
Mobil Composition of Matter No. 48 (MCM-48)	0.32	25	-	21
Santa Barbara Amorphous No. 15 (SBA-15)	0.40	25	1	22
Korea Advanced Institute of Science and Technology no.6 (KIT-6)	0.48	30	1	23
<b>MOFs</b>				24-26
Hong Kong University of Science and Technology-1 (HKUST-1)	7.19	30	10	27
	15.50	22	30	28
	3.60	25	1	29
Materials of Institute Lavoisier-101 (MIL-101)	0.85	25	10	30
	2.90	30	1.13	31
	1.17	30	2.41	27
	8.07	30	10	27
	21.28	22	36	28
	34	30	50	28,32
	40	30	50	
Metal organic framework-5 (MOF-5)	2.10	23	1	33
	15.22	24	14	34
MOF-210	54.5	298 K	50	35
	43.1		1	
MOF-177	33.5	298 K	35	36
	0.8		1	36
MOF-2	3.2		35	36
	0.7		1	
MOF-505	10.2		35	36
	3.3		1	
MOF-74	10.4		35	36
	4.9		1	
Ni-Datz-MOF-74	7.76		1	37
Cu <sub>3</sub> (BTC) <sub>2</sub>	10.7		35	36
	4.1		1	
IRMOF-11	14.7		35	36
	1.8		1	
IRMOF-3	18.7		35	36
	1.2		1	

IRMOF-6	19.5		35	36
	1.1		1	
IRMOF-1	21.7		35	36
	1.1		1	
<b>Targeted modification of the metal sites</b>				
Mg <sub>2</sub> (dobdc)	8-9	25	1	6,38-42
Mg <sub>2</sub> (dobpdc)	6.42	25	1	43
Ni <sub>2</sub> (dobdc)	7	25	1	40,44
Fe <sub>2</sub> (dobdc)	7.1	25	1	40,45
Co <sub>2</sub> (dobdc)	6.9	25	1	39,40,44
Zn <sub>2</sub> (dobdc)	~5.61	25	1	39,40,44
Mn <sub>2</sub> (dobdc)	~5.9	25	1	40
Cu <sub>2</sub> (dobdc)	~2.90	25	1	
Mg <sub>2</sub> (dondc)	6.42	25	1	46
Mg <sub>2</sub> (dotpdc)	~5.30	25	1	47
Mg <sub>2</sub> (pc-dobpdc)	~6.50	25	1	47
Mn <sub>2</sub> (dobpdc)		25	1	48
Co <sub>2</sub> (dobpdc)		25	1	48
Ni <sub>2</sub> (dobpdc)		25	1	48
Zn <sub>2</sub> (dobpdc)		25	1	48
Mg/Zn(dobdc)	5.10	25	1	49
Ni/Zn(dobdc)	4.80	25	1	49
Mg/Zn(dobpdc)	~5.90	25	1	50
SIFSIX-3-Cu	2.58	25	1	38,51
SIFSIX-3-Zn	2.51	25	1	38,52
SIFSIX-3-Ni	2.5	25	1	6
SIFSIX-18-Ni-β		25	1	53
SIFSIX-2-Cu-i	5.41	25	1	38,51,52
HKUST-1	2.5/4.1	25	1	6,36
UTSA-16	4.3	25	1	54
MIL-101(Cr)	1.22	25	1	55
ZU-16-Co	2.87	25	1	56
TIFSIX-3-Ni	2.32	25	1	53,56,57
NbOFFIVE-1-Ni	2.2	25	1	53,58
SIFSIX-14-Cu-i	4.70	25	1	59
dptz-CuTiF6	~4.51	25	1	60
MIL-53(Al)	2.58	25	1	38,51

### Amorphous/activated carbon

25,61

Walnut shell	1.86	25	1	62
Cotton wood	1.61	25	1	63
Rambutan peel	1.72	25	1	64
Chicken manure	10.15	20	1	65
Rice husk char	3.1	25	1	66
Date seeds	2.94	25	1	67
Olive stones	3.1	25	1	68
Coconut shell	3.9	25	1	69
nutshell	3.48	25	1	70
Palm date seeds	4.38	25	1	71
Urea and tobacco stem	29.5	25	30	72
Carrot peels	4.18	25	1	73
Camellia japonica	5	25	1	74
Pristine gelatine and starch	3.84	25	1	75
Paulownia sawdust	5.8	25	1	76
Argan hard shells	5.63	25	1	77
Coffee grounds	6.89	25	1	78
Wooden chopsticks	2.63	25	1	79
Polypodium vulgare	5.67	25	1	80
Lignin waste	2.4	25	1	81
Starch, cellulose and sawdust	4.8	25	1	82
Coconut shell-derived AC	0.18	20	1	83
	2.50	25	1	
	3.60	25	1	
	3.90	25	1	69

Oil palm biomass-derived AC	4.40	25	1	84
	3.71	25	1	85
Olive stone-derived AC	2.02	25	1	86
	3.10	25	1	68
Almond shell-derived AC	2.09	25	1	86
	2.70	25	1	68
Peanut shell-derived AC	4.41	25	1	87
Sawdust-derived AC	4.80	25	1	76
Bamboo-derived AC	4.50	25	1	88
Rice husk-derived AC	3.87	25	1	89
Coffee ground wastes-derived AC	2.67	35	1	90
	2.95	25	1.20	91
	2.90	25	1	92
Corn cob-derived AC	3.56	28	1	93
Wood-derived AC	4.10	30	1	94

## COF

Covalent triazine framework-1 (CTF-1)	2.25	25	1	95
Perfluorinated triazine-based COF (FCFT-1)	3.41	25	1	95
Dual functional (3-aminopropyl) triethoxysilane-based COF (APTES-COF-1)	3.99	30	37	96
				25
TMFPT-COF	0.86	25	1	97
TPE-COF-II	5.27	0	1	98
cCTF-500	1.82	25	1	99
cCTF-400	1.89	25	1	99
[HO]25%-H2P-COF	31	25	1	100
[HO2C]100%-H2PeCOF	76	25	1	100

## Silicates

25

SiO <sub>2</sub> -NS (Polyethyleneimine)	4.01	75	1	101
SBA-15 (Polyethyleneimine)	0.83	25	1	102
Silica monoliths (Tetraethylenepentamine)	2.23	25	1	103
Silica powder (Tetraethylenepentamine)	2.56	25	1	103
KIL-2 (Polyethyleneimine)	2.19	25	1	104
MSU-J (Tetraethylenepentamine)	3.73	25	1	105
Granular silica (Polyethyleneimine)	2.39	25	1	106
Microspherical silica (Polyethyleneimine)	2.27	25	1	106
Silica powder (3-aminopropyltrimethoxysilane)	1.14	25	1	103
SBA-15 N1-(3-trimethoxysilylpropyl) diethylenetriamine	1.88	25	1	107

## Carbon nanostructures

Mesoporous carbon nitride (CN) spheres	2.90	25	-	108
N-doped ordered mesoporous carbons (NMCs)	3.20	25	1	109
Resorcinol-formaldehyde (RF)-based carbon	3.13	25	1	110
Polyamide (PAN)-based ACF	1.92	25	1	111
Pitch-based ACFs	3.50	25	1	112
Graphene-based monolith	2.10	25	1	113
Mesophase pitches-based CMS	4.70	25	1	114
Cylindrical pelletized CMS	2.01	25	0.97	115
	3.43	25	9.99	
Polymeric based CMS	3.29	25	1	116
				25

PEI-graphite oxide APTS-graphite (Polyethyleneimine)	0.75	25	1	117
Graphite nanoplatelets (GNP)/Fe <sub>3</sub> O <sub>4</sub>	8.5	25	1	118
TEPA-graphite oxide	2.08	25	1	119
Graphite waste/Fe <sub>3</sub> O <sub>4</sub>	5.36	25	1	118
KOH activated graphite nanofibres	1.61, 1.35	25	1	120,121
MPD-graphene oxide	0.91	25	1	122
PEHA-graphene oxide	0.61	25	1	122
Monolithic reduced graphene oxide-based aerogels	6.31			123
TEPA-CNT	5.00	60	1	124
PEI-MWCNT	2.11	25	1	125
APS-CNT	2.19	25	1	126

### MXenes

Mo <sub>2</sub> C-Li	<b>3.66</b>		4 Mpa	127
Mo <sub>2</sub> C-Na	<b>3.47</b>			
Mo <sub>2</sub> C-K	<b>3.31</b>			
Mo <sub>2</sub> C-NH <sub>4</sub>	<b>3.34</b>			
Ti <sub>3</sub> C <sub>2</sub>	<b>1.33</b>		4 MPa	128
V <sub>2</sub> C	<b>0.52</b>		4 MPa	128
DMSO-Ti <sub>3</sub> C <sub>2</sub>	<b>5.79</b>		4 MPa	128
DMSO-V <sub>2</sub> C	<b>0.77</b>		4 MPa	128

### Clays

Cs-montmorillonite	1.4	-20	1	129
Cs-montmorillonite	0.9	20	1	129
Bentonite	0.23	10	10	130
Cs-Bentonite	1.25	10	10	
TMA-Bentonite	1.16	10	10	
MMA-Bentonite	0.92	10	10	
Mg, Cs and Ca -bentonite	0.6	20	1	131
Ni-fluorohectorite	<b>6.45</b>	20	55	132
Ni <sub>0.3</sub> -fluorohectorite	<b>2.14</b>	20	50	133

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