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# **Supplementary Information**

# CVD of MoS<sub>2</sub> Single Layer Flakes using Na<sub>2</sub>MoO<sub>4</sub> – Impact of Oxygen and Temperature-Time-Profile

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## 1. Synthesis of MoS<sub>2</sub>



Fig. S1 (a) Schematic drawing of the set-up to synthesize MoS<sub>2</sub> flakes. (b) Picture of the tube furnace.



**Fig. S2** *T-t*-profile (black) and gas flows (blue) during the synthesis of MoS<sub>2</sub> flakes, where light blue represents the pure N<sub>2</sub> and dark blue the O<sub>2</sub>/N<sub>2</sub> gas flow. The holding temperature  $T_{hold}$  indicates the maximum temperature during synthesis at the center of the tube furnace and the holding time  $t_{hold}$  the time the system is at  $T_{hold}$ . The synthesis time  $t_{syn}$ , which marks the time the center of the tube furnace is at a temperature higher than 650 °C, is flanked by red stars.

**Tab. S1** Investigated exposure times of O<sub>2</sub> in the gas flow (1.3%) and corresponding temperatures in the tube furnace when the O<sub>2</sub>/N<sub>2</sub> gas flow was turned off.  $T_{hold}$  was 710 °C and  $t_{hold}$  was set to 6 min for all exposure times of O<sub>2</sub>. The ramp-up time was 10 minutes. The asterisks \* indicate the syntheses, which were conducted a second time without sulfur present.

Exposure time of $O_2$ (min)	Temperature in the tube furnace (°C)
0*	-
2.5*	200
9	650
10*	710
11	710
12	710
13*	710

**Tab. S2** Synthesis time  $t_{syn}$  for different combinations of  $T_{hold}$  and  $t_{hold}$  investigated.  $t_{syn}$  increases with  $T_{hold}$  and  $t_{hold}$  due to longer time required for cooling down to 650 °C.

		Holding temperature T <sub>hold</sub> (°C)					
		710	730	750	770	790	810
	1	-	-	5.7	-	-	-
	2	4.9	5.8	-	-	8.4	9.5
	3	-	-	8.2	-	-	-
	4	7.0	-	-	-	-	11.7
nin	6	9.2	10.1	11.1	12.1	12.9	14.3
ld (r	7	-	-	-	-	-	15.3
$t_{hol}$	8	-	-	-	-	15.3	-
g time	9	-	-	-	-	16.4	-
	10	-	-	15.5	16.5	17.1	-
din	12	-	-	-	18.6	-	-
PH	15	18.4	19.4	21.0	21.9	-	-
	22	-	-	28.0	-	-	-
	30	33.7	35.1	36.3	-	-	-
	45	-	49.1	-	-	-	-
	60	64.5	65.1	-	-	-	-



**Fig. S3** The 14-sample test was performed by running a synthesis at 790 °C for 10 min with 14 growth substrates placed in a row in the reaction tube to show how the temperature gradient in the tube furnace and therefore the *T*-*t*-profiles influences the reaction process and flake quality.

# 2. XPS Analysis

#### 2.1 Samples

Sample	Labelling	Description	Remarks
P1	P-MoS2	MoS <sub>2</sub> powder (98 %, CAS: 1317-33-5, from Sigma-Aldrich, as delivered)	Reference sample for binding energy referencing
P2	P-Na2MoO4	Na <sub>2</sub> MoO <sub>4</sub> powder (98 %, CAS: 7631-95-0, from Sigma-Aldrich, as delivered)	Control sample for curve fitting model
SC	Na2MoO4	Spin-coated Mo-source (aqueous Na_2MoO_4 solution with 0.5 mg/ml)	Reference sample for Mo- source
Tt0	710C0M	T <sub>hold</sub> = 710 °C, <b>t<sub>hold</sub> = 0 min</b> , O <sub>exp</sub> = 2.5 min	
Tt1	710C2M	T <sub>hold</sub> = 710 °C, <b>t<sub>hold</sub> = 2 min</b> , O <sub>exp</sub> = 2.5 min	
Tt2	710C4M	T <sub>hold</sub> = 710 °C, <b>t<sub>hold</sub> = 4 min</b> , O <sub>exp</sub> = 2.5 min	
Tt3	710C6M	T <sub>hold</sub> = 710 °C, <b>t<sub>hold</sub> = 6 min</b> , O <sub>exp</sub> = 2.5 min	
Tt4	710C15M	T <sub>hold</sub> = 710 °C, <b>t<sub>hold</sub> = 15 min</b> , O <sub>exp</sub> = 2.5 min	Reference sample for binding energy referencing and curve fitting
Tt5	710C30M	T <sub>hold</sub> = 710 °C, <b>t<sub>hold</sub> = 30 min</b> , O <sub>exp</sub> = 2.5 min	
Tt6	710C60M	T <sub>hold</sub> = 710 °C, <b>t<sub>hold</sub> = 60 min</b> , O <sub>exp</sub> = 2.5 min	
Tt7	730C15M	<b>T</b> <sub>hold</sub> = <b>730 °C</b> , <b>t</b> <sub>hold</sub> = <b>15 min</b> , <b>O</b> <sub>exp</sub> = 2.5 min	
Tt8	750C15M	<b>T</b> <sub>hold</sub> = <b>750 °C</b> , <i>t</i> <sub>hold</sub> = <b>15 min</b> , <i>O</i> <sub>exp</sub> = 2.5 min	
Tt9	750C30M	<b>T</b> <sub>hold</sub> = <b>750 °C</b> , <b>t</b> <sub>hold</sub> = <b>30 min</b> , <b>O</b> <sub>exp</sub> = 2.5 min	
00	00M	T <sub>hold</sub> = 710 °C, <i>t</i> <sub>hold</sub> = 6 min, <i>O</i> <sub>exp</sub> = 0 min	
01	02.5M	T <sub>hold</sub> = 710 °C, <i>t</i> <sub>hold</sub> = 6 min, <i>O</i> <sub>exp</sub> = 2.5 min	Same sample as Tt3
02	09M	T <sub>hold</sub> = 710 °C, t <sub>hold</sub> = 6 min, <b>Ο</b> <sub>exp</sub> = 9 min	
03	010M	T <sub>hold</sub> = 710 °C, t <sub>hold</sub> = 6 min, <b>Ο</b> <sub>exp</sub> = <b>10 min</b>	
04	011M	T <sub>hold</sub> = 710 °C, <i>t</i> <sub>hold</sub> = 6 min, <i>O</i> <sub>exp</sub> = <b>11 min</b>	
05	012M	T <sub>hold</sub> = 710 °C, <i>t</i> <sub>hold</sub> = 6 min, <b>O</b> <sub>exp</sub> = 12 min	
06	013M	T <sub>hold</sub> = 710 °C, t <sub>hold</sub> = 6 min, <b>O</b> <sub>exp</sub> = 13 min	Reference sample for sulfate and curve fitting

**Tab. S3** Overview of samples investigated with XPS, where  $T_{hold}$  denotes the holding temperature,  $t_{hold}$  the holding time, and  $O_{exp}$  the exposure time of O<sub>2</sub>. The parameters in bold represent the parameters of interest.

SO	S-OOM	$T_{\text{hold}}$ = 710 °C, $t_{\text{hold}}$ = 6 min, $O_{\text{exp}}$ = 0 min No sulfur present	
S1	S-O2.5M	$T_{hold}$ = 710 °C, $t_{hold}$ = 6 min, $O_{exp}$ = 2.5 min No sulfur present	
S2	S-010M	$T_{hold}$ = 710 °C, $t_{hold}$ = 6 min, $O_{exp}$ = 10 min No sulfur present	
S3	S-013M	T <sub>hold</sub> = 710 °C, t <sub>hold</sub> = 6 min, <b>O</b> <sub>exp</sub> = 13 min No sulfur present	

#### 2.2 Survey Spectra



Fig. S4 XP survey spectra of (a) sample P1 (P-MoS2) and P2 (P-Na2MoO4), and (b)  $MoS_2$  flakes synthesized with different temperature-time-profiles as reported in Tab. S3.



Fig. S5 XP survey spectra of (a)  $MoS_2$  flakes synthesized with different exposure times of  $O_2$  and (b) all samples synthesized without sulfur present as reported in Tab. S1.

#### 2.3 Binding Energy Referencing of High-Resolution XP-Spectra

Due to the insulating nature of the growth substrate used (SiO<sub>2</sub>/Si), the following binding energy (BE) referencing method was adopted. First, the XP-spectrum of sample P1 (P-MoS2) was analyzed to obtain a reference spectrum of a conductive sample for the Mo 3d<sub>5/2</sub> peak position of a MoS<sub>2</sub> sample. The peak has its maximum at a binding energy of 229.1 eV and corresponds well to value given in literature.<sup>1,2</sup> Afterwards, the synthesized MoS<sub>2</sub> sample Tt4 (710C15M) was BE-referenced using the Mo 3d peak, set to the value found for P1. The main O 1s of sample Tt4, as for all other synthesized samples, is ascribed to the growth substrate (SiO<sub>2</sub>/Si) and was found at a BE of 532.5 eV, corresponding well to literature values.<sup>3</sup> Finally, this O 1s signal of the growth substrate was used for BE referencing of all other synthesized samples.

#### 2.4 Curve Fitting

#### Background and curve fitting of S 2p

The background of the S 2p signal of MoS<sub>2</sub> has a complex shape as its peaks sit right after the Si 2s signal caused by SiO<sub>2</sub>/Si (growth substrate). The effect of the characteristic inelastic scattering losses from the Si 2s signal impacts heavily the S 2p region due to the large difference in material thickness (bulk versus atomically thin layer). To consider the prominent convex shape of the background a quadratic fitting of the background is used. A conventional doublet is used to model the signal of MoS<sub>2</sub> flakes (Fig. S6a), though the signal may actually include a variety of additional minor constituents, such as elemental sulfur, amorphous sulfides (MoS<sub>x</sub> with 2<x<3), partially oxidized MoS<sub>2</sub> (MoS<sub>x</sub>O<sub>y</sub>), and different MoS<sub>2</sub> phases such as the 2H and 1T phase, which are thus ignored. The signal ascribed to sulfates is fitted with a single conventional doublet too (Fig. S6b), after subtraction of a linear background.



**Fig. S6** High-resolution XP-spectra of S 2p core level peak region from the sample (a) Tt4 (710C15M) and (b) O6 (O13M), which are used as reference samples for the curve fitting for the sulfur S 2p doublet (pink) and sulfate (green).

#### Background and curve fitting of Mo 3d and S 2s

The Mo 3d signals of  $Na_2MoO_4$ ,  $MoS_2$ , and Mo-oxides overlap over a wide BE range, which also encompasses the S 2s signals of  $MoS_2$  and of sulfates, when present.

The following curve-fitting model was applied to represent the signal envelope (Fig. S7).

- a.) Singlet and doublet for S 2s and Mo 3d, respectively, representing MoS<sub>2</sub> (pink and purple).
- b.) Singlet for S 2s representing sulfates (green). As this peak is entirely overlapped by the Mo 3d envelope, it is not possible to accurately see its presence. Therefore, its curve fitting is only added if a clear S 2p sulfate peak was detected in the high-resolution XP-spectra of the S 2p core level peak region.
- c.) Doublet for Mo 3d representing Na<sub>2</sub>MoO<sub>4</sub>/MoO<sub>3</sub>/secondary structure of the MoS<sub>2</sub> signal (grey).
- d.) Doublet for Mo<sub>def</sub> 3d representing defective states such as MoO<sub>3-x</sub>/MoS<sub>x</sub>O<sub>y</sub> (dark green).
- e.) Shirley background line with two steps at the position of the Mo  $3d_{5/2}$  and Mo  $3d_{3/2}$  peaks of the MoS<sub>2</sub> signal.



**Fig. S7** High-resolution XP-spectra of Mo 3d core level peak region of the sample (a) Tt4 (710C15M) and (b) O6 (O13M), which are used as reference samples for the curve fitting for the sulfur S 2s singlet (pink), Mo 3d doublet (purple), Mo doublet (grey),  $Mo_{def}$  doublet (dark green), and sulfate (green).

#### **Overview of fitting parameters**

Signal	Labelling	Source	Background	Position (eV)	Area	FWHM (eV)	Line
							Shape
Mo 3d <sub>5/2</sub>	A <sub>MoS</sub>	MoS <sub>2</sub>	Shirley	228.9 – 229.5	Free	0.9 - 1.1	LA(1.2,
							1.6, 260)
Mo 3d <sub>3/2</sub>	B <sub>MoS</sub>	MoS <sub>2</sub>	Shirley	A <sub>MoS</sub> + 3.15 <sup>a</sup>	A <sub>MoS</sub> * 2/3 <sup>b</sup>	A <sub>MoS</sub> * 1.15 <sup>a</sup>	LA (1.2,
							1.6, 260)
Mo 3d <sub>5/2</sub>	A <sub>Mo6</sub>	Na <sub>2</sub> MoO <sub>4</sub> ,	Shirley	232.3 – 233.1	Free	1.29 – 1.49	LA (1.3,
		MoO <sub>3</sub> ,					1.2, 399)
		Secondary					
		structure of					
		MoS <sub>2</sub> signal					
Mo 3d <sub>3/2</sub>	B <sub>Mo6</sub>	Na <sub>2</sub> MoO <sub>4</sub> ,	Shirley	A <sub>Mo6</sub> + 3.15 <sup>a</sup>	A <sub>Mo6</sub> * 2/3 <sup>b</sup>	А <sub>Моб</sub> * 1 <sup>а</sup>	LA (1.3,
		MoO <sub>3</sub> ,					1.2, 399)
		Secondary					
		structure of					
		MoS <sub>2</sub> signal					
Mo 3d <sub>5/2</sub>	A <sub>Modef</sub>	МоО <sub>з-х</sub> ,	Shirley	$A_{Mo6} - 1.2^d$	Free	A <sub>Mo6</sub> * 1.28 <sup>d</sup>	LA (1.5,
		MoO <sub>x</sub> S <sub>y</sub> <sup>c</sup>					243)
Mo 3d <sub>3/2</sub>	B <sub>Modef</sub>	MoO <sub>3-x</sub> ,	Shirley	A <sub>Modef</sub> + 3.15 <sup>a</sup>	A <sub>Modef</sub> * 2/3 <sup>b</sup>	A <sub>Modef</sub> * 1 <sup>a</sup>	LA (1.5,
		MoO <sub>x</sub> S <sub>y</sub> <sup>c</sup>					243)
S 2s	S <sub>MoS</sub>	MoS <sub>2</sub>	Shirley	A <sub>MoS</sub> – 2.85 <sup>e</sup>	A <sub>MoS</sub> * 0.41 <sup>e</sup>	A <sub>MoS</sub> * 2.11 <sup>e</sup>	LA (1.2,
							1.6, 100)
S 2s <sup>f</sup>	S <sub>Sul</sub>	Sulfate	Shirley	233.5 – 234-3	187 – 227 <sup>g</sup>	2.6 - 3.1	LA (1.5 <i>,</i>
							243)
S 2p <sub>3/2</sub>	SA <sub>MoS</sub>	MoS <sub>2</sub>	Quadratic	161.5 – 162.3	Free	0.79 – 1.19	LA(1.6, 4,
							308)
S 2p <sub>1/2</sub>	SB <sub>MoS</sub>	MoS <sub>2</sub>	Quadratic	$SA_{MoS} + 1.18^{a}$	SA <sub>MoS</sub> * 1 <sup>b</sup>	SA <sub>Mos</sub> * 1 <sup>a</sup>	LA(1.6, 4,
							308)
S 2p <sub>3/2</sub>	SA <sub>Sul</sub>	Sulfate	Linear	167.9 – 168.7	Free	1.2 – 1.6	LA(1.6,2,
							230)
S 2p <sub>1/2</sub>	SB <sub>Sul</sub>	Sulfate	Linear	SA <sub>Sul</sub> + 1.18 <sup>a</sup>	SA <sub>Sul</sub> * 1 <sup>b</sup>	SA <sub>Sul</sub> * 1 <sup>a</sup>	LA(1.5,
							243)

#### Tab. S4 All parameters and constraints used for the XPS curve fittings.

<sup>&</sup>lt;sup>a</sup> These constraints correspond well to values given in literature.<sup>1,2,7</sup>

<sup>&</sup>lt;sup>b</sup> Area ratio based on degeneracy.

<sup>&</sup>lt;sup>c</sup> Includes all defective states.

<sup>&</sup>lt;sup>d</sup> This constraint is derived from the sample O6 (O13M), which shows clearly oxidized MoS<sub>2</sub> flakes.

<sup>&</sup>lt;sup>e</sup> Assumption that MoS<sub>2</sub> is not structurally changing a lot among samples. Therefore, the relative distance on the binding energy scale, the ratio of area, and fwhm of two atoms of the same constituent must be constant. The values of the constraints are derived from the sample Tt4 (710C15M), which consists mainly of MoS<sub>2</sub> flakes.

<sup>&</sup>lt;sup>f</sup> This curve fitting is only added if a clear sulfate peak is visible in the high-resolution XP-spectra of the S 2p core level peak region.

<sup>&</sup>lt;sup>g</sup> As the intensity ratio between 2s and 2p stays the same for each element, the area range of the sulfate peak is derived as +/- 10 % of  $I_{S2s}^{sul}$  with  $I_{S2s}^{sul} = I_{S2s}^{s}/I_{S2p}^{s} * I_{S2p}^{sul}$  with *I* representing the intensity of the S 2s and S 2p peak of sulfur s and sulfate sul, respectively.



**Fig. S8** High-resolution XP-spectra of (a) S 2p and (b) Mo 3d core level peak regions of the sample P1 (P-MoS2).



**Fig. S9** High-resolution XP-spectra of (a) Mo 3d and (b) Na 1s core level peak regions of the sample P2 (P-Na2MoO4).



Fig. S10 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample SC (Na2MoO4).



Fig. S11 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt0 (710COM).



Fig. S12 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt1 (710C2M).



Fig. S13 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt2 (710C4M).



Fig. S14 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt3 (710C6M, and O1 (O2.5M), respectively).



Fig. S15 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt4 (710C15M).



Fig. S17 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt5 (710C30M).



Fig. S16 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt6 (710C60M).



Fig. S18 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt7 (730C15M).



Fig. S19 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt8 (750C15M).



Fig. S21 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample Tt9 (750C30M).



Fig. S20 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample O0 (OOM).



Fig. S22 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample O2 (O9M).



Fig. S23 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample O3 (O10M).



Fig. S24 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample O4 (O11M).



Fig. S25 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample O5 (O12M).



Fig. S26 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample O6 (O13M).



Fig. S28 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample S0 (S-OOM).



Fig. S27 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample S1 (S-O2.5M).



Fig. S29 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample S2 (S-O10M).



Fig. S30 High-resolution XP-spectra of (a) S 2p, (b) Mo 3d, and (c) Na 1s core level peak regions of the sample S3 (S-O13M).

#### 2.6 Deviation of Apparent Na/Mo-Ratio

The apparent Na/Mo-ratio of the sample SC (spin-coated Na<sub>2</sub>MoO<sub>4</sub>) is 4.4 and substantially larger than the expected stoichiometry for Na<sub>2</sub>MoO<sub>4</sub>. As pointed out in *Experimental* in the main manuscript, the quantitative formula used to determine atomic ratios in this work assumes homogeneous composition over the volume probed, which is not the case for our samples. For inhomogeneous compositions, large deviations are expected if the XPS-peaks used for quantification have a substantial difference in binding energy. This applies for the Na/Mo-ratio as the Na 1s and Mo 3d signals differ by about 840 eV in binding energy. An additional contribution to the excess of sodium measured by XPS on spin-coated Na<sub>2</sub>MoO<sub>4</sub> might be related to some degree of ionic exchange between the hydrogen and sodium cations at the Si-OH-rich surface of plasma cleaned SiO<sub>2</sub> substrates. By comparison, when the same approach to determine the Na/Mo-ratio is used to estimate the atomic ratio in a homogeneous powder sample, a value of 2.2 is measured, much closer to the expected stoichiometry.

# 3. Supplementary Figures and Tables



Fig. S31 Light microscopy image of a sample synthesized without  $O_2$  present in the gas flow. The scale bar represents 20  $\mu$ m.



**Fig. S32** Apparent S/Mo-ratio, determined by the formula described in the section *Experimental* in the main manuscript of the samples synthesized at 710 °C for 6 min and different exposure times of the  $O_2$ .



**Fig. S33** High-resolution XP-spectra of O 1s core level peak region of samples synthesized at 710 °C and various  $t_{hold}$ . The O 1s region is dominated by the signal of the growth substrate used (SiO<sub>2</sub>/Si). The inset shows a zoom-in of the shoulder of the O 1s signal to indicate the decrease in the Mo-source (Na<sub>2</sub>MoO<sub>4</sub>), in line with the appearance of the Mo 3d peaks attributed to MoS<sub>2</sub> (see main manuscript).



**Fig. S34** Light microscopy images of  $MoS_2$  flakes synthesized with different holding temperatures  $T_{hold}$  and holding times  $t_{hold}$  as indicated directly in the images. Regardless of the *T*-*t*-profiles all synthesized  $MoS_2$  flakes have a triangular shape, show a homogeneous colour, and are evenly distributed over the entire growth substrate. The scale bar represents 20  $\mu$ m.



**Fig. S35** Light microscopy images of  $MoS_2$  flakes synthesized in the 14-sample test. Regardless of the position of the growth substrate (indicated directly in the images), all synthesized  $MoS_2$  flakes except for the most upstream and most downstream samples (sample *I* and *XIV*) have a triangular shape, show a homogeneous colour, and are evenly distributed over the entire growth substrate. The scale bar represents 20  $\mu$ m.



**Fig. S36** Raman spectrum of the sample synthesized at 790 °C for 8 min as a representative for a typical Raman spectrum of MoS<sub>2</sub> single layer flakes with the relevant peaks at around 384 and 404 cm<sup>-1</sup>, representing the  $E_{2g}^{1}$  and  $A_{1g}$  Raman mode, respectively.

**Tab. S5** The difference between the  $A_{1g}$  and  $E_{2g}^1$  Raman modes of all synthesized MoS<sub>2</sub> flakes except for the most upstream and most downstream samples (sample *I* and *XIV*) from the 14-sample test is around 20 cm<sup>-1</sup>, indicating the presence of monolayers.<sup>4,5</sup> For each sample, 4 to 10 different flakes were measured.

T <sub>hold</sub> (°C)	t <sub>hold</sub> (min)	$A_{1g} - E_{2g}^{1}$ (cm <sup>-1</sup> )
710	2	17.7 (±0.4)
	4	17.3 (±0.5)
	6	17.7 (±0.5)
	15	19.1 (±0.4)
	30	20.2 (±0.1)
	60	20.3 (±0.1)
730	2	18.6 (±0.2)
	6	19.1 (±0.2)
	15	20.4 (±0.1)
	30	20.8 (±0.3)
	45	20.6 (±0.1)
	60	20.9 (±0.8)
750	1	18.9 (±0.2)
	3	18.9 (±0.3)
	6	19.0 (±0.5)
	10	19.9 (±0.3)
	15	20.3 (±0.04)
	22	20.9 (±0.5)
	30	20.6 (±0.2)
770	6	20.3 (±0.3)
	10	20.8 (±0.3)
	12	20.7 (±0.5)
	15	20.5 (±0.3)
790	2	19.3 (±0.1)
	6	20.7 (±0.5)
	8	20.9 (±0.4)
	9	20.5 (±0.2)
	10	19.9 (±0.1)
810	2	19.5 (±0.3)
	4	20.9 (±0.2)
	6	20.4 (±0.4)
	7	20.1 (±1.1)

Number of sample in the 14-sample test	$A_{1g} - E_{2g}^{1}$ (cm <sup>-1</sup> )
	20.3 (±0.3)
	20.2 (±0.6)
IV	20.8 (±0.3)
V	20.2 (±0.4)
VI	20.2 (±0.6)
VII	20.5 (±0.2)
VIII	20.3 (±0.3)
IX	20.3 (±0.3
X	20.8 (±0.4)
XI	20.3 (±0.5)
XII	20.6 (±0.9)
XIII	19.7 (±0.2)



**Fig. S37** (a) AFM height image and (b) statistical height analysis of  $MoS_2$  flakes synthesized at 750 °C for 15 min as a representative for the height measurements based on the description in [6]. The left peak (•) represents the substrate and the right peak (•) the  $MoS_2$  flakes. The scale bar represents 5  $\mu$ m.



**Fig. S38** AFM images of  $MoS_2$  flakes synthesized at 710 °C for 6 min and picked-up by a  $SiO_2/Si$  substrate while floating on the water surface after being detached (transfer by the pick-up method, see *Experimental* in the main manuscript). The scale bar represents (a) 5  $\mu$ m and (b) 500 nm.



Fig. S39 Photoluminescence (PL) spectrum of the sample synthesized at 790 °C for 6 min as a representative for the PL spectrum of a  $MOS_2$  single layer flake with its characteristic peaks at around 1.85 and 2.0 eV, representing the A- and B-exciton, respectively.



**Fig. S40** Light microscopy images of MoS<sub>2</sub> flakes synthesized with different holding temperatures  $T_{hold}$  and holding times  $t_{hold}$  as indicated directly in the images to show the influence of the *T*-*t*-profiles on the detachability of the synthesised MoS<sub>2</sub> flakes. The blue lines mark the immersion depth of the growth substrates into water to detach the MoS<sub>2</sub> flakes. The scale bar represents 100  $\mu$ m.



**Fig. S41** Light microscopy images of  $MoS_2$  flakes synthesized in the 14-sample test to show the influence of the position of the growth substrate in the tube furnace and, therefore, how the *T*-*t*-profile influences the detachability of the synthesised  $MoS_2$  flakes. The blue lines mark the immersion depth of the growth substrates in water to detach the  $MoS_2$  flakes. Samples *V* and *VII* were completely immersed in water. The scale bar represents 100  $\mu$ m.



Fig. S42 MoS<sub>2</sub> flakes picked-up after detaching with a (a) SiO<sub>2</sub>/Si and (b) gold substrate. The scale bar represents 200  $\mu$ m.



![](_page_23_Figure_1.jpeg)

**Fig. S43** Light microscopy images of MoS<sub>2</sub> flakes stored (a) at ambient conditions and (b) in vacuum (300 mbar) for different storage times as indicated directly in the images to test the long-term stability of the MoS<sub>2</sub> flakes detachability. The blue lines mark the immersion depth of the growth substrate in water. Samples without a blue line were immersed completely in water. The scale bar represents 20  $\mu$ m.

![](_page_24_Figure_0.jpeg)

**Fig. S43-Continuation** Light microscopy images of MoS<sub>2</sub> flakes stored in (c) a desiccator for different storage times as indicated directly in the images to test the long-term stability of the MoS<sub>2</sub> flakes detachability. The blue lines mark the immersion depth of the growth substrate in water. The scale bar represents 20  $\mu$ m.

![](_page_24_Figure_2.jpeg)

Fig. S44 AFM measurements of a sample synthesized at 770 °C for 12 min and after different storage time. The scale bar represents 500 nm.

![](_page_25_Figure_0.jpeg)

**Fig. S45**  $I_{\rm B}/I_{\rm A}$ -ratio versus resting time of samples synthesized with different *T*-*t*-profiles, where different colors indicate the corresponding  $T_{\rm hold}$ . The samples in (a) are 100 % detachable (yellow circle), (b) are partial detachable (orange triangle), and (c) are not detachable at all (red cross).

![](_page_25_Figure_2.jpeg)

**Fig. S46** Elemental mapping of S-and Mo<sup>+</sup>-ions of samples synthesized with different *T-t*-profiles measured with ToF-SIMS operated in negative (top row) and positive (bottom row) mode.

![](_page_26_Figure_0.jpeg)

**Fig. S47** ToF-SIMS spectra of samples synthesized with different *T*-*t*-profiles, with ToF-SIMS operated in (a) negative and (b) positive mode.

![](_page_26_Figure_2.jpeg)

**Fig. S48** Elemental mapping of Na<sup>+</sup>-ion of samples synthesized with different *T*-*t*-profiles measured with ToF-SIMS operated in positive mode.

![](_page_26_Figure_4.jpeg)

**Fig. S49** ToF-SIMS spectra operated in the positive mode indicating decreased counts for Na<sup>+</sup> for samples with increasing  $T_{hold}$  and  $t_{hold}$ .

![](_page_27_Figure_0.jpeg)

**Fig. S50** High-resolution XP-spectra of (a) Mo 3d and (b) Na 1s core level peak regions of samples treated at 710 °C for 6 min in absence of sulfur and different exposure times of  $O_2$ . As a reference, the sample synthesized at 710 °C for 6 min with sulfur present is shown in purple.

![](_page_27_Figure_2.jpeg)

**Fig. S51** (a) Photoluminescence (PL) spectra of a  $MoS_2$  flake after transfer onto a fresh  $SiO_2/Si$  substrate to eliminate any influence of the growth substrate on the PL measurement. The spectrum changes depending on the laser irradiation time (561 nm, 7.6 mW) prior to PL measurement due to removal of adsorbed water and PMMA residues of the transfer.<sup>8</sup> (b) PL spectra of a  $MoS_2$  flake on the growth substrate. The spectra only change in intensity with increasing laser irradiation, while the intensity ratio of the B-/A-exciton remains constant at 0.06, corroborating that laser irradiation for cleaning of the transferred flake has no detrimental effects.

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