Electronic Supplementary Information (ESI)

A new metalorganic chemical vapor deposition process for MoS_2 with a 1,4-diazabutadienyl stabilized molybdenum precursor and elemental sulfur

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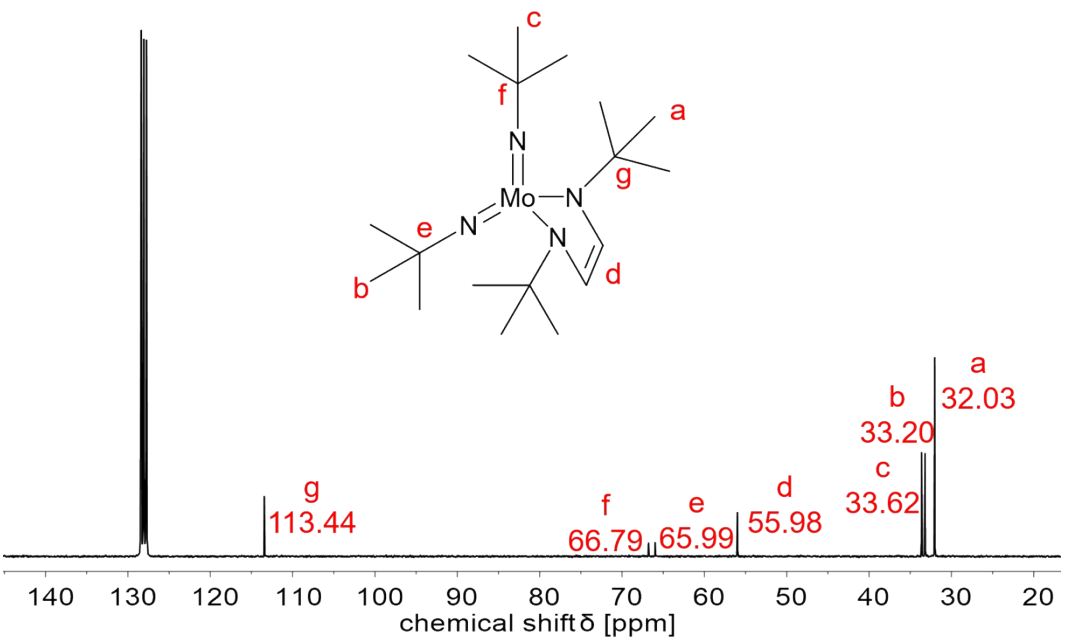

Figure S1. ^{13}C NMR spectrum and "chair" structure of [Mo(N^{t}Bu)$_2$(^{t}Bu$_2$DAD)].

Table S1. Crystal data and structure refinement for [Mo(N^{t}Bu)$_2$(^{t}Bu$_2$DAD)].

Empirical formula	**$C_{18}H_{38}MoN_4$**
Molecular weight (g/mol)	406.46
Temperature (K)	100.00(10)
Crystal system	monoclinic
Space group	$P2_1/c$
a (Å)	10.3048(2)
b (Å)	10.5005(3)
c (Å)	20.7712(5)
α (°)	90
β (°)	93.033 (2)
γ (°)	90
Volume (Å^3)	2244.41(9)
Z	4
Calc. density (g cm^{-3})	1.203
μ (1 mm^{-1})	4.800
F(000)	864.0
Crystal size (mm^3)	0.275 x 0.15 x 0.061
Radiation	CuKα (λ = 1.54184)
2 θ range for data collection (°)	8.526 to 155.522
Index ranges	-13 ≤ h ≤ 13, -12 ≤ k ≤ 13, -26 ≤ l ≤ 21
Reflections collected	23518
Independent reflections	4678 [R_{int} = 0.0238, R_{sigma} = 0.0154]
Data/restrains/parameters	4678/3/259
Goodness-of-fit on F^2	1.092
Final R indexes (I ≥ 2σ (I))	R_1 = 0.0309, wR_2 = 0.0841
Final R indexes (all data)	R_1 = 0.0322, wR_2 = 0.0850
Largest diff. peak (hole/e Å^{-3})	0.43/-0.70

Table S2. Bond lengths for [Mo(N^{t}Bu)$_2$(^{t}Bu$_2$DAD)].

Atom	Atom	Length (Å)	Atom	Atom	Length (Å)
Mo1	C1	2.492(3)	C3	C18	1.481(7)
Mo1	N1	2.016(2)	C3	C17A	1.531(12)
Mo1	N2	2.019(2)	C3	C19A	1.471(11)
Mo1	C2	2.497(3)	C3	C18A	1.556(10)
Mo1	N3	1.744(2)	N4	C4	1.457(3)
Mo1	N4	1.758(2)	C4	C7	1.512(4)
C1	N1	1.374(3)	C4	C6	1.512(4)
C1	C2	1.393(4)	C4	C5	1.504(4)
N1	C14	1.464(3)	C9	C15	1.523(4)
N2	C2	1.364(3)	C8	C15	1.528(4)
N2	C15	1.477(3)	C11	C14	1.536(4)
N3	C3	1.449(3)	C12	C14	1.533(4)
C3	C19	1.489(6)	C13	C14	1.521(4)
C3	C17	1.551(7)	C15	C16	1.522(4)

Table S3. Bond angles for [Mo(N^tBu)$_2$($^tBu_2$DAD)].

Atom	Atom	Atom	Angle(˚)	Atom	Atom	Atom	Angle(˚)
C1	Mo1	C2	32.42(9)	N3	C3	C18	110.9(4)
N1	Mo1	C1	33.41(9)	N3	C3	C17A	108.3(5)
N1	Mo1	N2	85.75(8)	N3	C3	C19A	112.5(8)
N1	Mo1	C2	62.99(9)	N3	C3	C18A	108.1(5)
N2	Mo1	C1	62.67(8)	C19	C3	C17	108.9(5)
N2	Mo1	C2	33.05(8)	C18	C3	C19	114.6(6)
N3	Mo1	C1	143.13(9)	C18	C3	C17	107.6(6)
N3	Mo1	N1	113.77(9)	C17A	C3	C18A	102.7(10)
N3	Mo1	N2	112.97(9)	C19A	C3	C17A	114.8(13)
N3	Mo1	C2	142.41(9)	C19A	C3	C18A	109.8(9)
N3	Mo1	N4	113.85(10)	C4	N4	Mo1	161.1(2)
N4	Mo1	C1	99.39(10)	N4	C4	C7	109.8(2)
N4	Mo1	N1	112.96(9)	N4	C4	C6	108.4(2)
N4	Mo1	N2	114.60(9)	N4	C4	C5	109.8(2)
N4	Mo1	C2	100.08(10)	C7	C4	C6	109.8(3)
N1	C1	Mo1	53.89(12)	C5	C4	C7	108.6(3)
N1	C1	C2	119.8(2)	C5	C4	C6	110.5(3)
C2	C1	Mo1	73.98(15)	N1	C14	C11	109.4(2)
C1	N1	Mo1	92.70(15)	N1	C14	C12	110.8(2)
C1	N1	C14	120.6(2)	N1	C14	C13	107.3(2)
C14	N1	Mo1	136.99(16)	C12	C14	C11	110.1(2)
C2	N2	Mo1	93.10(15)	C13	C14	C11	109.4(2)
C2	N2	C15	122.0(2)	C13	C14	C12	109.9(2)
C15	N2	Mo1	136.68(16)	N2	C15	C9	110.6(2)
C1	C2	Mo1	73.60(15)	N2	C15	C8	106.3(2)
N2	C2	Mo1	53.85(12)	N2	C15	C16	109.4(2)
N2	C2	C1	119.5(2)	C9	C15	C8	109.2(3)
C3	N3	Mo1	166.18(19)	C16	C15	C9	111.3(3)
N3	C3	C19	107.6(4)	C16	C15	C8	110.0(3)
N3	C3	C17	107.0(3)				

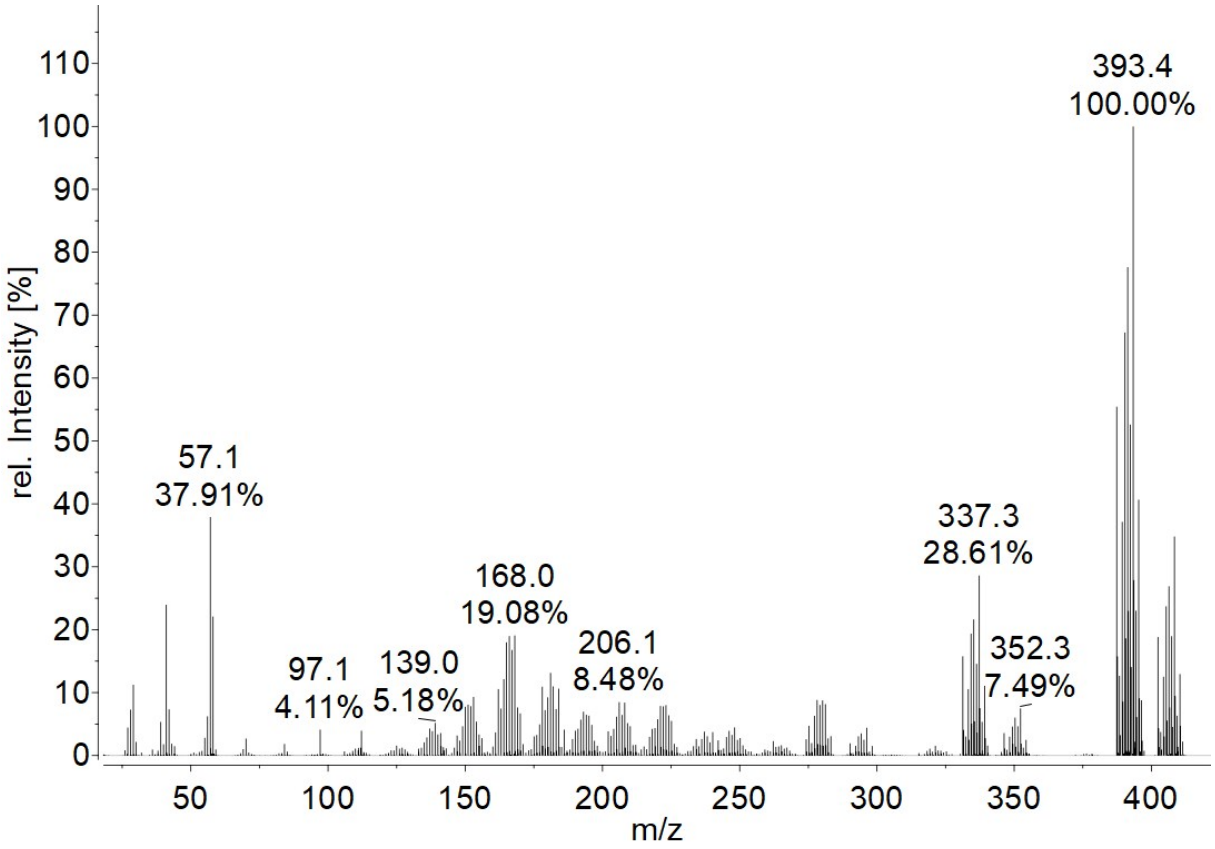

Figure S2. EI-MS spectrum (70 eV) of [Mo(N^tBu)$_2$(^{t}Bu$_2$DAD)].

Table S4. Composition of MoS_2 films estimated from RBS/NRA measurements. Depositions were done at 800 °C with varying deposition parameters. The process parameters in green were used for the main depositions. Variation of certain parameters are marked in blue.

T_{evap} S_8 (°C)	T_{evap} Prec. (°C)	Flow rate (sccm)	Mo (at.%)[a]	S (at.%)[a]	O (at.%)[a]	N (at.%)[a]	C (at.%)[a]	Mo/S Ratio [b]
130	50	25	31.1	63.1	1.4	3.8	0.6	0.49
120	50	25	31.2	62.5	2.6	2.2	1.6	0.50
110	50	25	31.8	63.0	0.8	1.9	2.5	0.50
130	40	25	23.9	51.2	5.7	19.3	0.0	0.47
130	60	25	32.3	59.8	0.6	7.0	0.2	0.54
130	70	25	31.3	61.8	0.5	5.3	1.1	0.51
130	50	50	30.2	56.2	1.7	11.8	0.0	0.54

[a]for all concentration values an error of ± 2 at.% can be considered

[b]the Mo/S ratio has an overall error of 3 %

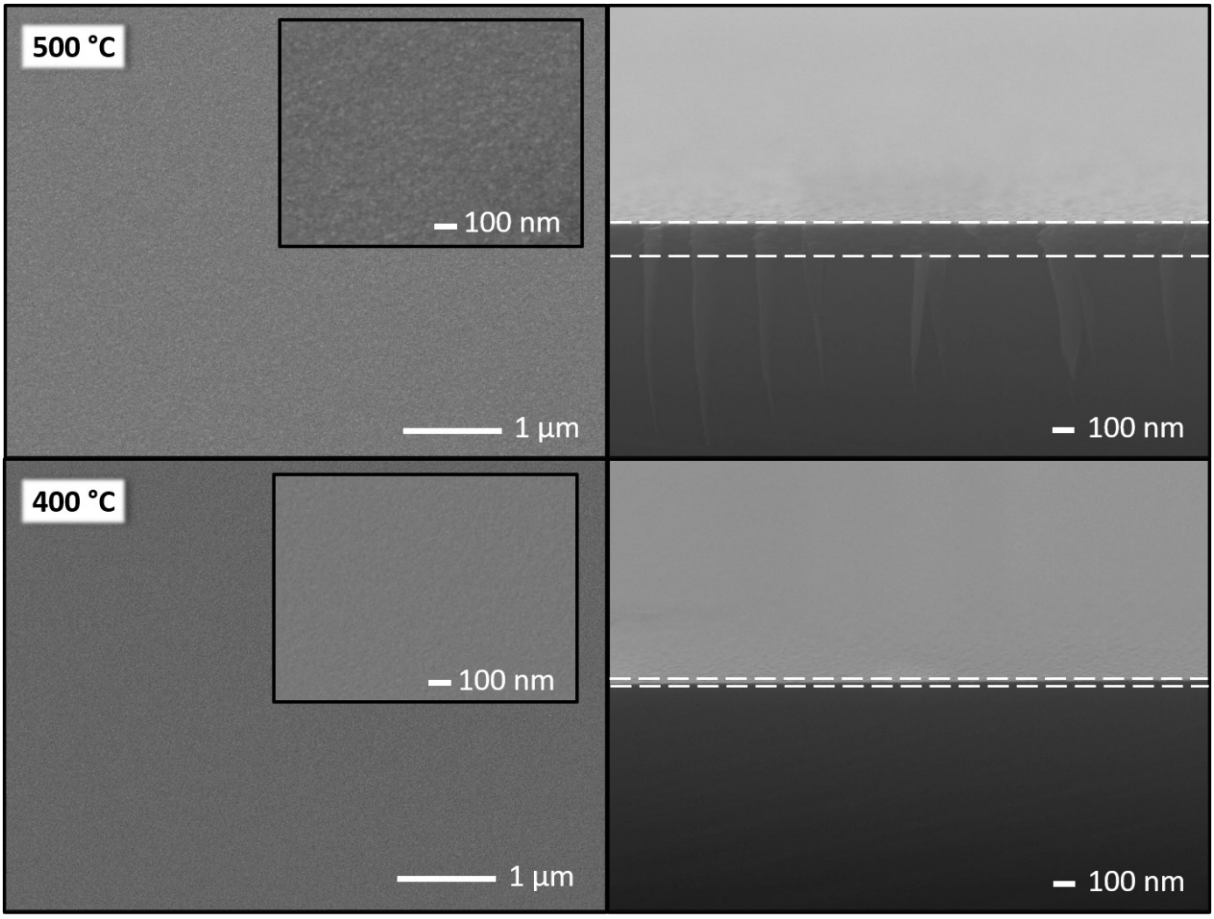

Figure S3. SEM images of MoS_2 films deposited on Si(100) at T_{dep} = 500 °C (top), T_{dep} = 400 °C (bottom) with deposition times of 10 min, respectively. Cross-section images are shown on the right side and top-view images with two different magnifications on the left side. White dashed lines in the cross-section images demarcate the deposited layers from the underlying substrate.

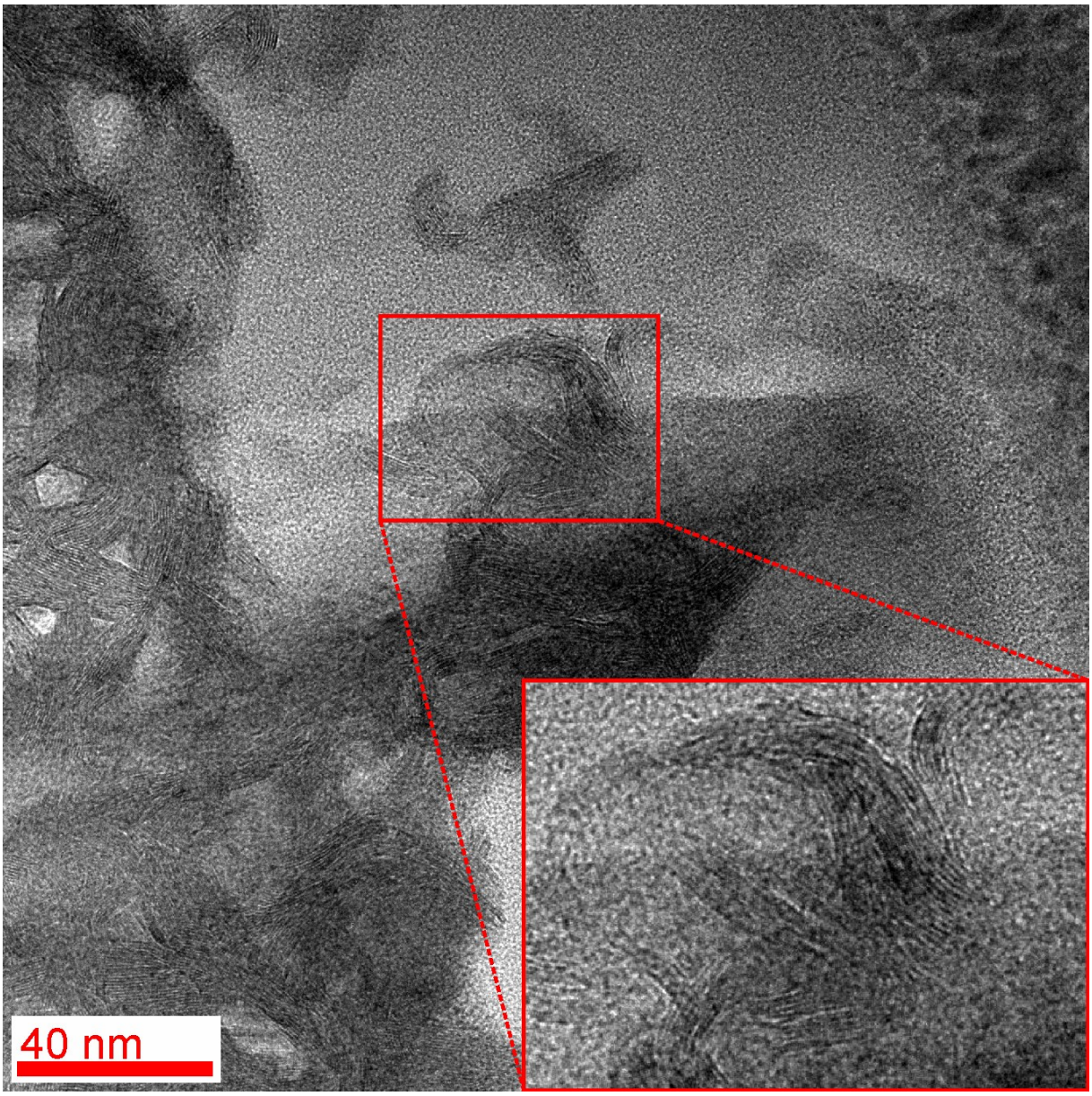

Figure S4. HR-TEM micrograph of a MoS_2 film deposited on Si(100) at T_{dep} = 800 °C. The image shows the cross-section of the top of the surface with a vertically grown flake with a magnified view in the red rectangle. The typical layered structure is visible.

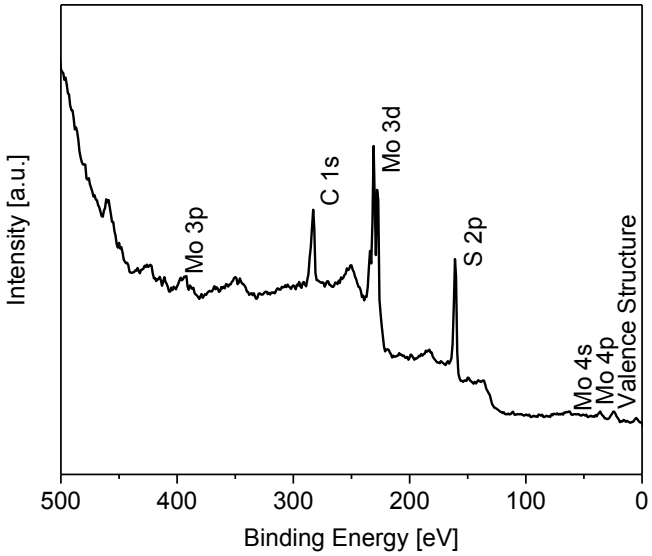

Figure S5. XPS survey spectrum of a MoS_2 film deposited at 600 °C on Si(100) with a thickness of 200 nm.

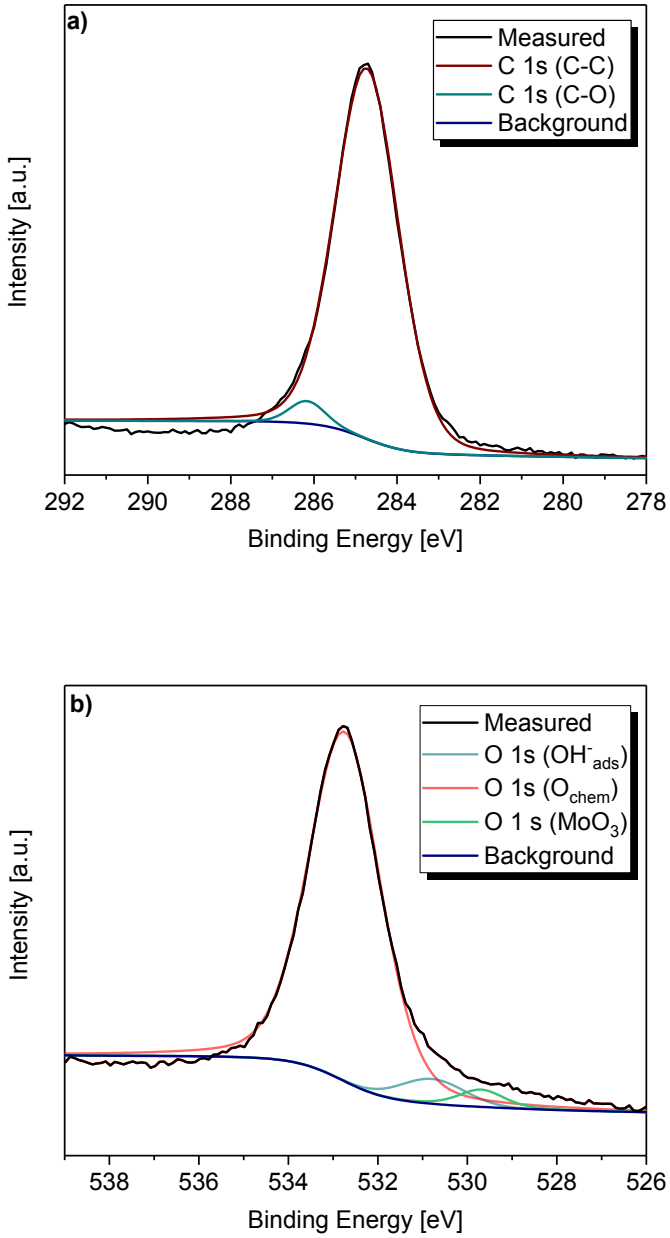

Figure S6. High resolution XPS spectra of a MoS_2 film deposited at 600 °C on Si(100) with a thickness of 200 nm. Core level spectrum of (a) C 1s peaks excited with a photon energy of 380 eV and of (b) O 1s peak excited with a photon energy of 610 eV.