Supporting Information

Single Mo atoms paired with neighboring Ti atoms catalytically decompose ammonium bisulfate formed in low-temperature SCR

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Figures and Tables

Table R1 Relevant parameters from TPDC experiments

Samples	S / ppm	A / mL	yield / %
Mo_1/TiO_2	1045	0.52	67
VWTi	936	0.47	60
MoO_3	592	0.30	38
TiO_2	390	0.20	25

The corresponding total concentration of SO_2 (denoted as S) was obtained from TPDC experiments. Multiplying S by the flow rate (500 mL min^{-1}) can get the total release amount of SO_2 (denoted as A). Meanwhile, based on the loading of ABS (2%) and the mass of catalysts (0.2 g), the maximum release amount of SO_2 was calculated to be 0.78 mL, assuming that HSO_4^- is completely decomposed into SO_2. The corresponding yield of the produced SO_2 can be obtained by dividing A by 0.78 mL. The yields are 67%, 60%, 38% and 25% for Mo_1/TiO_2, VWTi, MoO_3 and TiO_2, respectively.

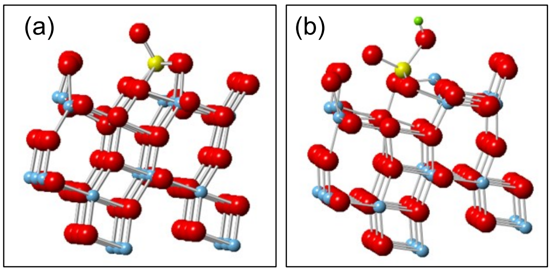

Fig. S1 Side view of structure model of Mo adatom on TiO_2 (101) plane from DFT calculation (a) together with that after removing an oxygen atom in MoO_5 motif (b). Light blue represents Ti atoms, red O atoms, green H atoms, and yellow Mo atoms.

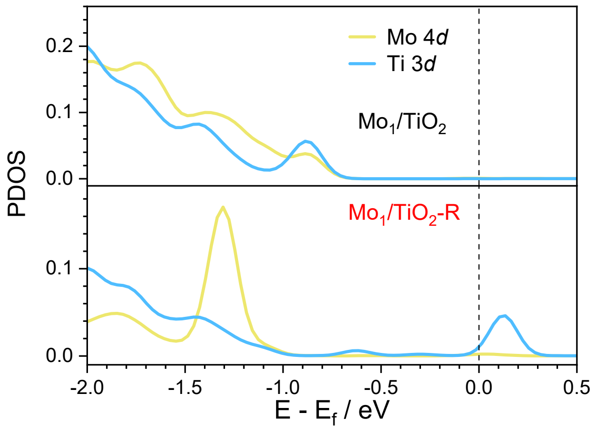

Fig. S2 PDOS of Mo site (4*d*) and Ti site (3*d*) in Mo_1/TiO_2 (up) and Mo_1/TiO_2-R (down).

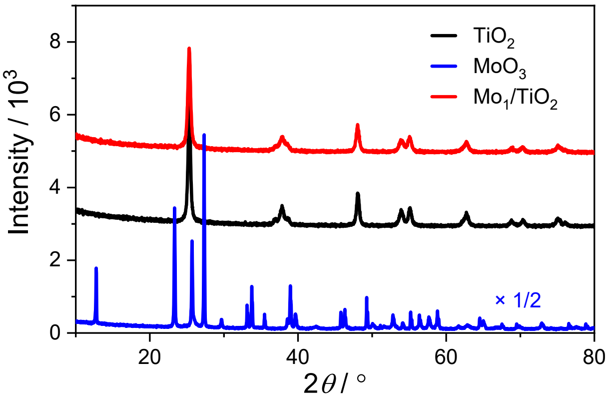

Fig. S3 XRD patterns of Mo_1/TiO_2, MoO_3 and TiO_2 catalysts. No peaks corresponding to MoO_3 appear on the XRD pattern of Mo_1/TiO_2, indicating a highly-dispersed state of Mo species on TiO_2.

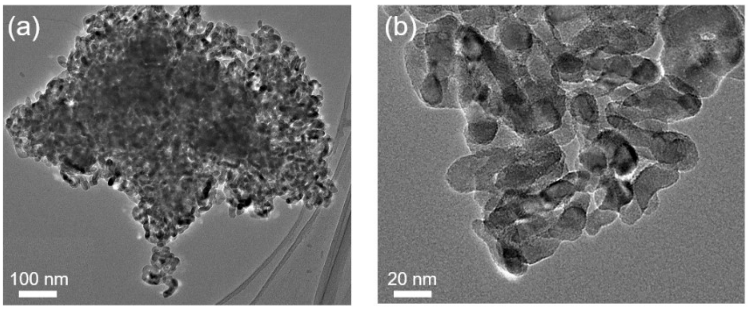

Fig. S4 (a,b) TEM images of Mo_1/TiO_2. No MoO_3 particles can been observed in the images of Mo_1/TiO_2.

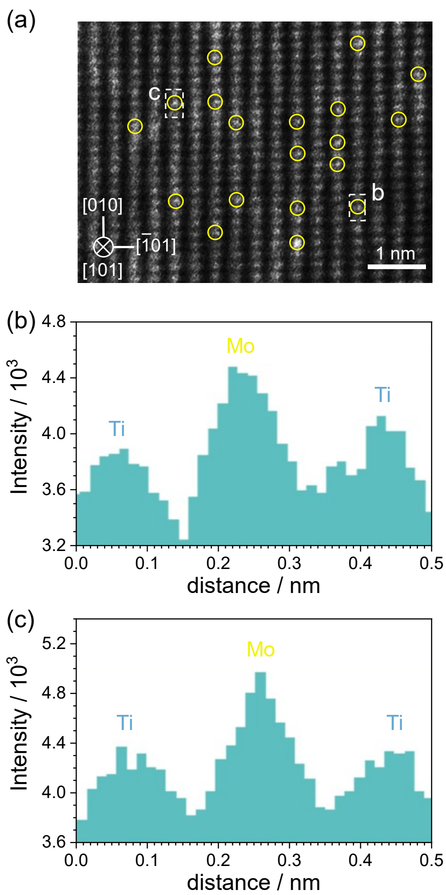

Fig. S5 (a) Atomic-resolution AC-STEM image of Mo_1/TiO_2. (b) and (c) Normalized image intensities in sites b and c shown in (a), respectively.

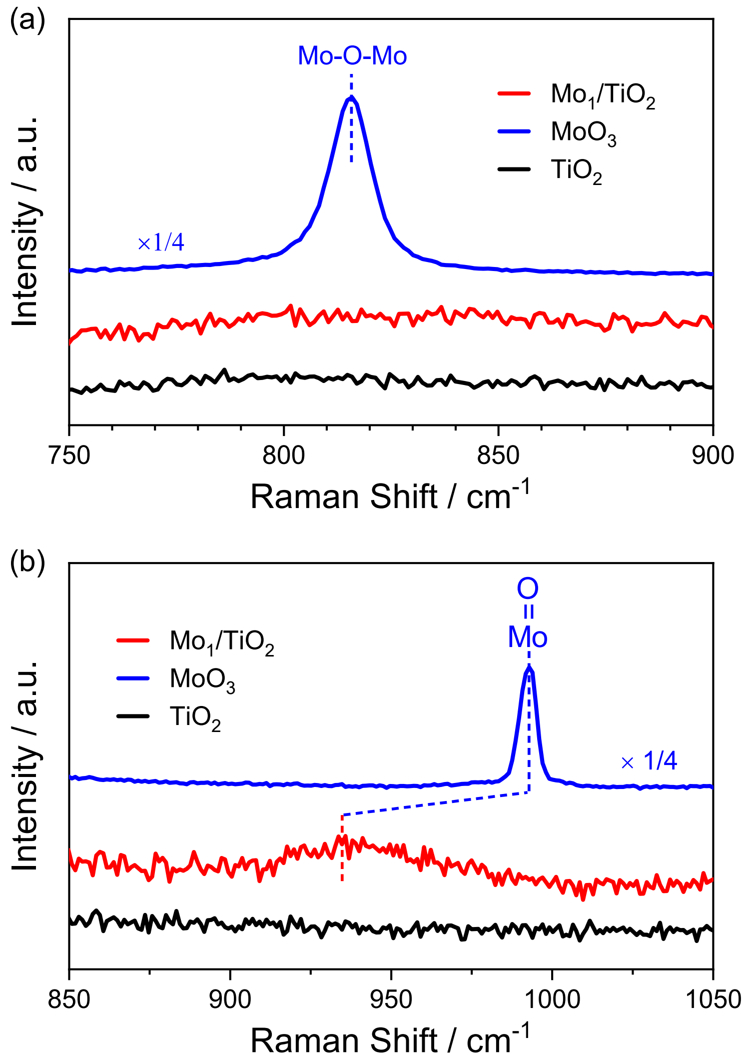

Fig. S6 (a, b) Raman spectra of Mo_1/TiO_2, MoO_3, and TiO_2 catalysts.

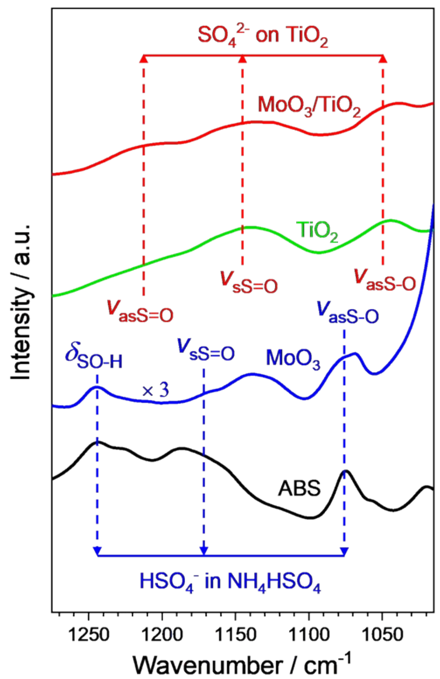

Fig. S7 FT-IR spectra of ABS-deposited catalysts together with pure ABS. The characteristic peaks on the spectra of ABS-MoO_3 and pure ABS are nearly the same.

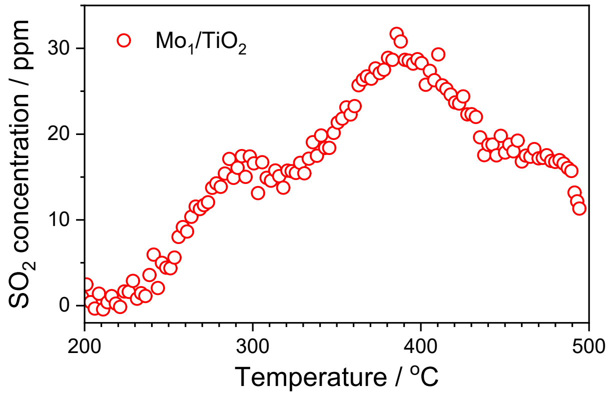

Fig. S8 SO_2 signal of pre-reduced Mo_1/TiO_2 with 2% ABS deposited during TPDC experiment.

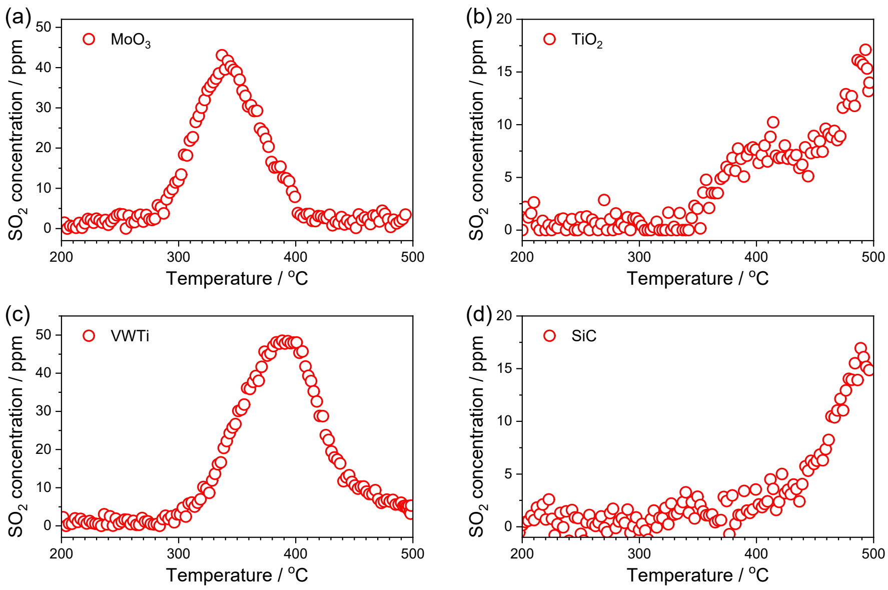

Fig. S9 SO_2 signal of (a) pre-reduced MoO_3, (b) pre-reduced TiO_2, (c) V_2O_5-WO_3/TiO_2 (VWTi) and (d) SiC with 2% ABS deposited as referenced samples during TPDC experiment. SiC processes nearly no redox property, so it was chosen to depict the pristine SO_2 release temperature of ABS.

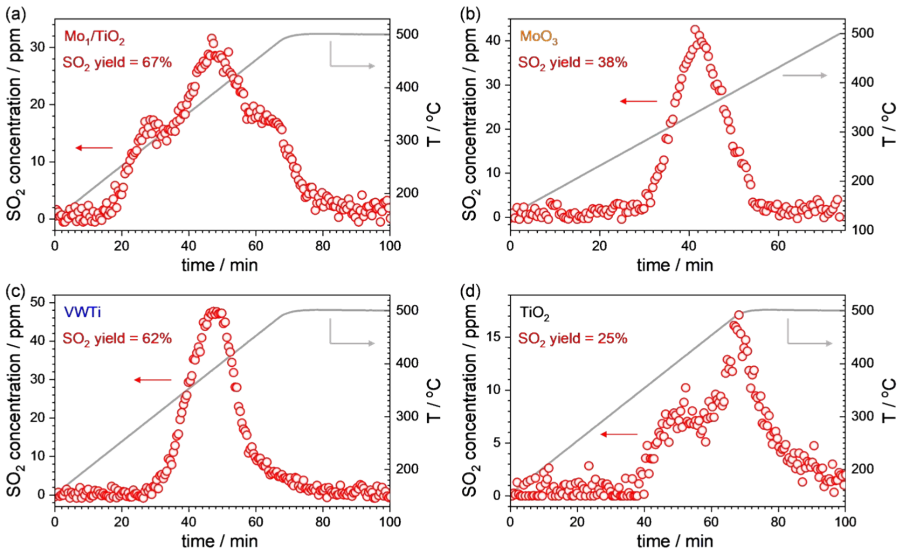

Fig. S10 SO_2 signal as a function of time over ABS-deposited Mo_1/TiO_2 (a), MoO_3 (b), VWTi (c) and TiO_2 (d) during TPDC experiments.

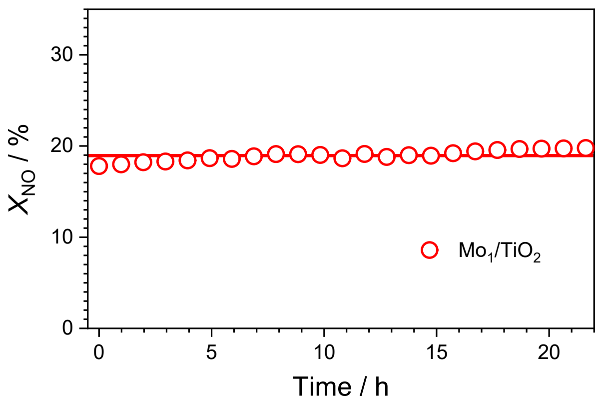

Fig. S11 NO conversion as a function of time over Mo_1/TiO_2 catalyst. Conditions: 0.50 g catalyst, T = 260 °C, 500 ppm NH_3 = NO, 500 ppm SO_2, 5 vol.% H_2O, 3 vol.% O_2, N_2 balanced. The catalyst was per-treated for 10 hours to exclude the effect of surface acidification on the activity.

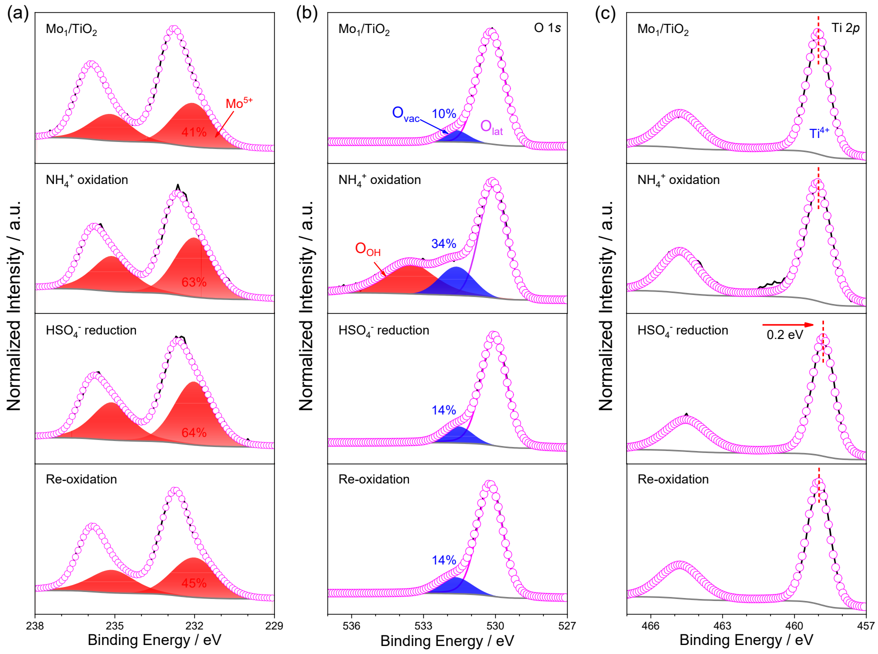

Fig. S12 (a) Mo 3*d*, (b) O 1*s* and (C) Ti 2*p* XPS of Mo_1/TiO_2 catalyst during different stages. On the process of NH_4^+ oxidation, Mo gains electrons and oxygen vacancies increase along with the formation of high-energy H_2O molecular. On the process of HSO_4^- reduction, electron was transferred from O to Ti and S, resulting in decrease of O vacancies and the release of SO_2. After re-oxidation process, electronic states of all elements go back to the pristine levels.

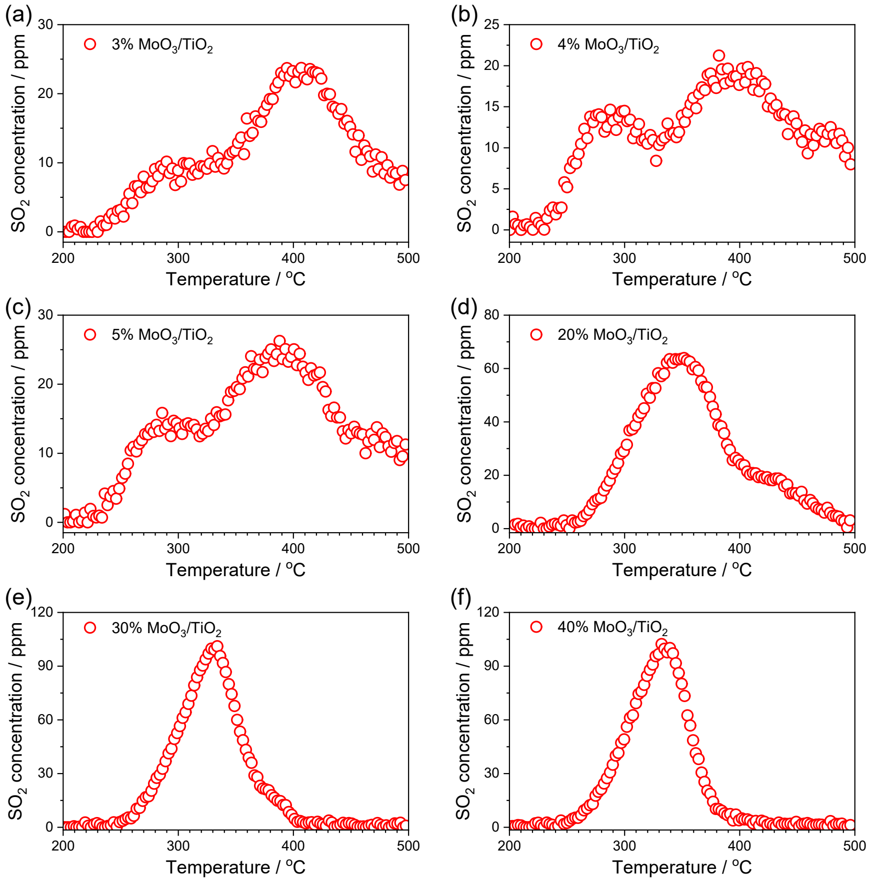

Fig. S13 SO_2 signal of a series of ABS-deposited pre-reduced MoO_3/TiO_2 catalysts with different MoO_3 loading during TPDC experiment.

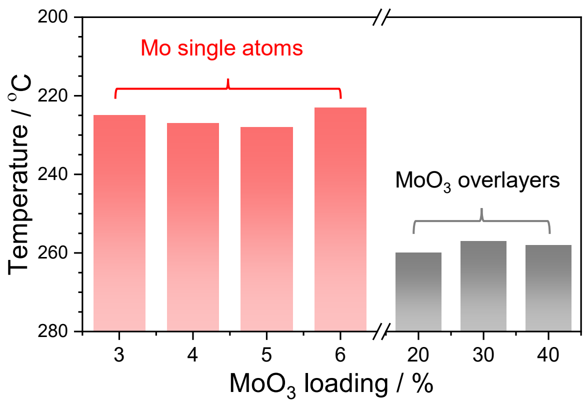

Fig. S14 Onset temperature of the SO_2 release curves over ABS-deposited pre-reduced MoO_3/TiO_2 catalysts as a function of MoO_3 loading. Mo_1/TiO_2 corresponds to sample with 6 wt.% MoO_3 loading. At low loadings (Mo-Ti dual sites on surface), the SO_2 release temperatures are almost the same, which are located at ~ 225 °C, while at high loading (only MoO_3 on surface), the temperature all delay to ~260 °C.

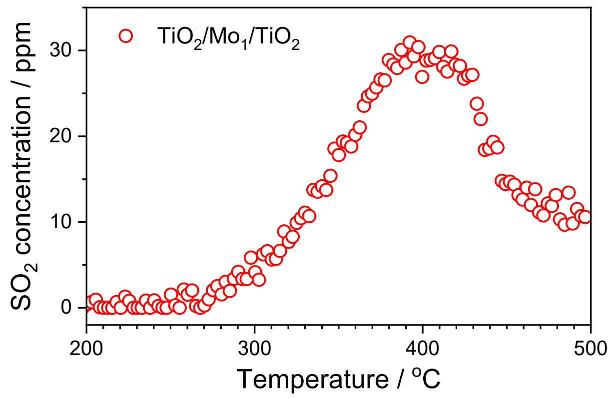

Fig. S15 SO_2 signal of pre-reduced $TiO_2/Mo_1/TiO_2$ with 2% ABS deposited during TPDC experiment. After the coverage of TiO_2 on Mo single atoms, the onset of SO_2 release delays a lot.

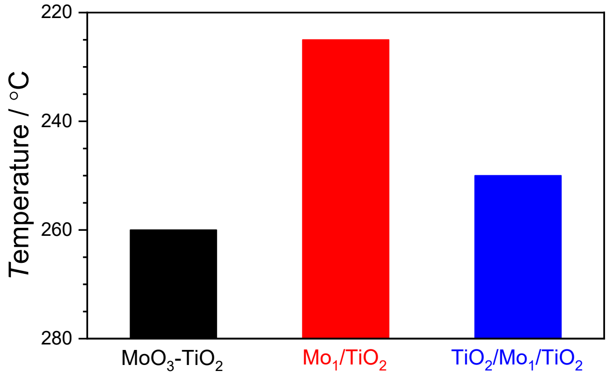

Fig. S16 Onset temperature of the SO_2 release curves over ABS-deposited pre-reduced MoO_3/TiO_2 (40 wt.% MoO_3 loading), Mo_1/TiO_2, and $TiO_2/Mo_1/TiO_2$ catalysts. When MoO_3 or TiO_2 mainly exposed on the surface, the onset of the SO_2 release delays a lot compared with that of MoO_3/TiO_2.