Supporting Information

NO disproportionation over defective 1T′-MoS_2 monolayers

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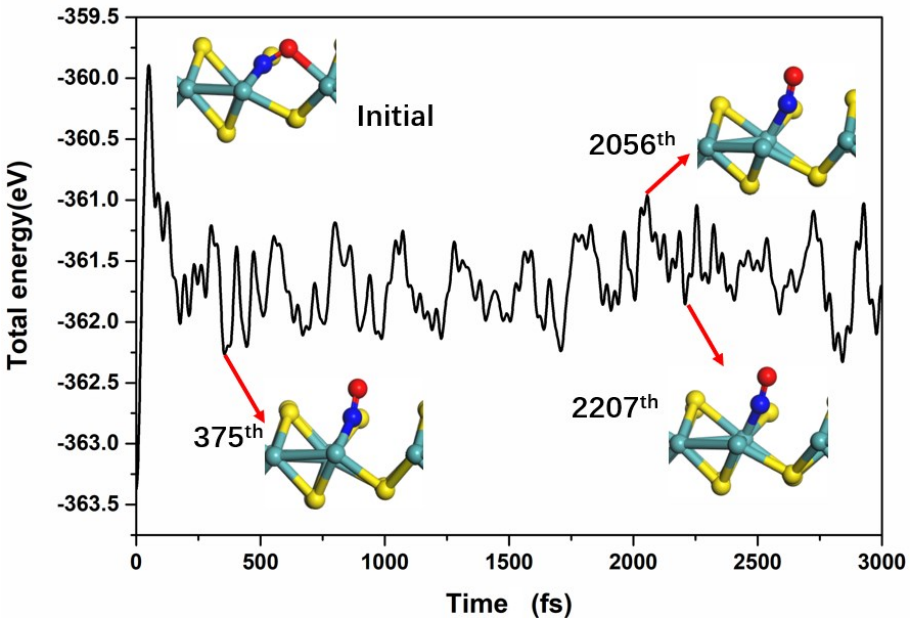

Figure S1. AIMD simulations of the defective 1T′-MoS_2 monolayer adsorbed with an side-on NO molecule. Insets: snapshots of the side views at the initial and 375th, 2056th and 2207th steps of the simulation at 300 K.

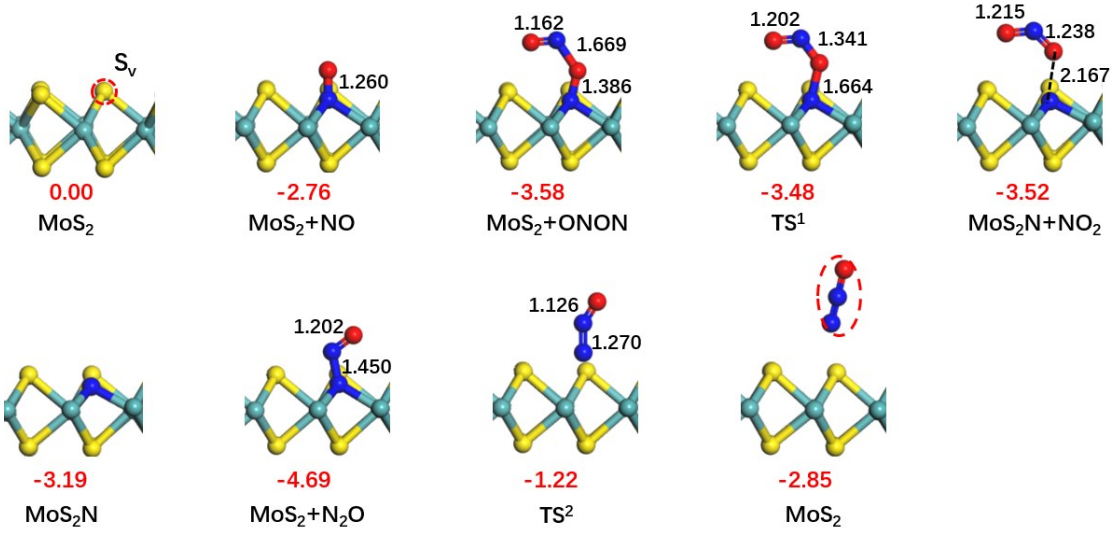

Figure S2. Key structures in the catalytic cycle of NO disproportionation on 2H-MoS_2 monolayers with S vacancies. The key bond length (Å) and relative energy (eV) are

labeled. Transition states are denoted as TS.

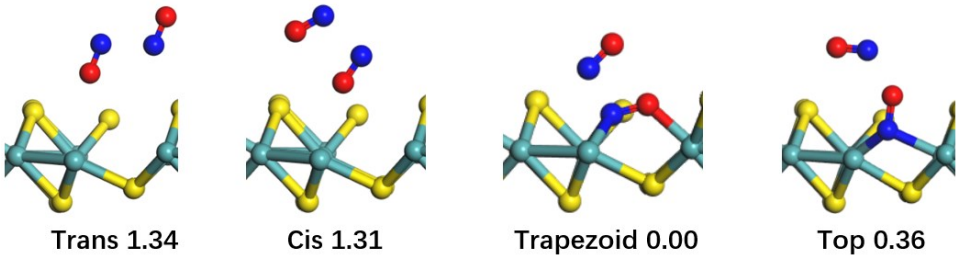

Figure S3. Possible adsorption configurations of two NO molecules adsorbed on defective 1T′-MoS_2 monolayer: trans-, cis- and trapezoid-2NO. The numbers labelled in figure represent the relative energies (eV) of the different configurations. The trans- and cis-$(NO)_2$ configurations are too high in energy to occur. Although the trapezoid-$(NO)_2$ configuration is energetically more stable, it is more difficult for N^2 to grab the O^1 atom to form NO_2. In addition, the readily transformation from the side-on configuration of a single adsorbed NO to the N end-on configuration makes the existence of trapezoid-2NO unlikely.

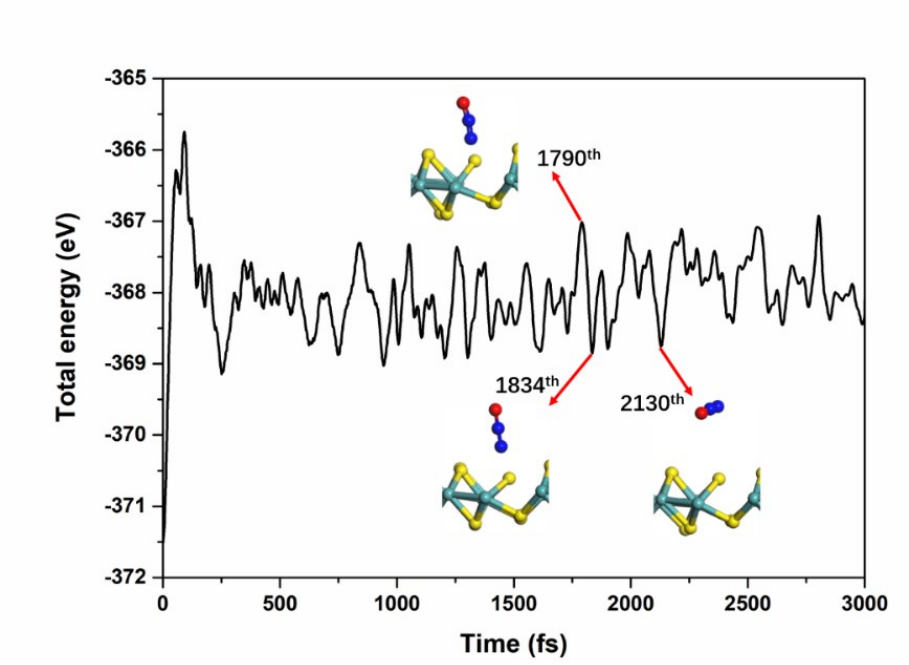

Figure S4. AIMD simulations of the adsorption of N_2O on defective 1T′-MoS_2 monolayer. Insets: snapshots of the side views at the 1790^{th}, 1834^{th} and 2130^{th} steps of the simulation at 500 K.

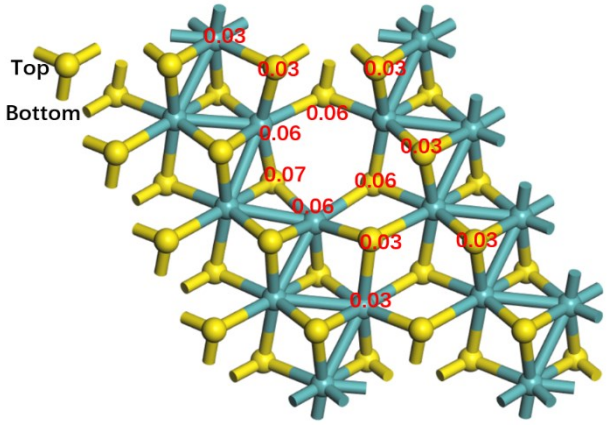

Figure S5. The electron change of the defective 1T′-MoS_2 monolayer. The red numbers represent the negative charge gained by the atoms.

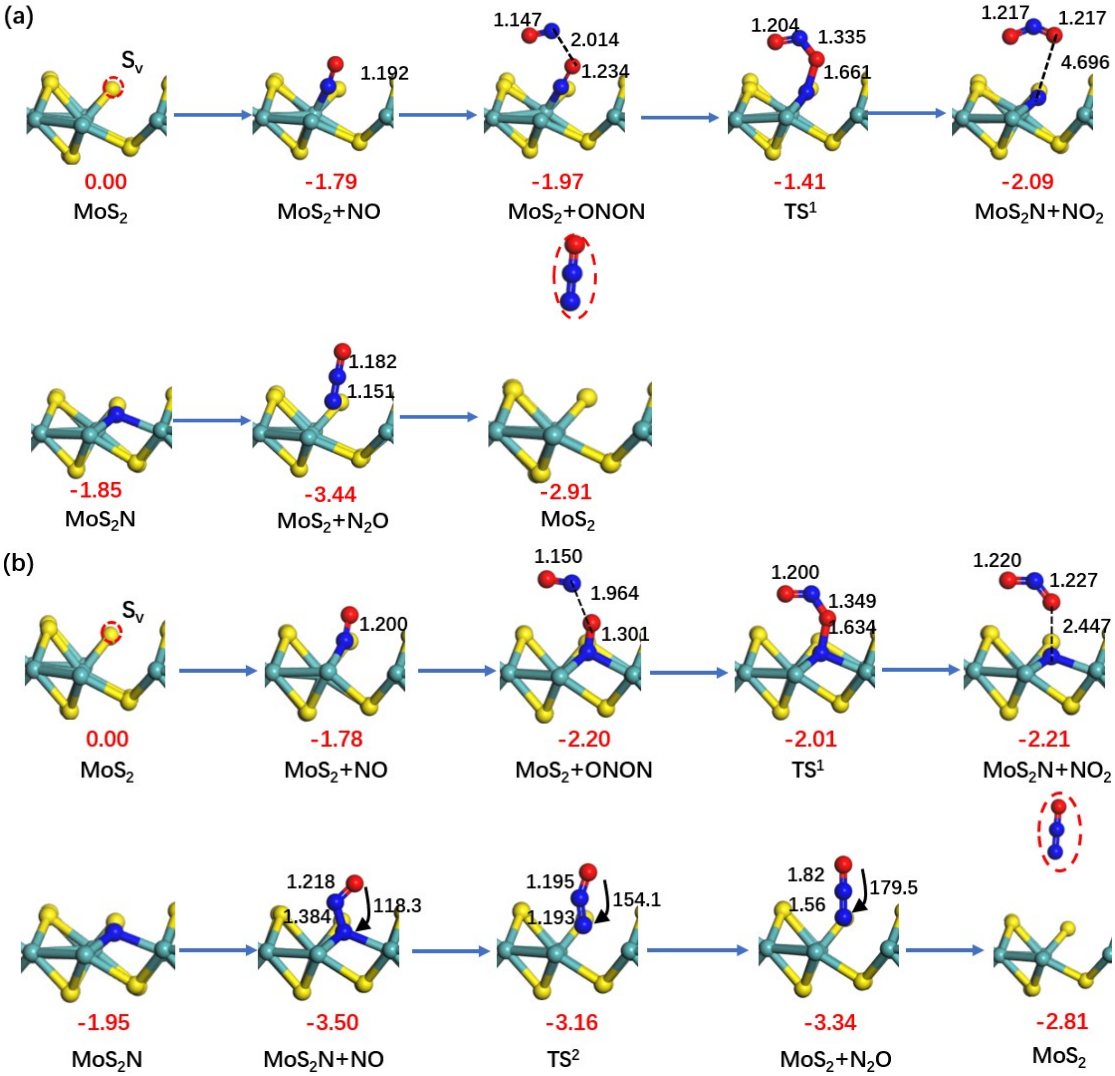

Figure S6. NO disproportionation pathways over defective 1T′-MoS_2 monolayers

under (a) 3% (tensile) strain and (b) -3% (compressive) strain.

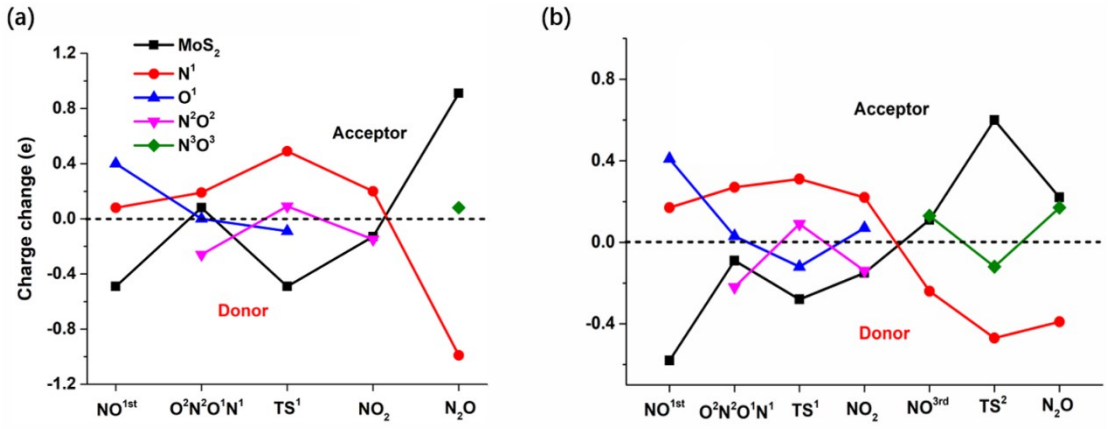

Figure S7. The change of negative charge in the defective 1T′-MoS_2 monolayer and the adsorbents during the NO disproportionation under (a) 3% (tensile) strain and (b) -3% (compressive) strain. NO^{1st} and NO^{3rd} represent the bindings of the first and third NO molecule, respectively. N^1 and O^1 atoms are from the first adsobed NO molecule and N^2O^2 (N^3O^3) denotes the second (third) adsorbed NO molecule. The positive (negative) value means gain (loss) electrons.

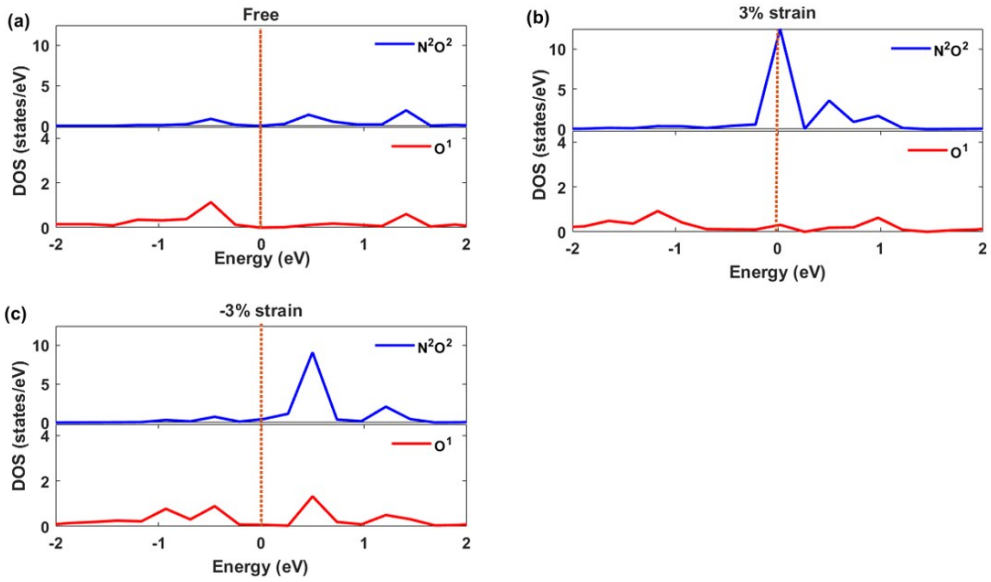

Figure S8. Local density of states (DOS) (projected on the adsorbed second N^2O^2 molecule and O^1 atom from first NO molecule) of defective 1T′-MoS_2 monolayer with S vacancy adsorbed by two NO molecules. The Fermi level is indicated by the red dashed line.

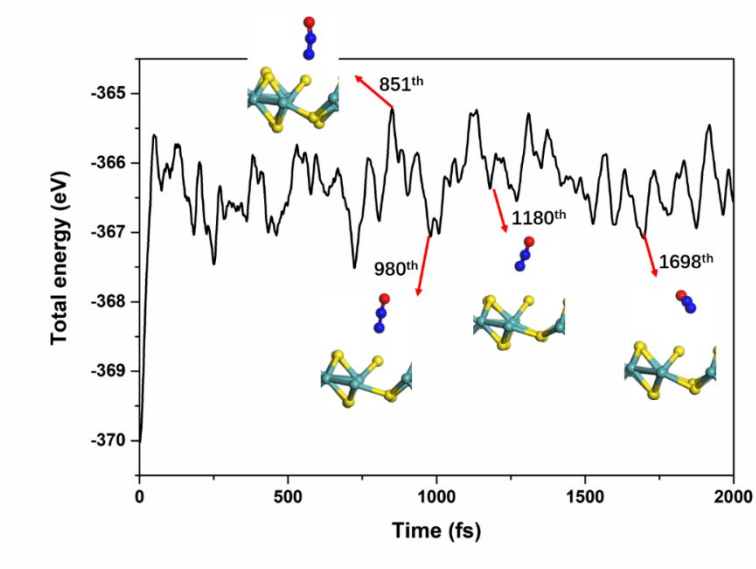

Figure S9. AIMD simulations of the adsorption of N_2O on 3% strained defective 1T′-MoS_2 monolayer. Insets: snapshots of the side views at the 851^{th}, 980^{th}, 1180^{th} and 1698^{th} steps of the simulation at 500 K.

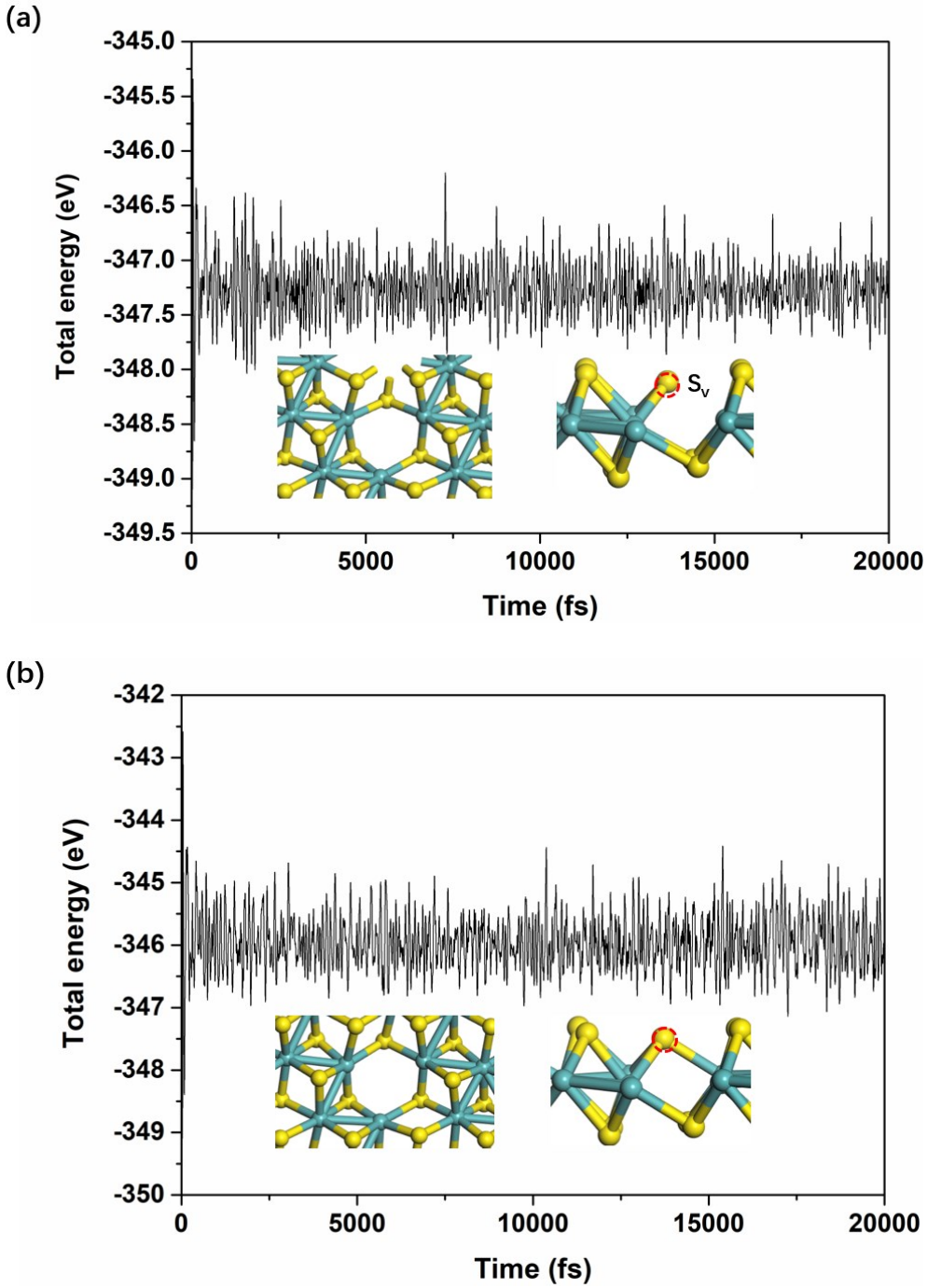

Figure S10 AIMD simulations of -3% strained defective 1T′-MoS_2 monolayer at (a) 300K and (b) 500 K. Insets: snapshots of the top and side views at the end of the simulation, respectively.