Supporting information for

Acid sites on silica-supported molybdenum oxides probed by ammonia adsorption: Experiment and theory

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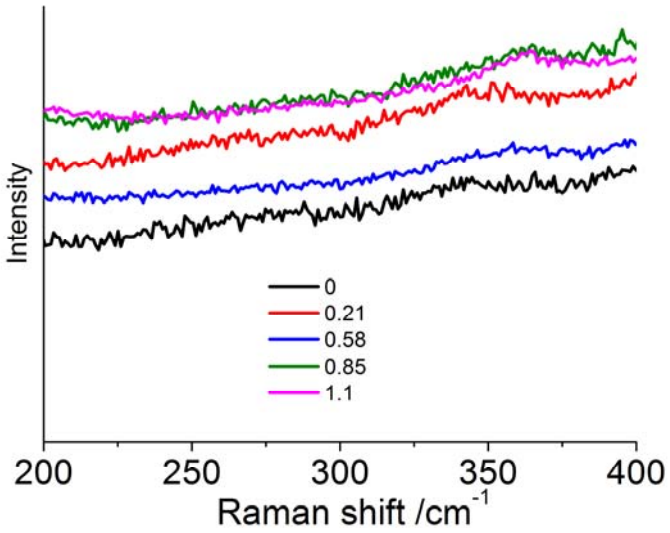

Fig. S1. Raman spectra (excitation at 1.96 eV (632 nm)) in the low-energy range of dehydrated MoO_x/SBA-15 measured at room temperature. The samples were pretreated in 20% O_2 in Ar at 823 K for 0.5 h.

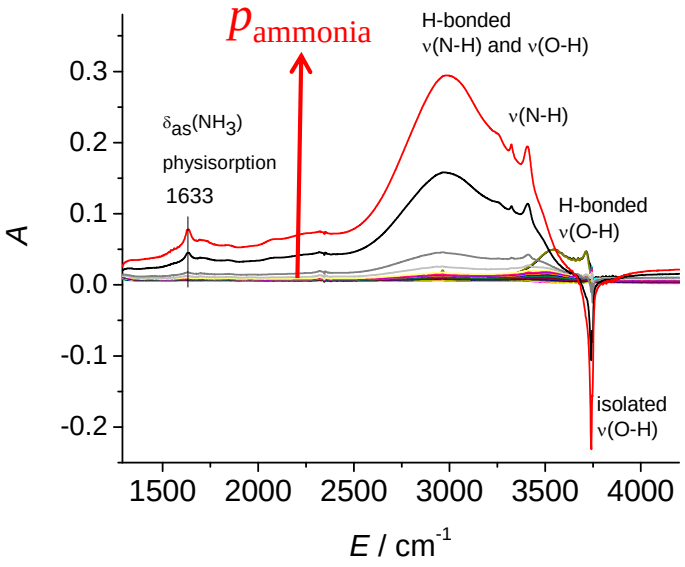

Fig. S2. IR spectra recorded after ammonia adsorption on SBA-15 at 353 K. Ammonia pressure was increased stepwise up to 7 hPa. The sample was pretreated in O_2 at 823 K and at 20 kPa for 0.5 h. The spectrum recorded before ammonia adsorption was used as reference.

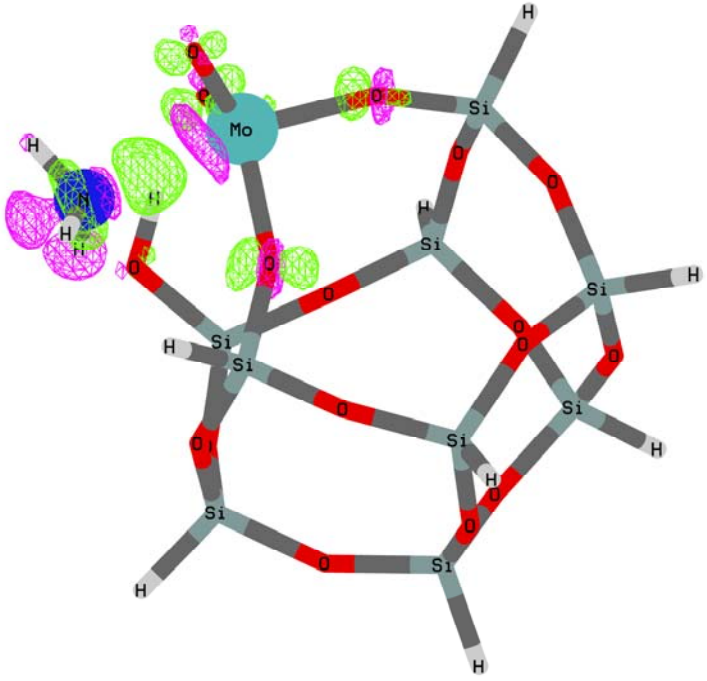

Fig. S3. Plot of electron density difference Δρ of supported molybdenum oxide species as shown in Figure 9A in the main text. The electron density differenceΔρis defined as $\Delta\rho = \rho_{NH_3+Mo/SBA-15} - (\rho_{NH_3} + \rho_{Mo/SBA-15})$, where ρ represents electron density. Pink and green colours show regions (isovalue=0.005) of decreased and increased electron density, respectively.

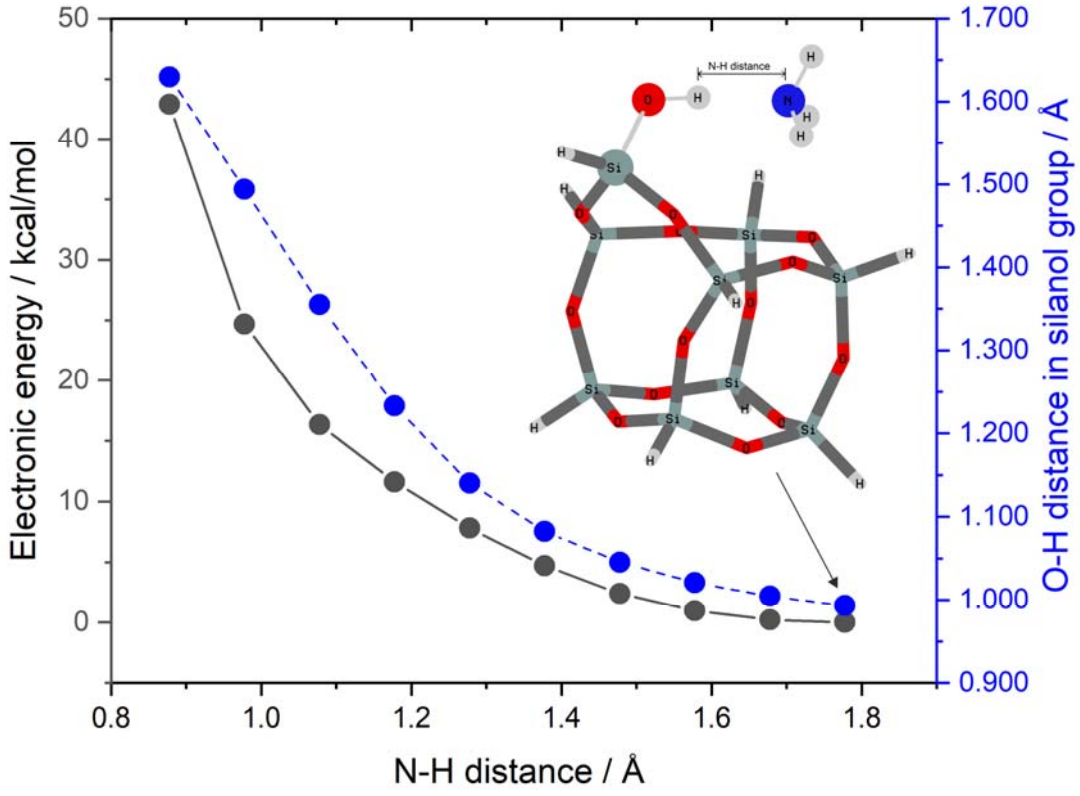

Fig. S4. Relaxed potential energy scan (PES) along the N-H distance of an ammonia molecule approaching the Si-OH group in the silsesquioxane cluster ($Si_8O_{12}H_8$), which represents bare SBA-15, as shown in the inset.

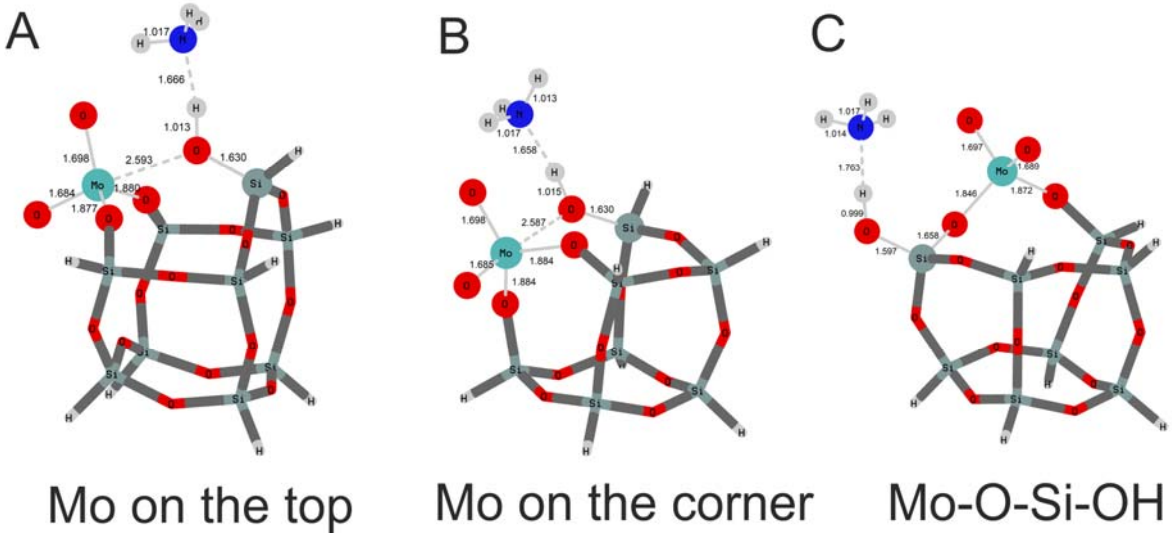

Fig. S5. Various optimized structures of supported molybdenum oxide with an ammonia molecule located near the silanol group. The optimized structures show a hydrogen bond between ammonia and the silanol group N---H-O-Si. The values shown here represent bond distances (Å).

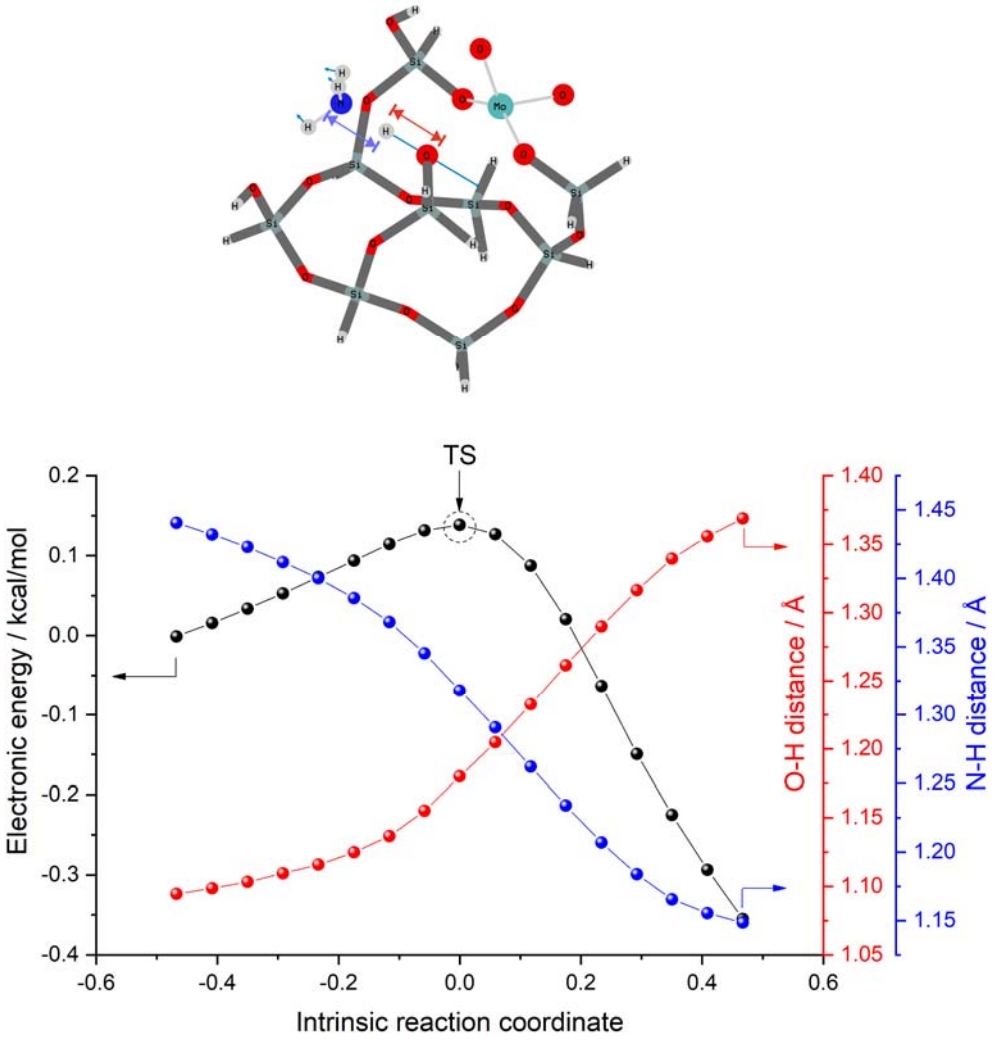

Fig. S6. Intrinsic reaction coordinate **(**IRC) calculation for the transition state (TS) structure. Changes in the O-H distance and N-H distance are illustrated in the representation of the TS structure (red and blue arrows). Small light blue arrows in the representation of the TS structure show displacement vectors of the H atoms in the ammonia molecule and the silanol group for the vibrational mode, which generate imaginary frequencies.