SUPPORTING INFORMATION

Chemical vapor deposition of trimethylaluminium on dealuminated faujasite zeolite

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1. Supplementary results

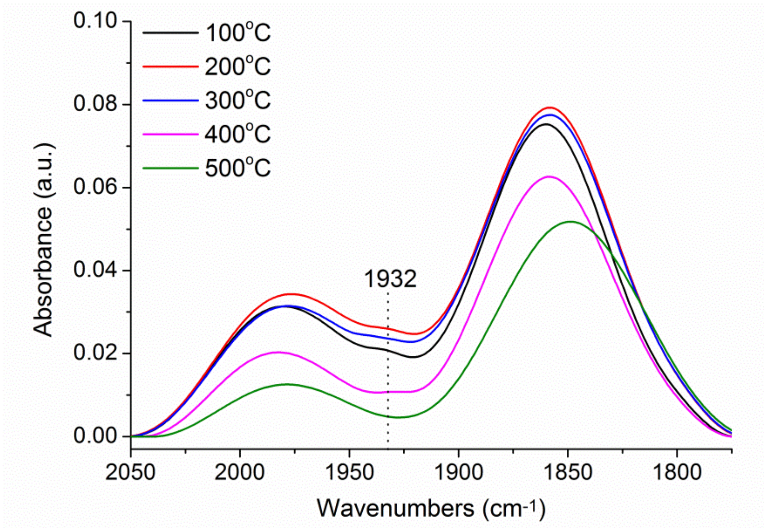

Figure S1. In situ FTIR spectra in the region of *v*(Al-H) collected upon the decomposition of the grafted TMA species on AHFSY zeolite via reduction in 50 mbar H_2 at different temperatures.

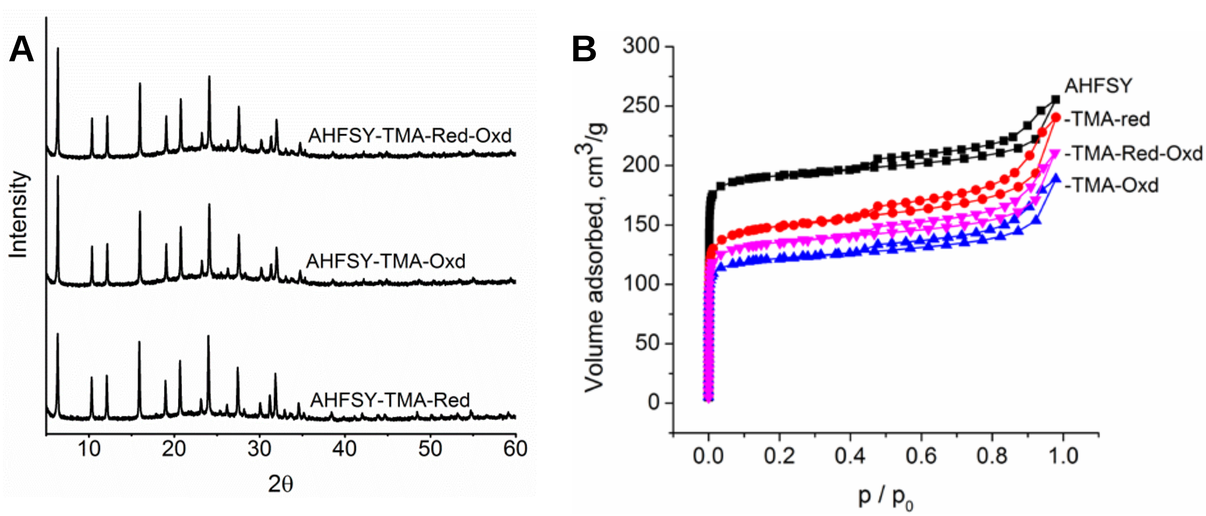

Figure S2. (A) Powder XRD patterns and (B) adsorption-desorption Ar isotherms of TMA-modified zeolites.

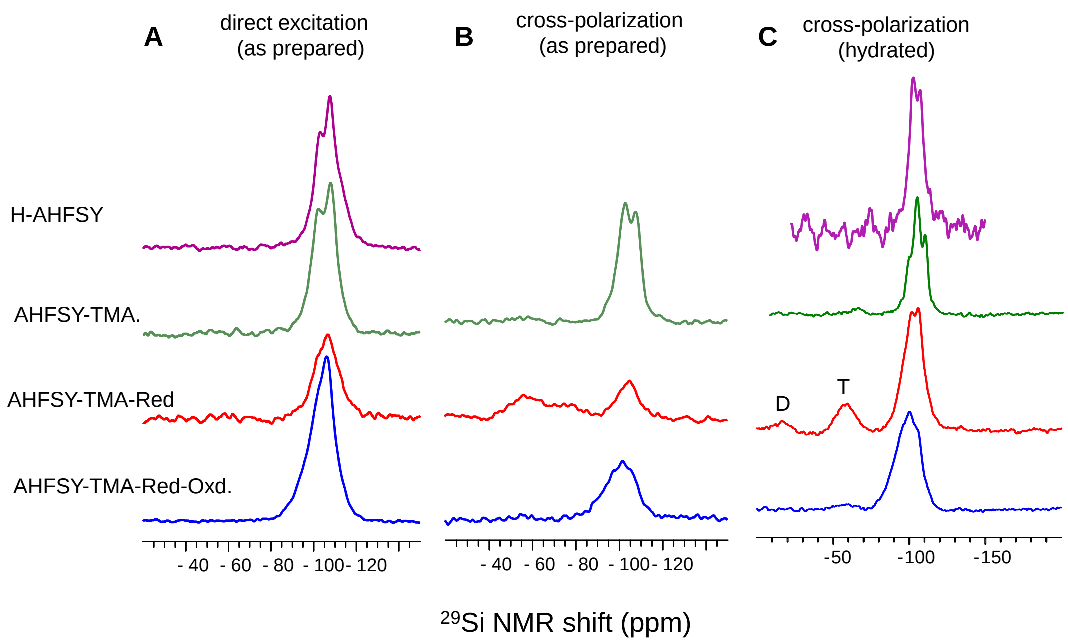

Figure S3. ^{29}Si MAS NMR spectra of AHFSY, AHFSY-TMA, AHFSY-TMA-Red and AHFSY-TMA-Red-Oxd zeolites: (A) high power proton decoupled direct-excitation - and (B) cross-polarization ^{29}Si NMR spectra of the as–prepared zeolites;(C) cross-polarization spectra of air-exposed, hydrated samples

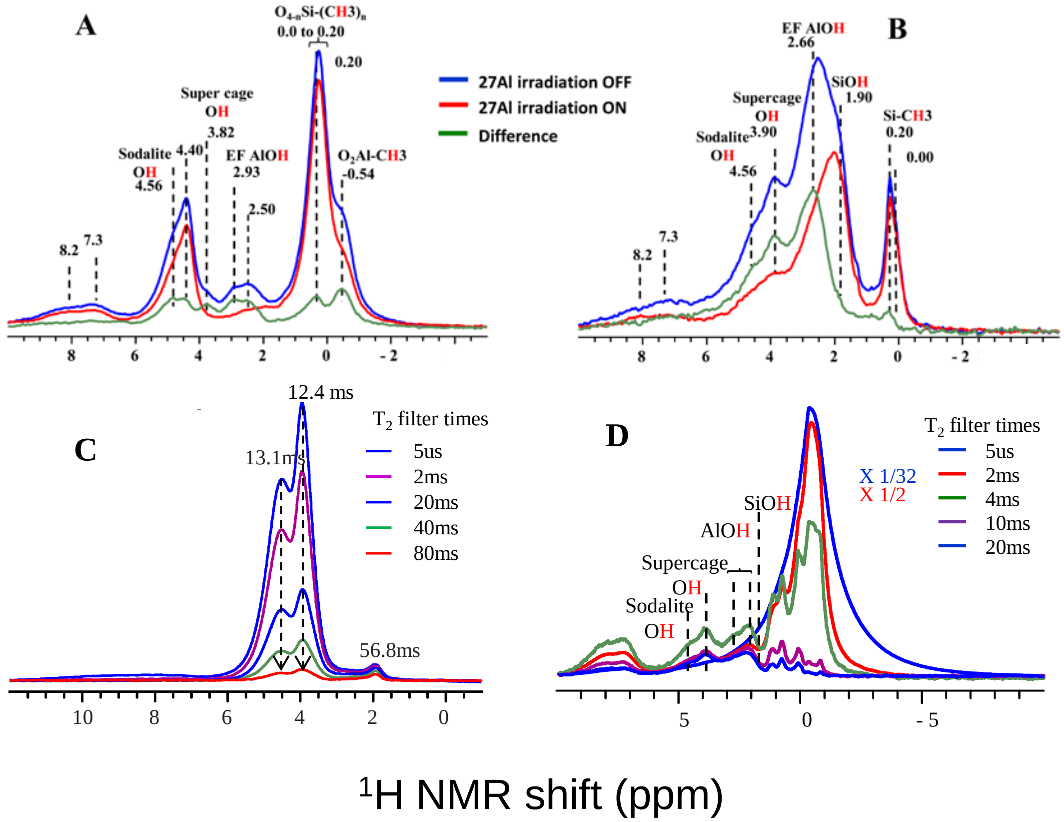

Figure S4. ^{1}H-{^{27}Al} TRAPDOR effect in (A) reduced and (B) reduced-oxidized TMA modified AHFSY zeolite: Blue and red lines represent ^{1}H MAS NMR spectra without and without ^{27}Al irradiation, respectively, while the green line represent a difference spectrum. (C,D) Proton T_2 filtered of dehydrated (C) AHFSY and (D) AHFSY-TMA at varied echo times 2τ= 0.05, 2, 4,10 and 20 ms. The T_2 (1H) values of different sites in dehydrated AHFSY zeolite are mentioned above the peaks. The silanol moiety in AHFSY with δ ~ 2 ppm has a relatively long T_2 value (56 ms).

*** Are silanol signals (~2 ppm) in AHSFY-TMA absent, or hidden under the intense Al-(CH_3)$_n$ signal?**
From the stack plot of the T_2(^{1}H)-filtered spectra of AHFSY-TMA (Fig. S4D) it can be seen that the silanol signal is absent, whereas a small BAS and AlOH fraction is visible. This may be compared with the T_2-filtered spectra of the AHFSY parent zeolite (Fig. S4C), in which the silanol protons decay relatively slowly as a function of the echo time in comparison to the BAS and AlOH fraction. This indicates, that, if *unperturbed* SiOH would be present in AHFSY-TMA, it should be clearly recognizable at long filter time. Since this is not the case, we conclude that the silanol signal is not just hidden under the intense Al(CH_3)$_n$ signal, but is really absent and the silanols have reacted with TMA, as well.

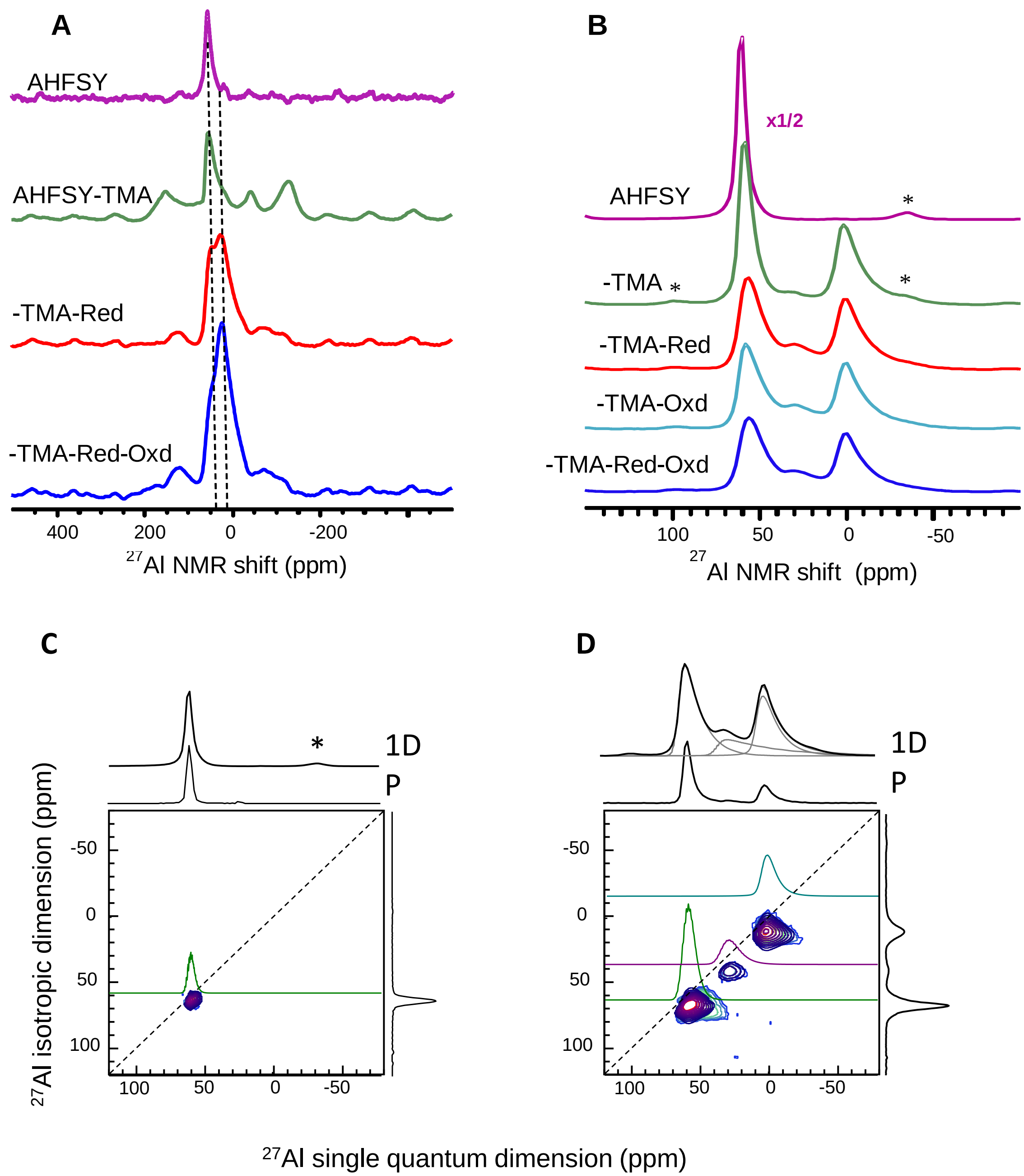

Figure S5. (A, B) 1D single pulse excitation ^{27}Al MAS NMR spectra of (A) dehydrated and (B) hydrated parent and TMA-modified zeolites and (C,D) MQMAS NMR spectra of hydrated (C) AHFSY and (D) AHFSY-TMA-Oxd (spinning side bands are marked with *).

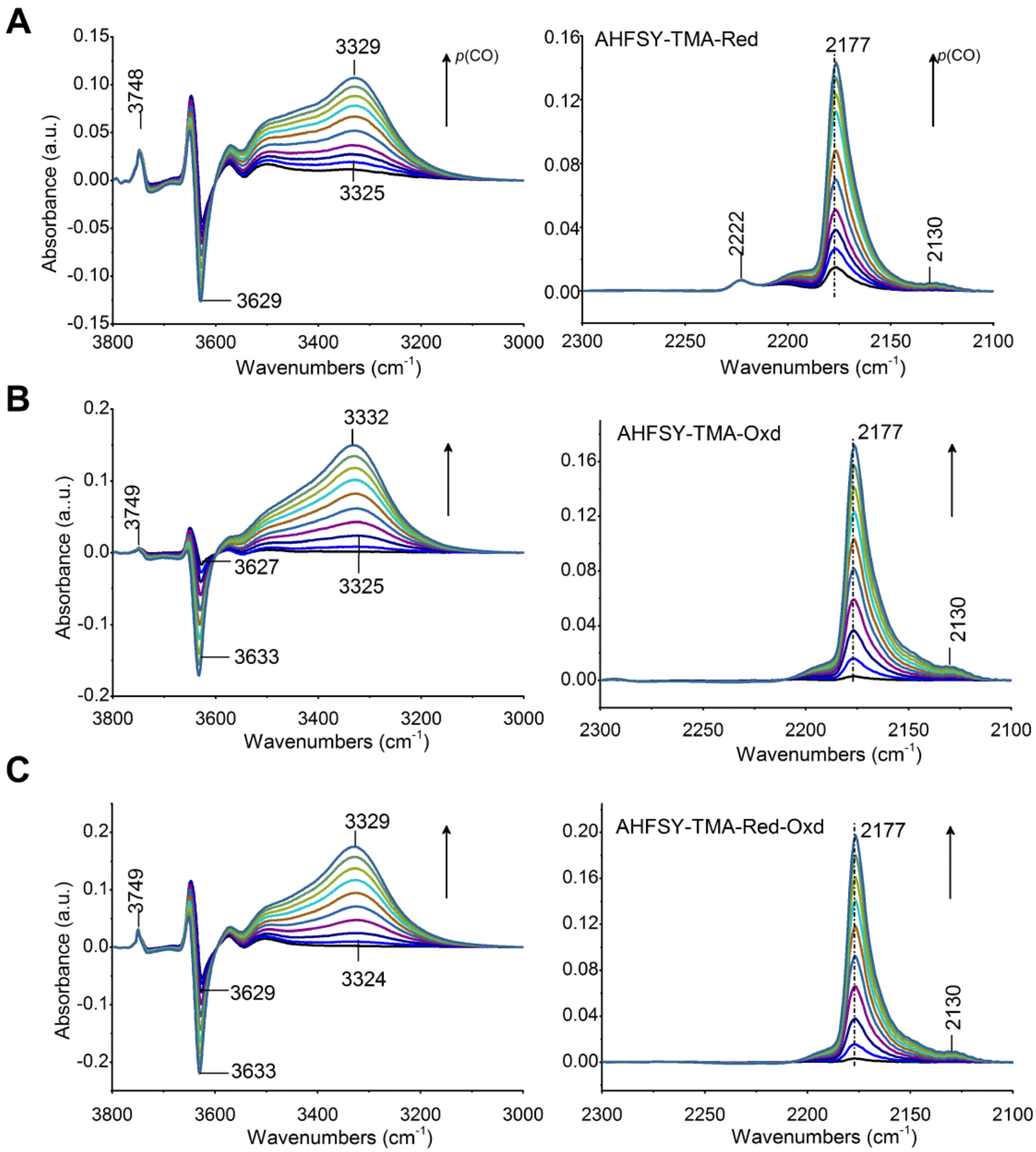

Figure S6. The evolution of *v*(OH) (left panel) and v(CO) (right panel) upon the progressive adsorption of CO on dehydrated (A) AHFSY-TMA-Red, (B) AHFSY-TMA-Oxd, (C) AHFSY-TMA-Red-Oxd at the liquid nitrogen temperature.

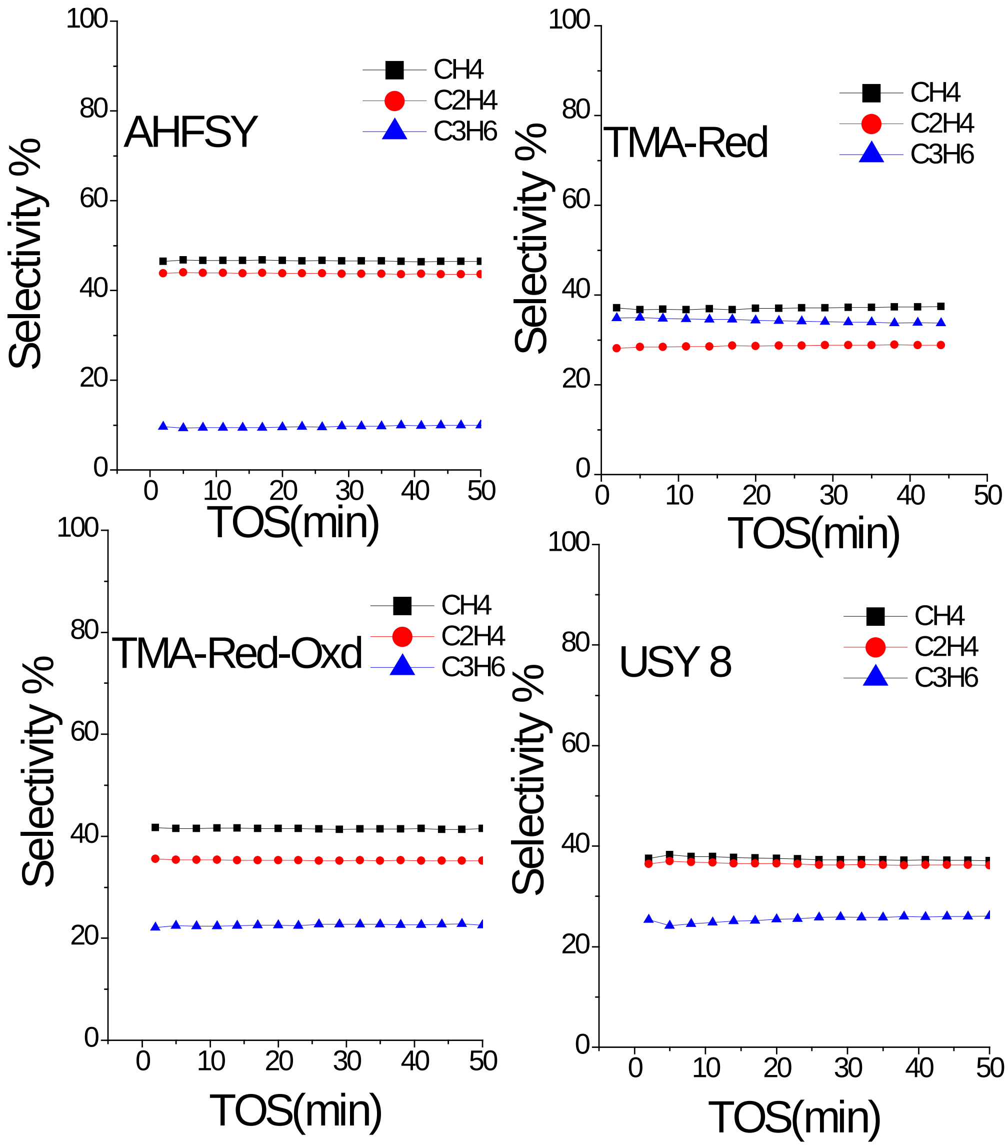

Figure S7. The selectivities of propane cracking at 590ºC.

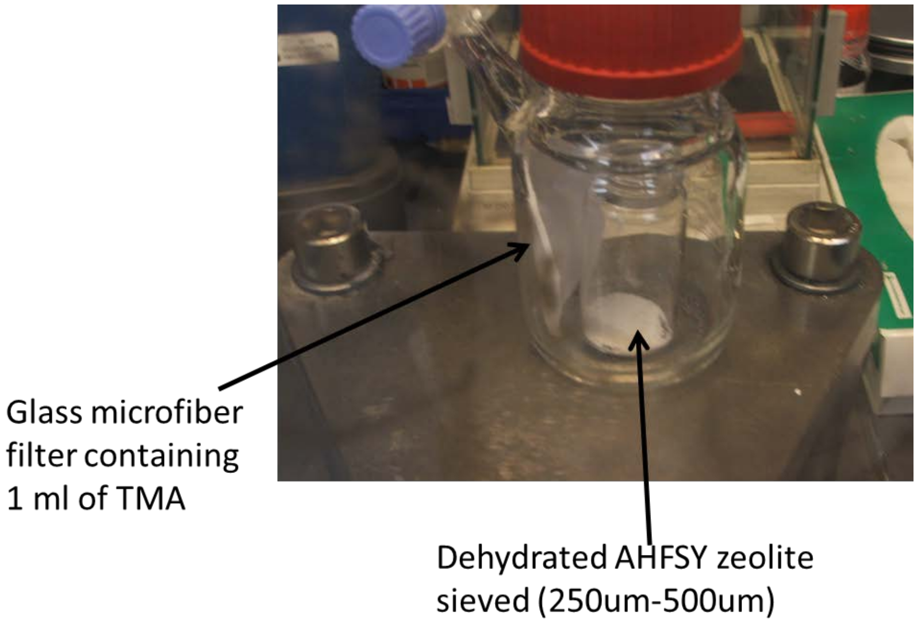

Figure S8: The setup used for CVD of TMA on dehydrated AHFSY zeolite inside the nitrogen-flushed glove-box.