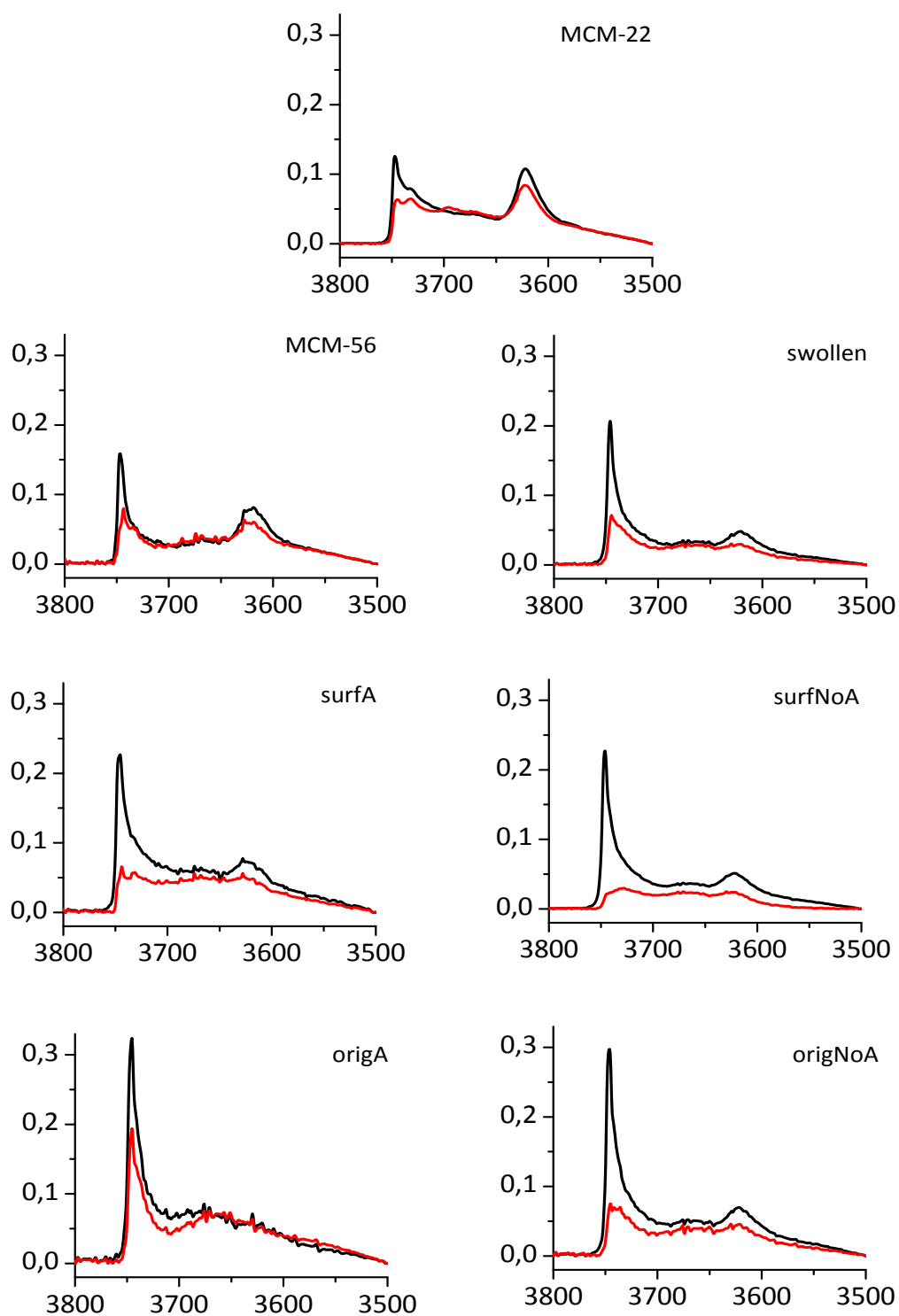
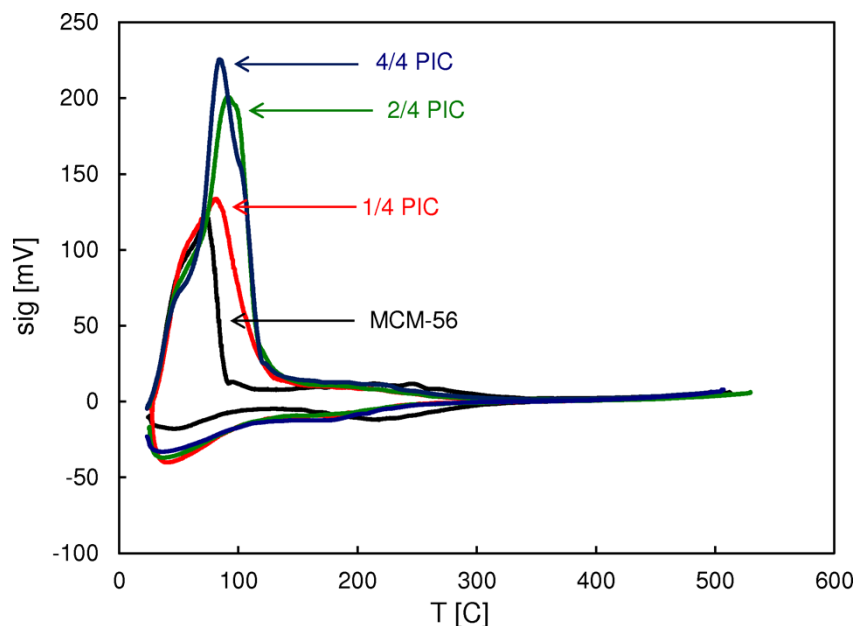


### Supplementary Information

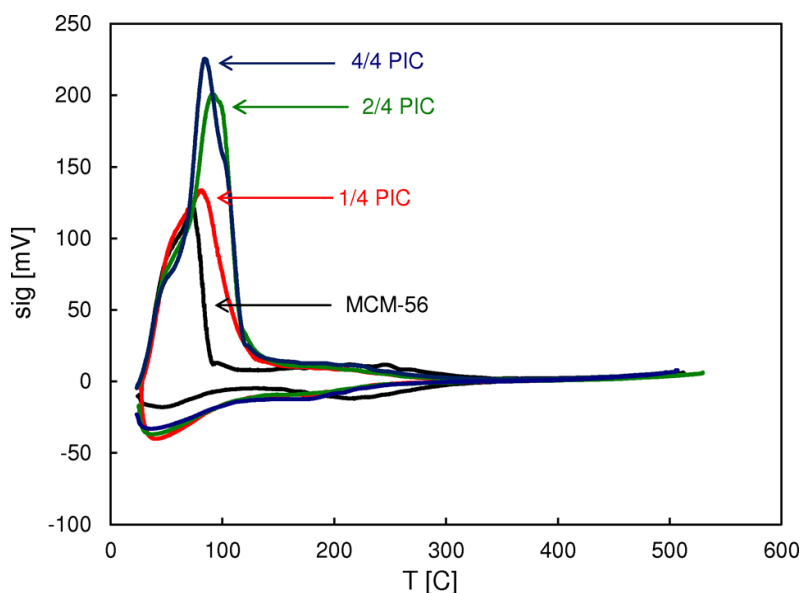
**Figure S1** Adsorption of pivalonitrile on MCM-22, MCM-56 and its delaminated derivatives. Red lines are the spectra of activated samples (activation 1 hour, vacuum 10<sup>-5</sup> Torr, spectra at RT, normalized to the same mass of the pellet: 10 mg)



**Figure S2** The QE-TPDA profiles of the parent and the pillared MCM-56 zeolites recorded at heating/cooling rate of 10°C/min. The micropore volumes were calculated by integration of the desorption profiles in the temperature range 150-350°C.



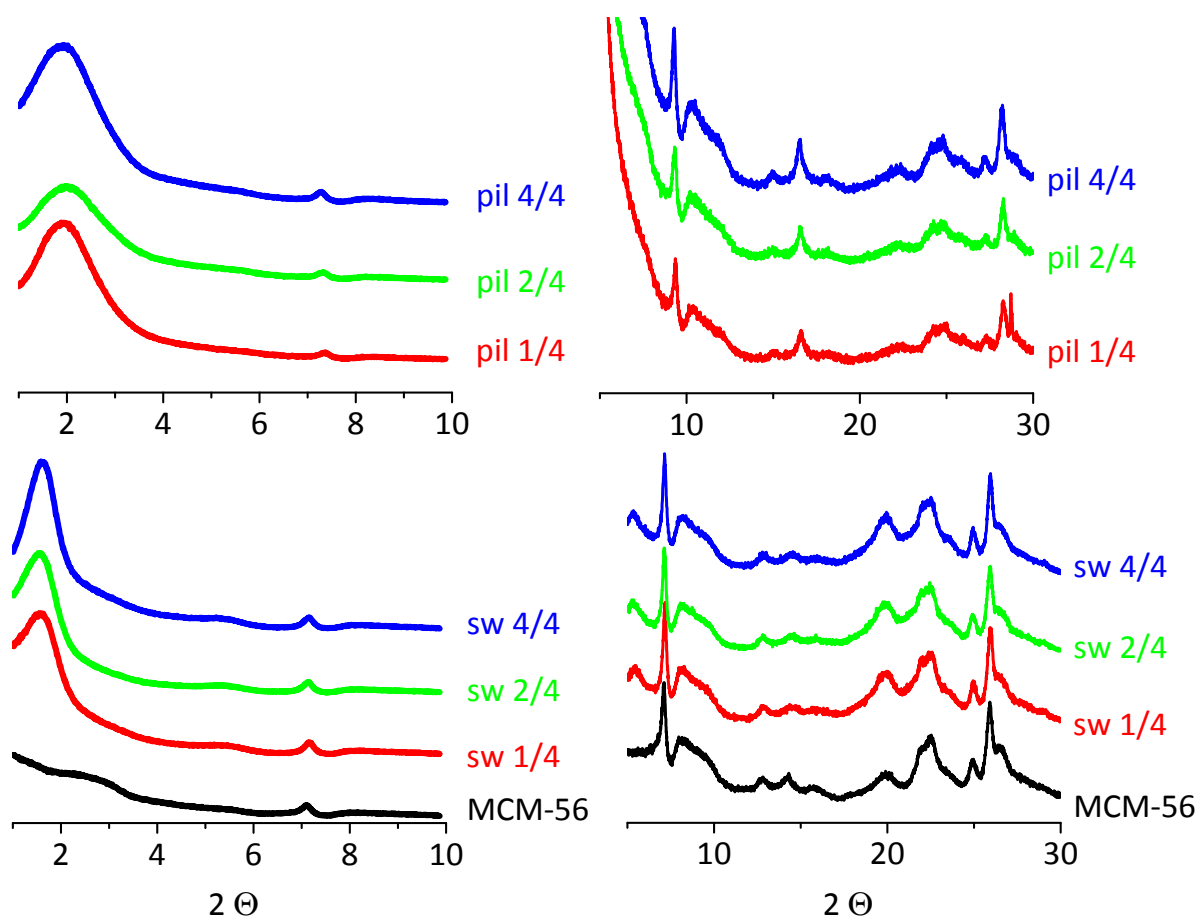
**Figure S3** The QE-TPDA profiles of the parent and the pillared MCM-56 zeolites recorded at heating/cooling rate of 10°C/min. The micropore volumes were calculated by integration of the desorption profiles in the temperature range 150-350°C.



Quasi-equilibrated temperature programmed desorption and adsorption (QE-TPDA) of nonane was employed as a complementary method of porosity characterization. The QE-TPDA measurements were performed in a flow thermodesorption system equipped with the chromatographic thermal conductivity detector (TCD). Prior the QE-TPDA measurement the sample (ca 10 mg) was activated by heating in flow of He up to 500°C, at 10°C/min. After cooling the sample the carrier gas was changed from pure He to He saturated at room

temperature with nonane vapor (ca 0.4 mol%). When the initial adsorption at room temperature was completed, the QE-TPDA experiment was performed by cyclic heating and cooling the sample according to a programmed temperature profile. This method and details of the experiments are described in more details elsewhere [W. Makowski, P. Kuśtrowski, Probing pore structure of microporous and mesoporous molecular sieves by quasi-equilibrated temperature programmed desorption and adsorption of n-nonane, *Micropor.Mesopor. Mater.*, 102 (2007) 283-289, doi: 10.1016/j.micromeso.2007.01.009].

**Figure 2** – enlarged version. Low and high angle XRD's of MCM-56 swollen with CTMA-OH of different concentration and pillared.



**Figure 3** - enlarged version. Nitrogen isotherms for parent and pillared MCM-56. Isotherms are not shifted for MCM-56 and 4/4 samples, the offset for 2/4 is 100 and for 1/4 is 200 cm<sup>3</sup>/g STP.

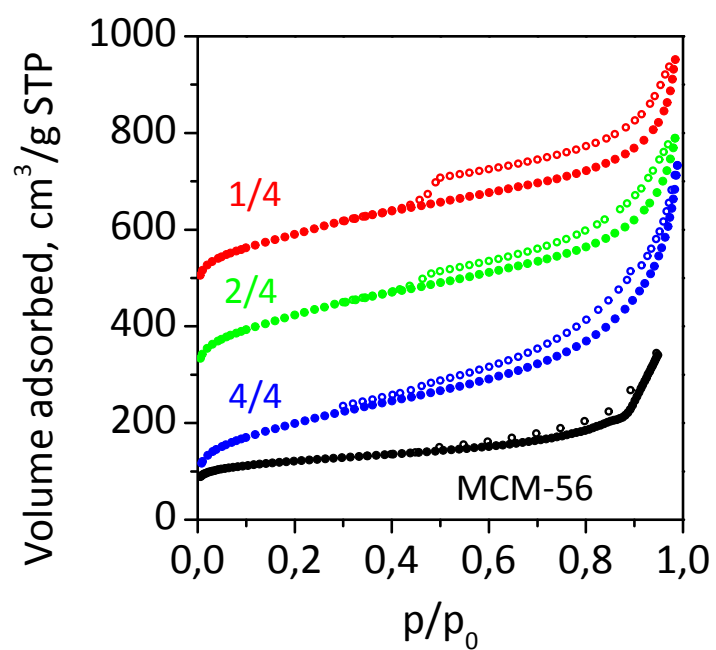


Figure 4 – enlarged version. FTIR spectra of pillared MCM-56 in the region of skeletal (A) and OH (B) vibrations

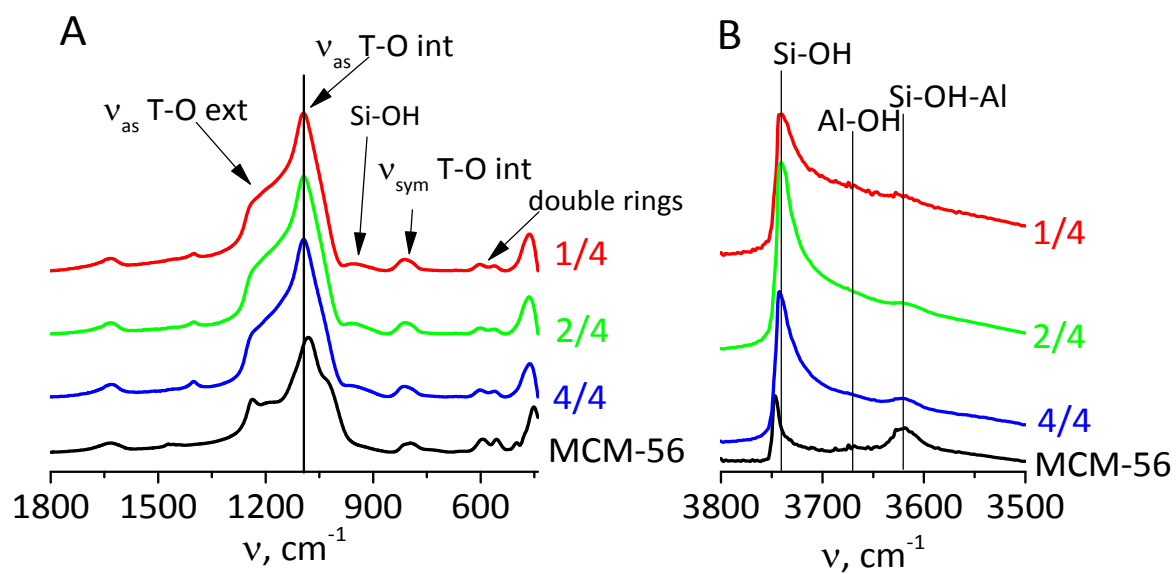


Figure 5 – enlarged version.  $^{29}\text{Si}$  MAS NMR (A) and  $^{27}\text{Al}$  MAS NMR (B) spectra of selected MCM-56 materials.

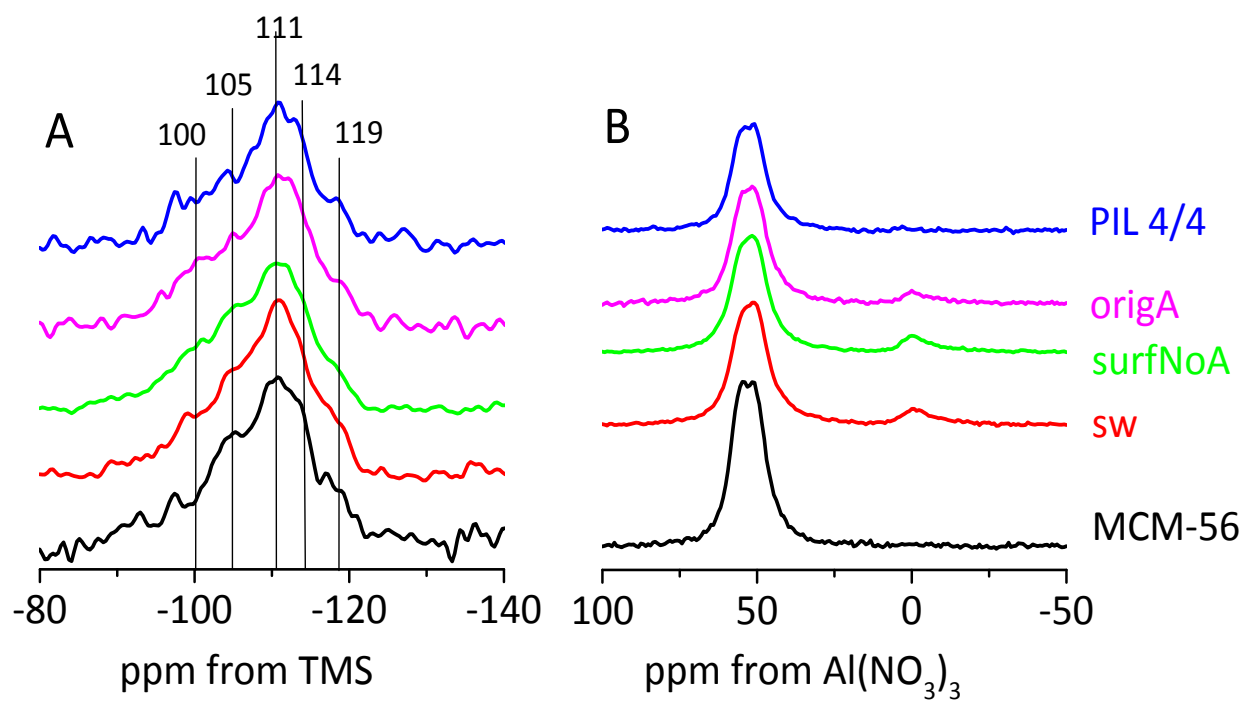
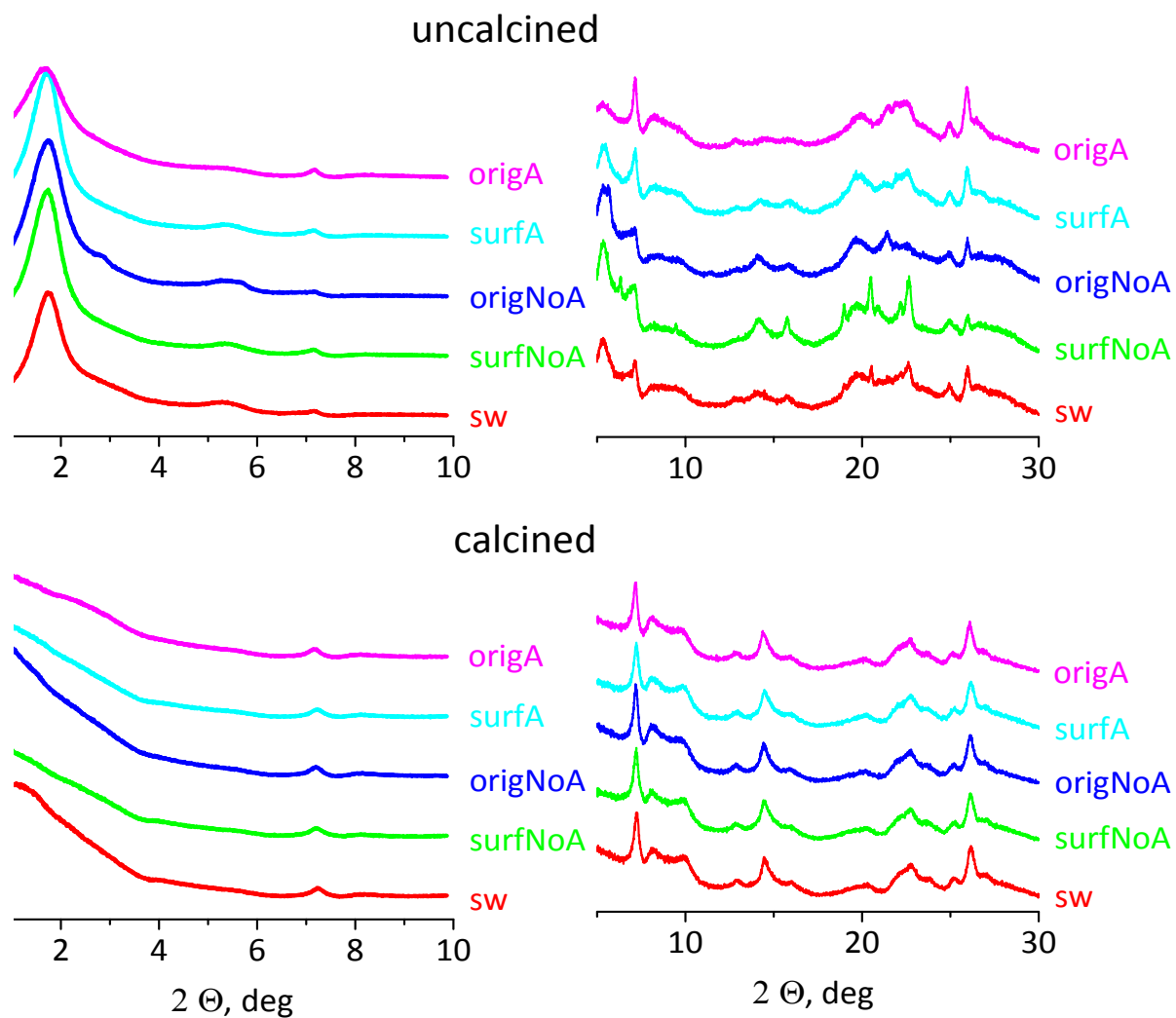


Figure 6 – enlarged version. Low and high angle XRD's of swollen and sonicated MCM-56 samples.



1. Figure 7 – enlarged version (A) and offset scale (B). Nitrogen isotherms for parent and delaminated MCM-56 samples

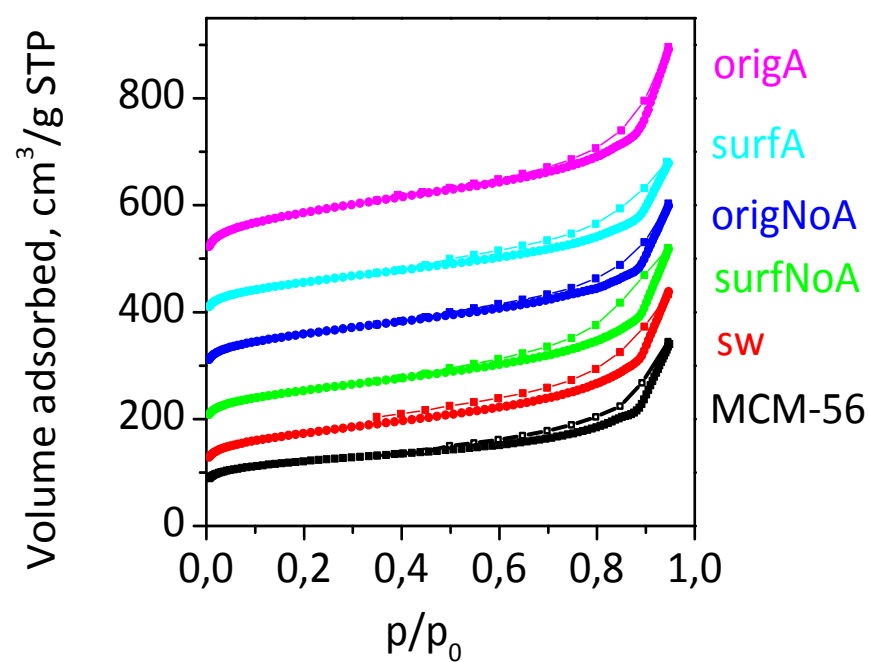
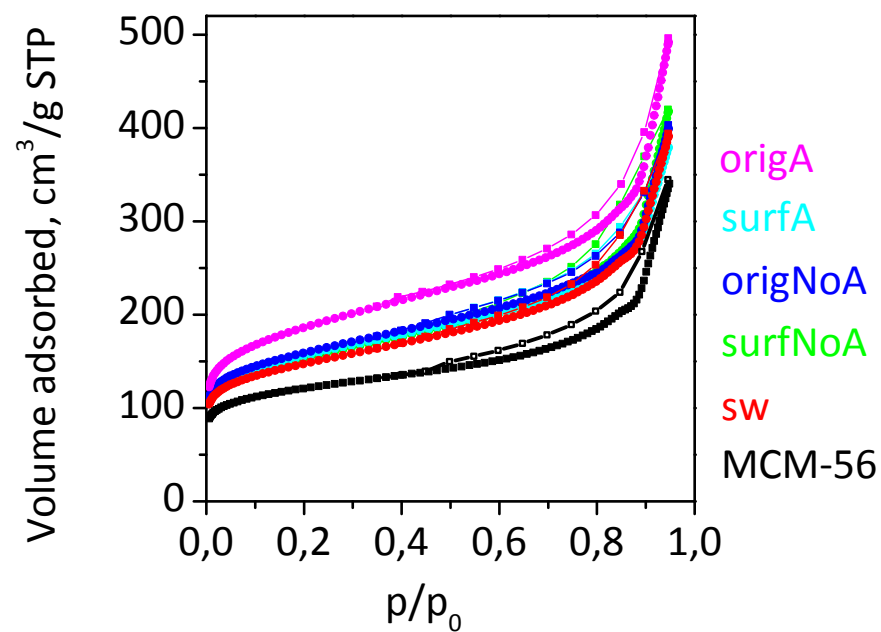


Figure 8 enlarged version. FTIR spectra of MCM-56 and delaminated derivatives. A – region of skeletal vibrations, B – region of OH vibrations.

