

## Supporting information for:

Consequences of secondary zeolite growth on catalytic  
performance in DMTO studied over DDR and CHA

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### Product calculations.

Conversion, selectivities and yields were calculated on a molar carbon basis. Thus, conversion (%) was defined as the fraction of DME consumed during the reaction:

$$X = \frac{n_{C,DME_{in}} - n_{C,DME_{out}}}{n_{C,DME_{in}}} \cdot 100\% \quad (1)$$

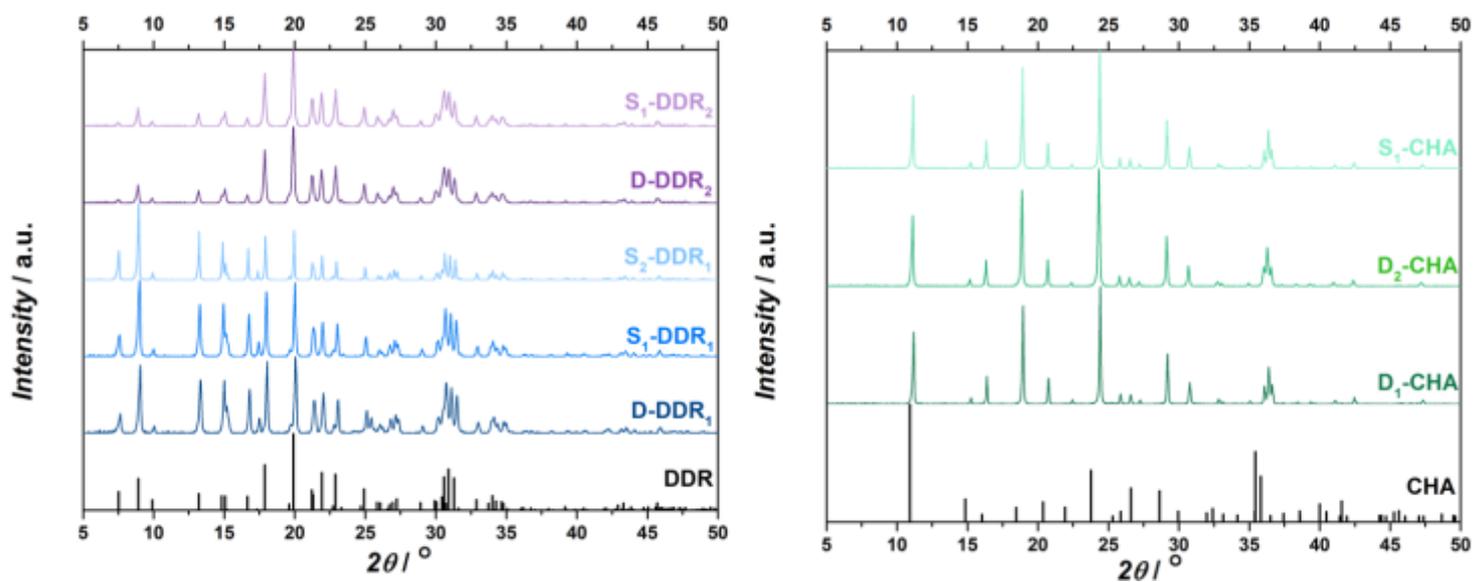
the selectivity (%) towards ethylene (2) and propylene (3) was calculated based on the carbon number as follows:

$$S_{ethylene} = \frac{2 \cdot n_{C_2H_4}}{2 \cdot n_{C,DME_{in}} - 2 \cdot n_{C,DME_{out}}} \cdot 100\% \quad (2)$$

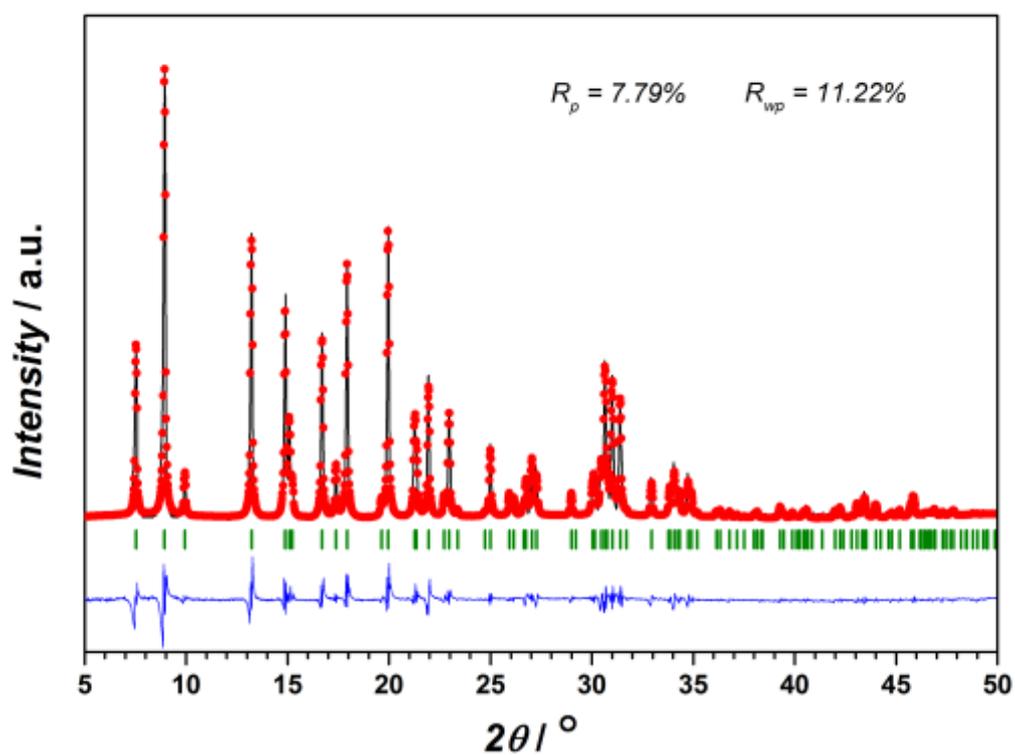
$$S_{propylene} = \frac{3 \cdot n_{C_3H_6}}{2 \cdot n_{C,DME_{in}} - 2 \cdot n_{C,DME_{out}}} \cdot 100\% \quad (3)$$

and the yield (%) of a component  $i$  was defined from its selectivity and DME conversion:

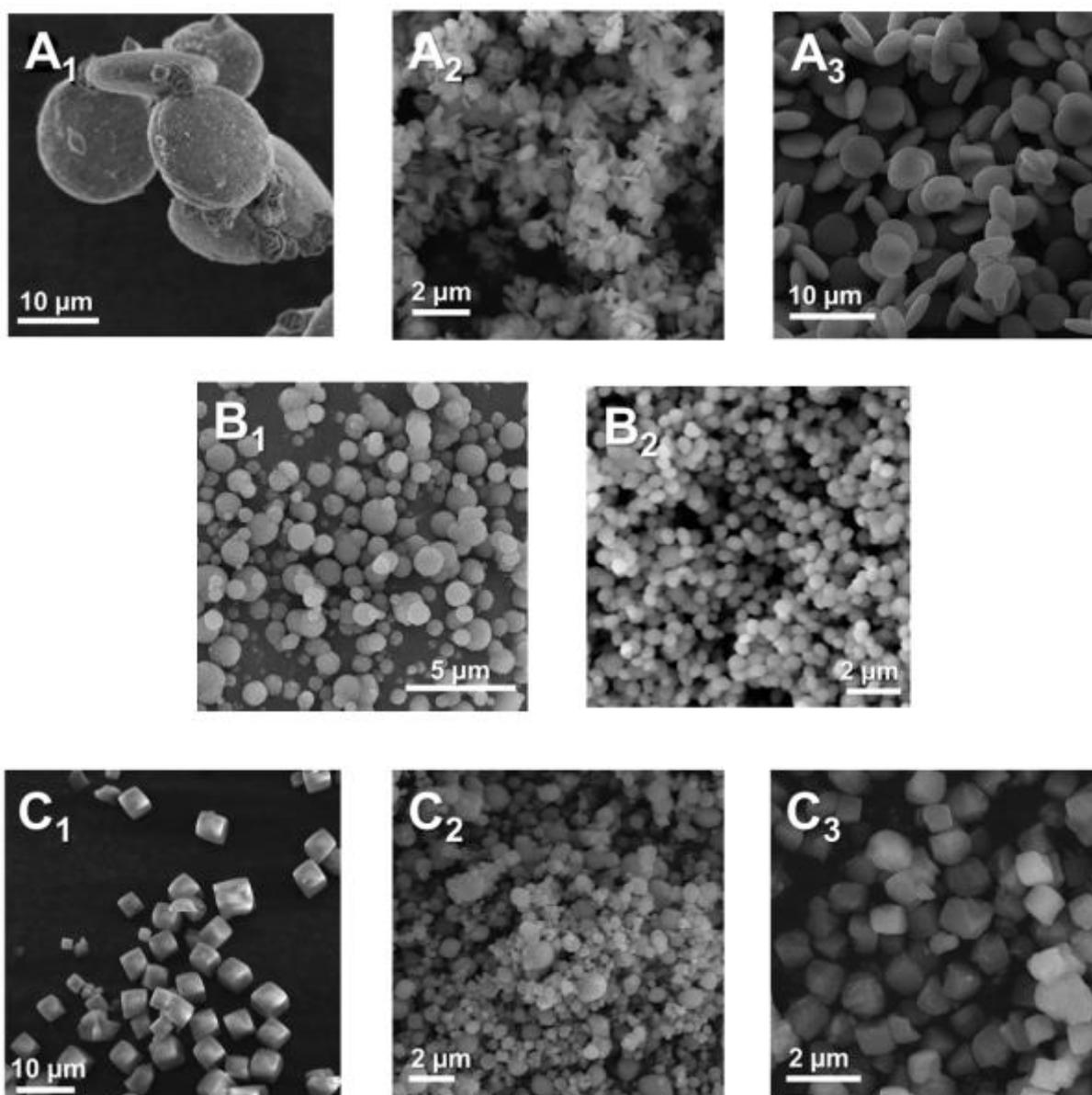
$$Y_i = \frac{S_i \cdot X}{100} \quad (4)$$



**Figure S1.** XRD patterns of zeolites with (*left*) DDR and (*right*) CHA topology together with reference patterns from database of zeolite structures.



**Figure S2.** Pawley fitting plot of Sigma-1 zeolite. Experimental data are presented as a black solid line, calculated one by red dots and difference between them as blue solid line. The Bragg positions of the peaks are represented as green sticks. The blue line represents the difference between experimental data and fitting.



**Figure S3.** SEM images of (A) Sigma-1, (B) ZSM-58 and (C) SSZ-13 zeolites prepared by direct hydrothermal and seeded synthesis.

(A<sub>1</sub>) – D-DDR<sub>1</sub>

(A<sub>2</sub>) – S<sub>1</sub>-DDR<sub>1</sub>

(A<sub>3</sub>) – S<sub>2</sub>-DDR<sub>1</sub>

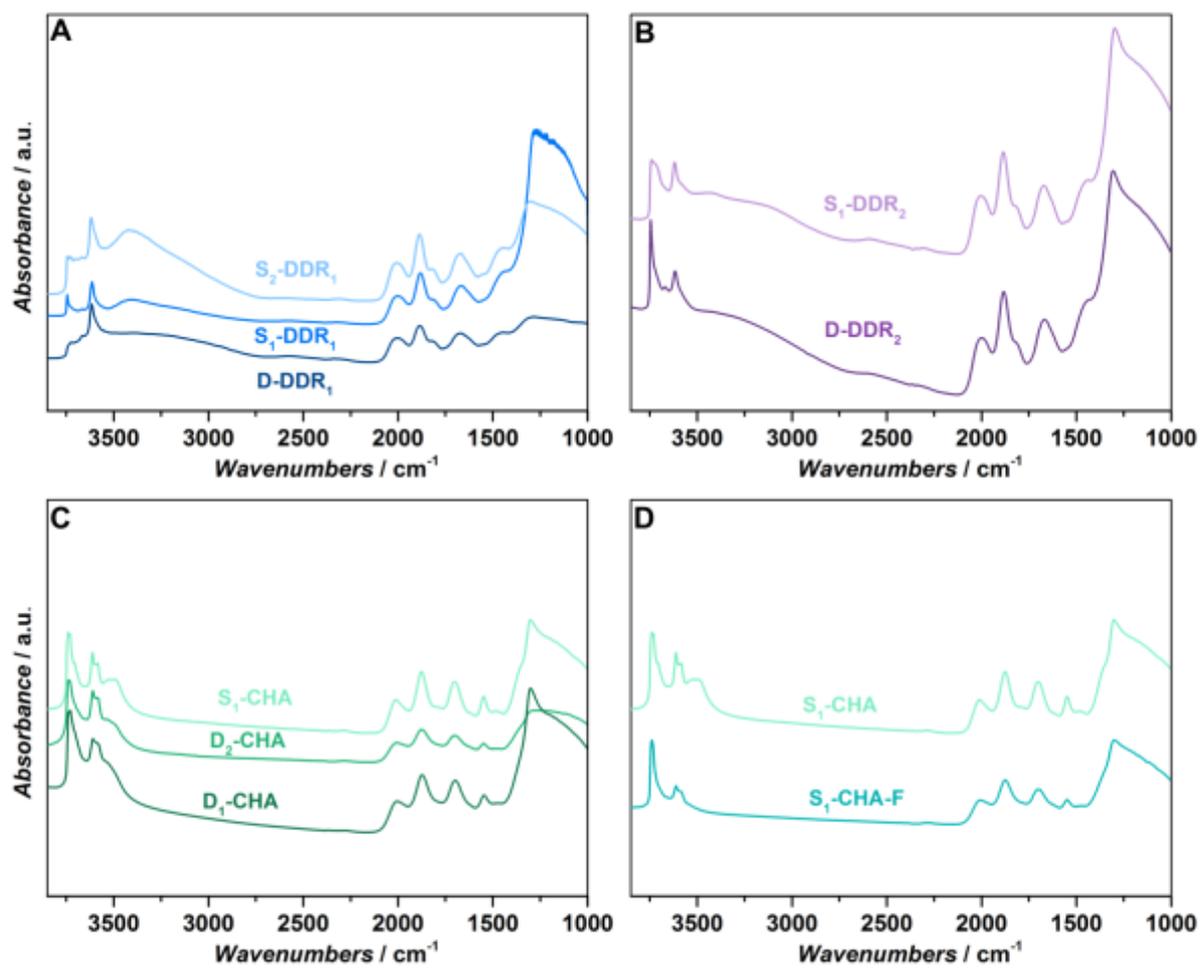
(B<sub>1</sub>) – D-DDR<sub>2</sub>

(B<sub>2</sub>) – S<sub>1</sub>-DDR<sub>2</sub>

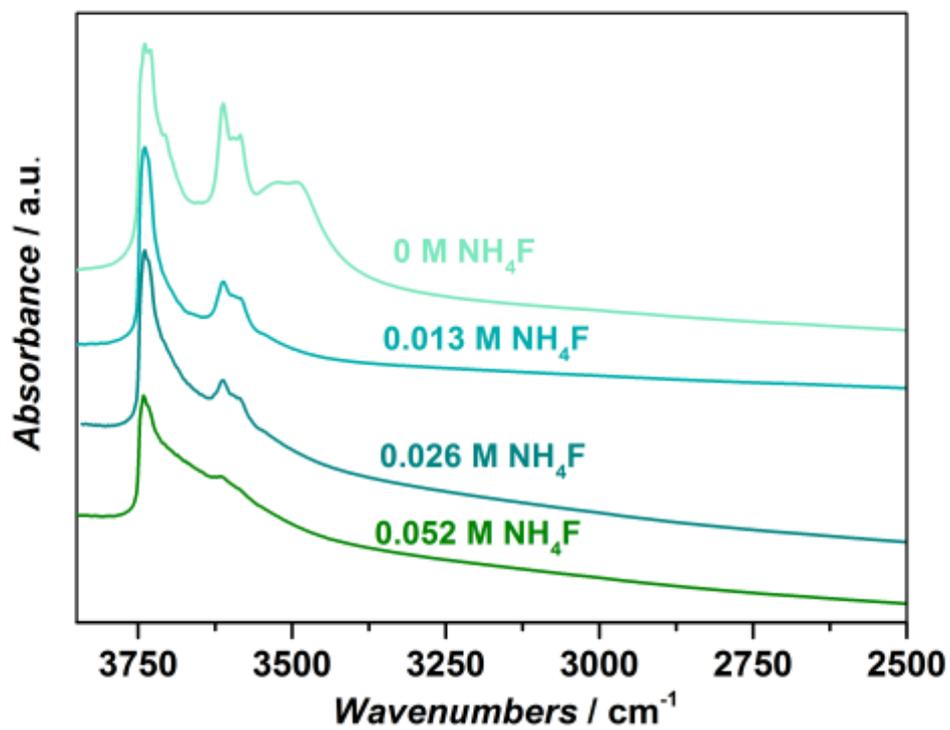
(C<sub>1</sub>) - D<sub>1</sub>-CHA

(C<sub>2</sub>) – D<sub>2</sub>-CHA

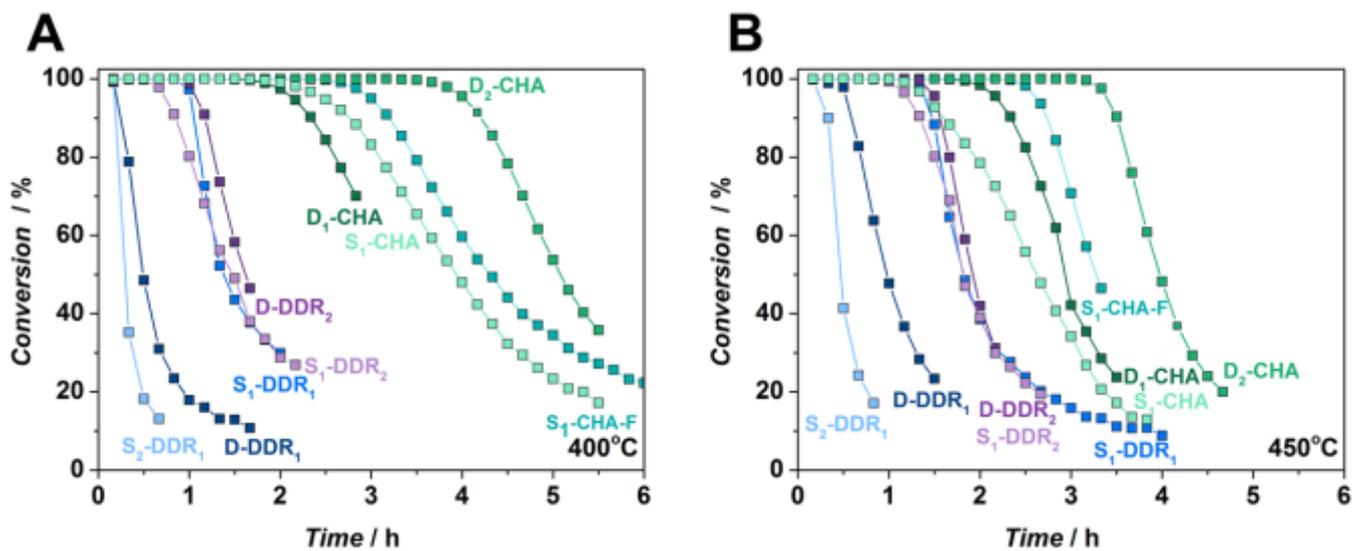
(C<sub>3</sub>) – S<sub>1</sub>-CHA



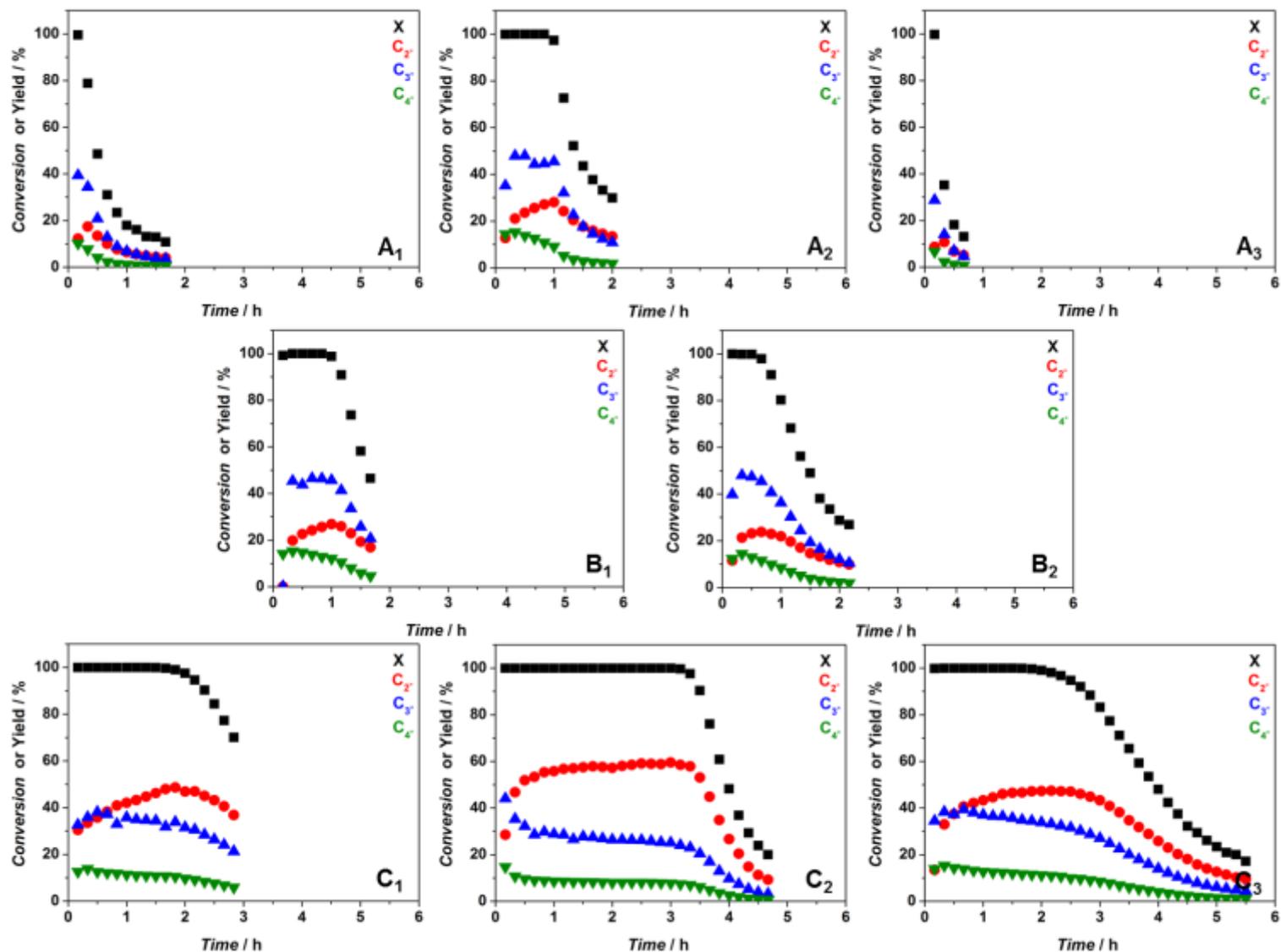
**Figure S4.** FTIR spectra of activated Sigma-1 (A), ZSM-58 (B), SSZ-13(C) and fluoride-modified SSZ-13 (D) zeolites.



**Figure S5.** FTIR spectra of S1-CHA treated with different concentrations of  $\text{NH}_4\text{F}$ .



**Figure S6.** Dimethyl ether conversion as a function of time on stream at (A) 400°C and (B) 450°C for the zeolites under study.  $M_{cat} = 0.5$  g,  $WHSV = 1.23$  g<sub>DME</sub> g<sub>cat</sub><sup>-1</sup>h<sup>-1</sup>.



**Figure S7.** Dimethyl ether conversion and yields of ethylene, propylene and butenes as a function of time at 400°C for the zeolites under study.  $M_{cat} = 0.5$  g,  $WHSV = 1.23$  g<sub>DME</sub> g<sub>cat</sub><sup>-1</sup> h<sup>-1</sup>.

(A<sub>1</sub>) – D-DDR<sub>1</sub>

(A<sub>2</sub>) – S<sub>1</sub>-DDR<sub>1</sub>

(A<sub>3</sub>) – S<sub>2</sub>-DDR<sub>1</sub>

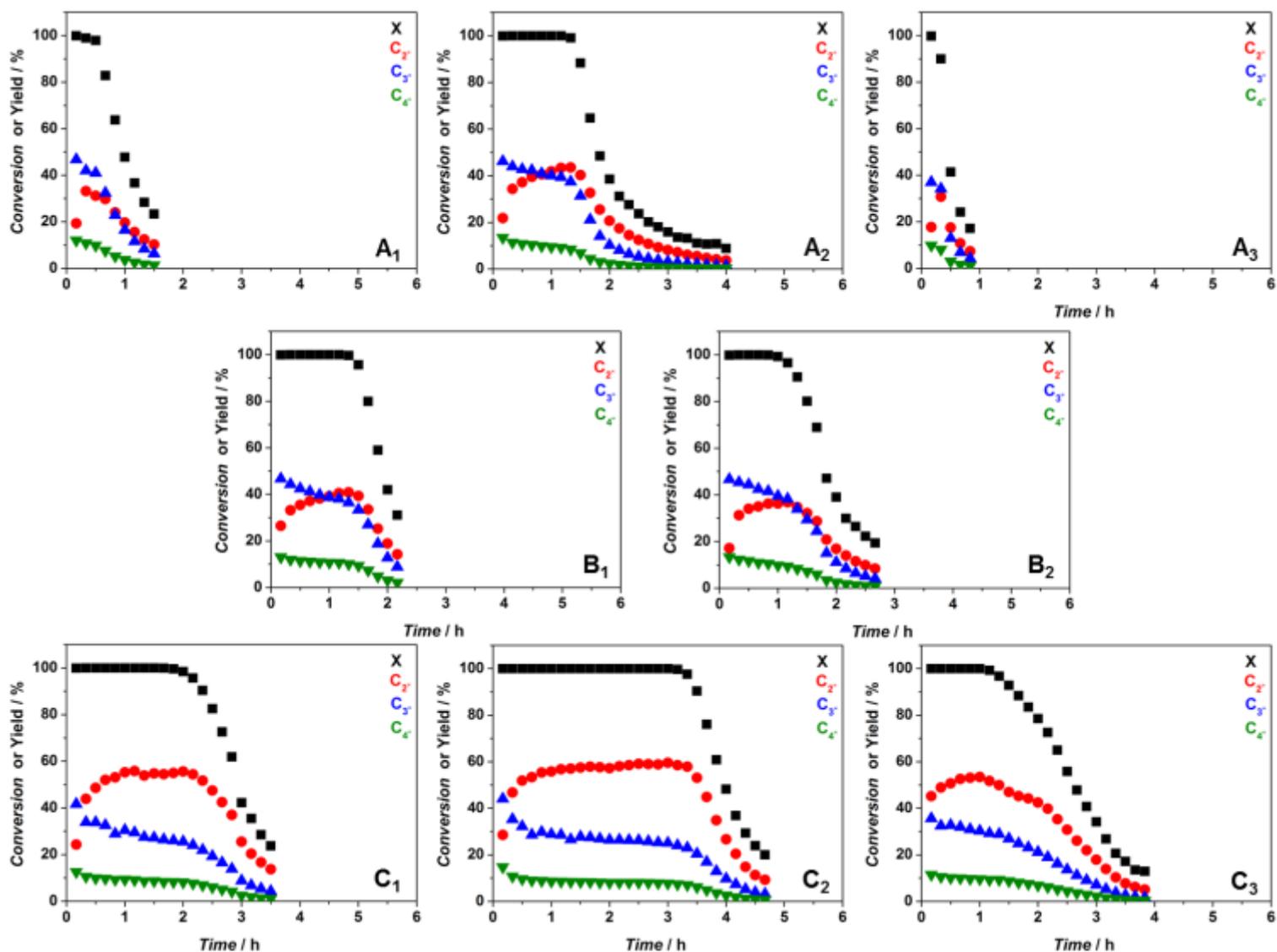
(B<sub>1</sub>) – D-DDR<sub>2</sub>

(B<sub>2</sub>) – S<sub>1</sub>-DDR<sub>2</sub>

(C<sub>1</sub>) - D<sub>1</sub>-CHA

(C<sub>2</sub>) – D<sub>2</sub>-CHA

(C<sub>3</sub>) – S<sub>1</sub>-CHA



**Figure S8.** Dimethyl ether conversion and yields of ethylene, propylene and butenes as a function of time at 450°C for the zeolites under study.  $M_{cat} = 0.5$  g,  $WHSV = 1.23$  g<sub>DME</sub> g<sub>cat</sub><sup>-1</sup> h<sup>-1</sup>.

(A<sub>1</sub>) – D-DDR<sub>1</sub>

(A<sub>2</sub>) – S<sub>1</sub>-DDR<sub>1</sub>

(A<sub>3</sub>) – S<sub>2</sub>-DDR<sub>1</sub>

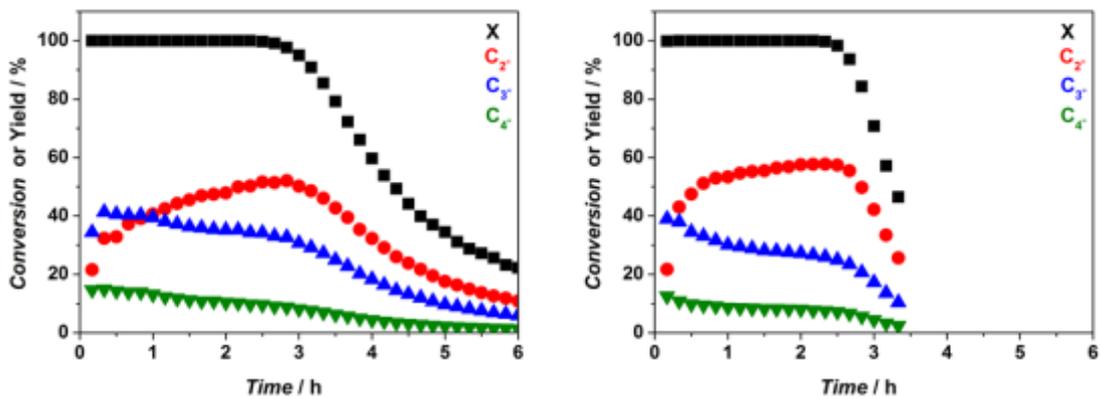
(B<sub>1</sub>) – D-DDR<sub>2</sub>

(B<sub>2</sub>) – S<sub>1</sub>-DDR<sub>2</sub>

(C<sub>1</sub>) – D<sub>1</sub>-CHA

(C<sub>2</sub>) – D<sub>2</sub>-CHA

(C<sub>3</sub>) – S<sub>1</sub>-CHA



**Figure S9.** Dimethyl ether conversion and yields of ethylene, propylene and butenes as a function of time obtained over S<sub>1</sub>-CHA-F (*left*) at 400°C and (*right*) at 450°C.  $M_{cat} = 0.5$  g,  $WHSV = 1.23$  g<sub>DME</sub> g<sub>cat</sub><sup>-1</sup>h<sup>-1</sup>.