## Supporting Information



Figure S1. XPS spectra of Ni-MCM-41 (a) Calcined, (b) after reduction and (c) during the reaction in $\mathrm{scCO}_{2}$


Figure S2. FTIR spectra of Ni-MCM-41 (a) Calcined, and (b) during reaction in $\mathrm{scCO}_{2}$

## S3. Computational methods

The calculations were performed using the $\mathrm{DMOl}^{3}$ program [1] of Accelrys Inc. The geometry of the all the interacting molecules in all the cases were optimized using the double numerical with polarization basis set DNP [2] which is equivalent to the $6-311 \mathrm{G}^{* *}$ basis set with BLYP as the exchange correlation functional $[3,4]$. Transition state calculations were performed using a LST/QST transition search methodology [5]. Halgren and

Lipscomb introduced the linear synchronous transit (LST) and quadratic synchronous transit (QST) methods for searching for the transition state of molecules as incorporated in the Dmol3 program of Accelrys Inc. Calculating analytical gradients and numerical Hessians optimized all geometries including those of the TS. Hessian Eigen values have either all positive values (ground state) or only one negative value (transition state). We also confirmed that the ground state geometries had all positive vibrational frequencies and that the transition-state geometries had a one and only one imaginary frequency.

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