Supporting Information

N-Doped Carbon as a Solid Base Catalyst for Continuous Flow Knoevenagel Condensation

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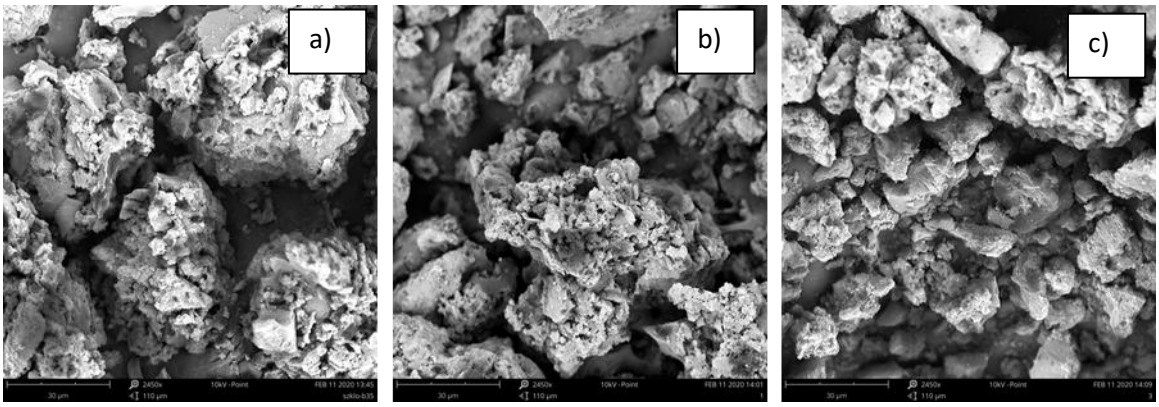
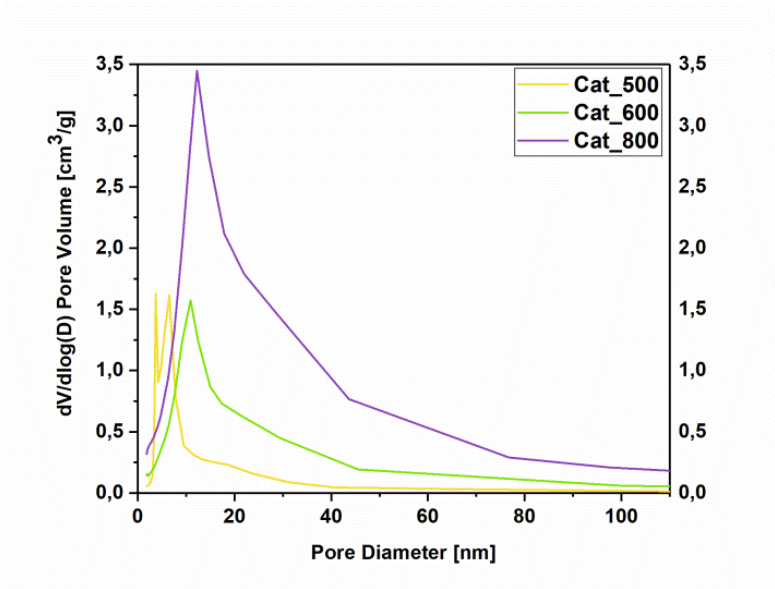


S1. SEM images of a) Cat_500, b) Cat_600, c) Cat_800.



S2. Pore-size distribution for Cat_500, Cat_600 and Cat_800.

S3. Determination of the exact volume of the reactor for Knoevenagel condensation in presence of Cat_500:

The exact volume of the reactor was calculated from the difference between the volume of the empty reactor and the volume of catalyst. For the volume determination, the Omnifit column (6.6 x 100mm) with a filter at the column ends was densely packed with 0.250 g of Cat_500 (additionally separated with cotton wool) and weighed. Ethanol was pumped through the reactor for 30 min to ensure air removal from the catalyst pores. The amount of solvent

was made level with the front of the catalytic bed and the reactor was weighed once again. Using a solvent with a known density, the exact volume of the reactor was determined to be 0.67 mL.

M_0= mass of rector with catalyst = 30.71g

M_z= mass of rector with catalyst and ethanol=31.24 g

$$M_{EtOH}=M_z-M_0= 0.53 \text{ g}$$

$$V=m/d_{EtOH}= 0.53/ 0.79 = 0.67 \text{ ml}$$

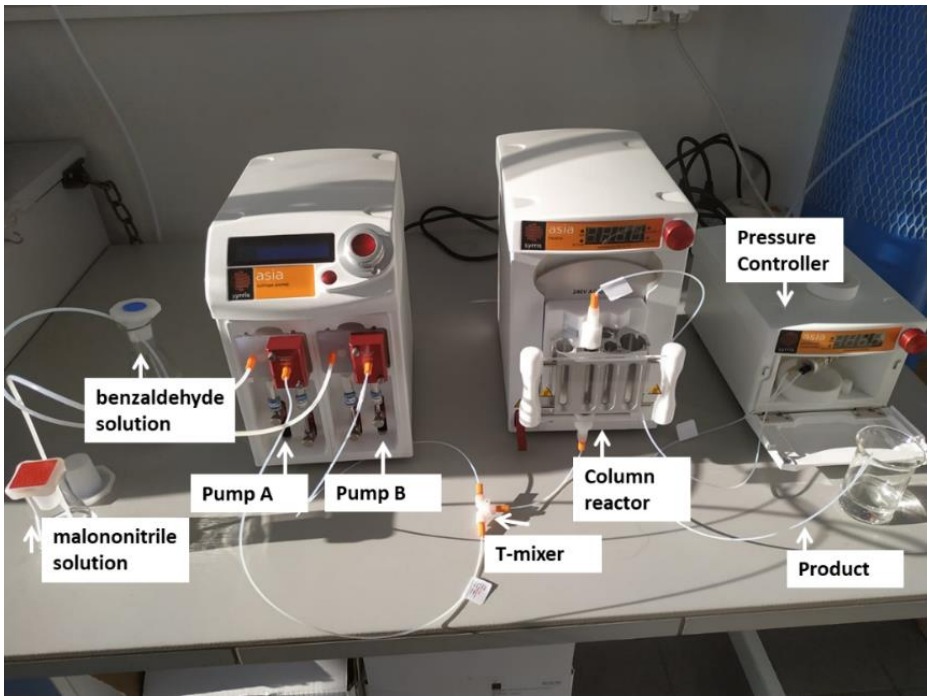
S4. Explanation for calculation of flow reaction rates.

The rates of benzaldehyde conversion (*r*) were calculated according to the following equations:

$$r(mmol\ min^{-1}\ g^{-1}) = \frac{c \cdot f}{m_{cat}}$$

where c refers to the concentration of benzaldehyde in mmol·cm^{-3} in the withdrawn sample, m_{cat} is catalyst mass in g and f is the molar flow in cm^3·min^{-1}.

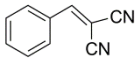
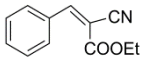
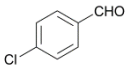
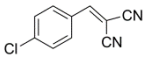
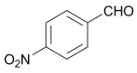
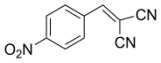
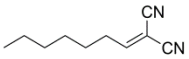
S5. Continuous flow reaction set-up for Knoevenagel condensation: Syrris Asia flow system equipped with two pumps.



S6. GC analysis

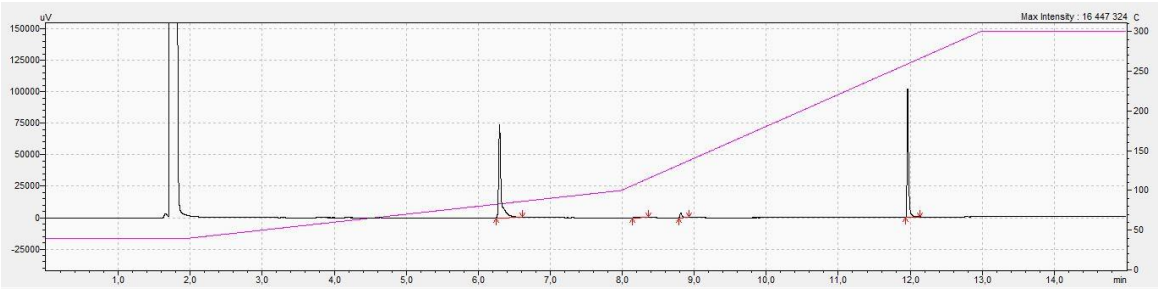
Gas Chromatography analysis was performed using SHIMADZU 2010 chromatograph equipped with ZB-50 (30 m × 0.25 mm × 0.25 µm) column. The parameters set during analysis: an injection temperature of 250 °C, a FID detector temperature of 300 °C, the initial temperature of 40 °C (hold for 2 min), with a rate of increasing temperature 20°C/min and the final temperature of 280 °C (hold for 10 min), linear flow set to 35 mL/min, a split ratio of 100:1, and a run time of 18 min. The conversion (α) was calculated from the peak area ratio. The chromatograms with retention times of corresponding aldehydes and products are provided in S7.

S7. Retention times of investigated aldehydes and products on GC analysis.

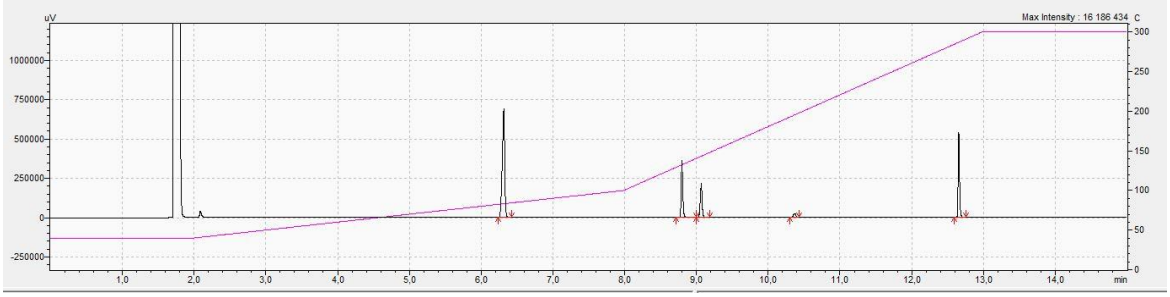
Aldehyde	Product	Retention times, min (I aldehyde; II product)
	a 	(I) 8.8 (II) 12.0
	b 	(I) 8.8 (II) 12.7
	c 	(I) 10.3 (II) 12.6
	d 	(I) 11.7 (II) 13.5
	e 	(I) 6.7 (II) 11.0

Retention time of decane: 6.300

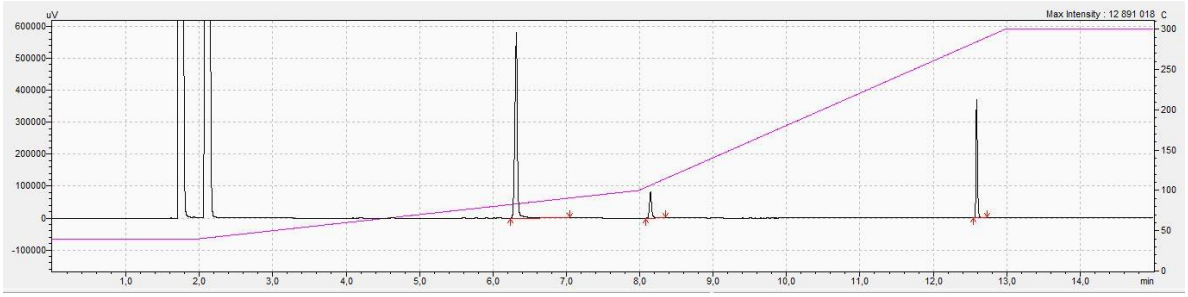
a)



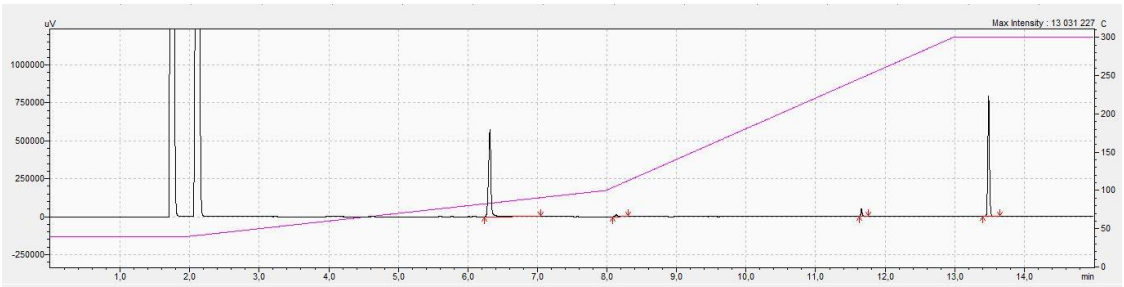
b)



c)

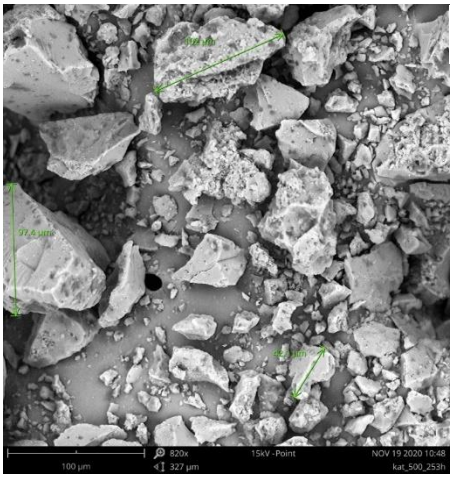


d)

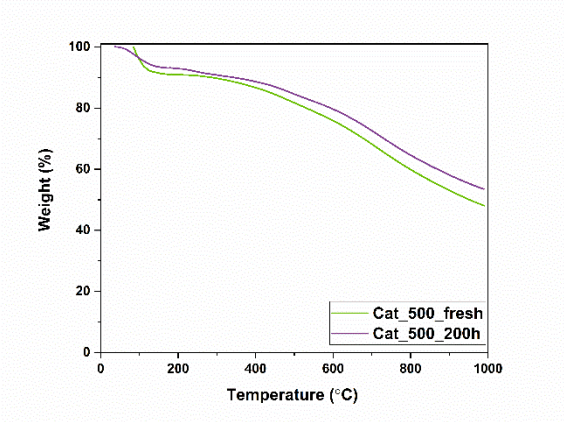


S8. a) SEM image of Cat_500 after 200 h performance under continuous flow conditions; b) Thermogravimetric analysis of fresh Cat_500 and Cat_500 after 200 h performance under continuous flow conditions.

a)



b)



S9. Percentage contribution of different N species on the Cat_500 surface after 200 h performance under continuous flow conditions obtained from XPS analysis.

	398 eV (%)	399 eV (%)	401 eV (%)	404 eV (%)

Cat_500_200h	32.64	35.39	27.83	4.14

S10. TOF determination

The number of accessible basic sites was determined with acid–base titration using PH/ION METER S220-UMIX KIT SEVENCOMP. TOF was calculated according to the following equation:

$$TOF = \frac{c_{product}F}{n_{cat}}$$

where $c_{product}$ refers to the concentration of product in mmol $\cdot L^{-1}$, F is the flow rate in L $\cdot h^{-1}$ and n_{cat} is is the amount of catalyst inside the reactor column.