Supplementary Materials

Table S1. Composition of hybrid zeolites

	Phase ratio	Si/Al, mol/mol
MFI-MEL	55 (MFI):45 (MEL)	53
MFI-MTW	60 (MFI):40 (MTW)	53
MFI-MCM-41	80 (MFI):20 (MCM-41)	55
MFI	100 (MFI)	48

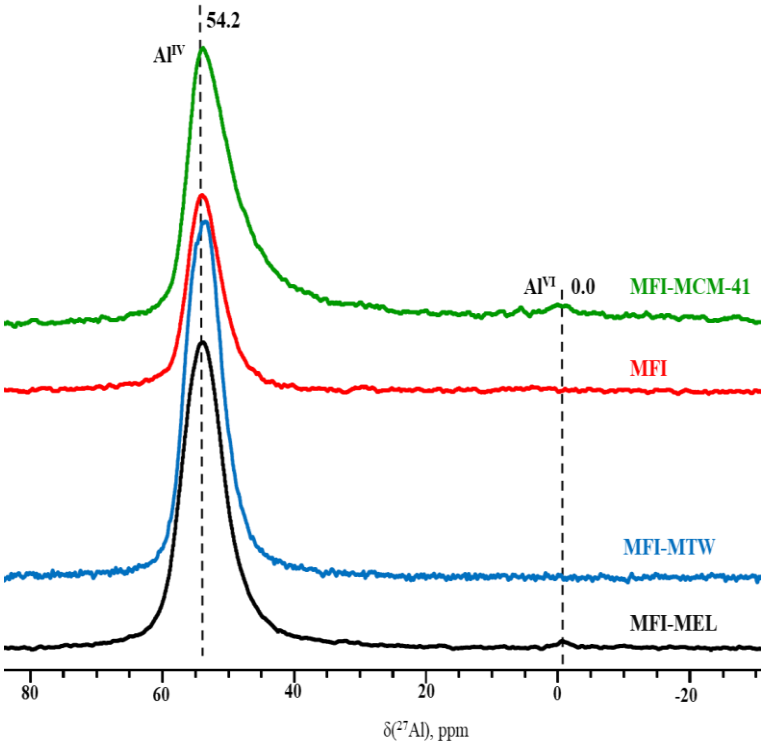

Figure S1. ^{27}Al MAS NMR spectra of hybrid zeolites.

Figure S1: An intense peak with chemical shifts at ~54 ppm. corresponds to the 4-coordinated Al atoms in the zeolite framework. The spectra of the MEL-MFI and MFI-MCM-41 have a weak peak with chemical shifts at ~0 ppm. It corresponds to the octahedral Al atoms, which are not incorporated in the zeolite framework.

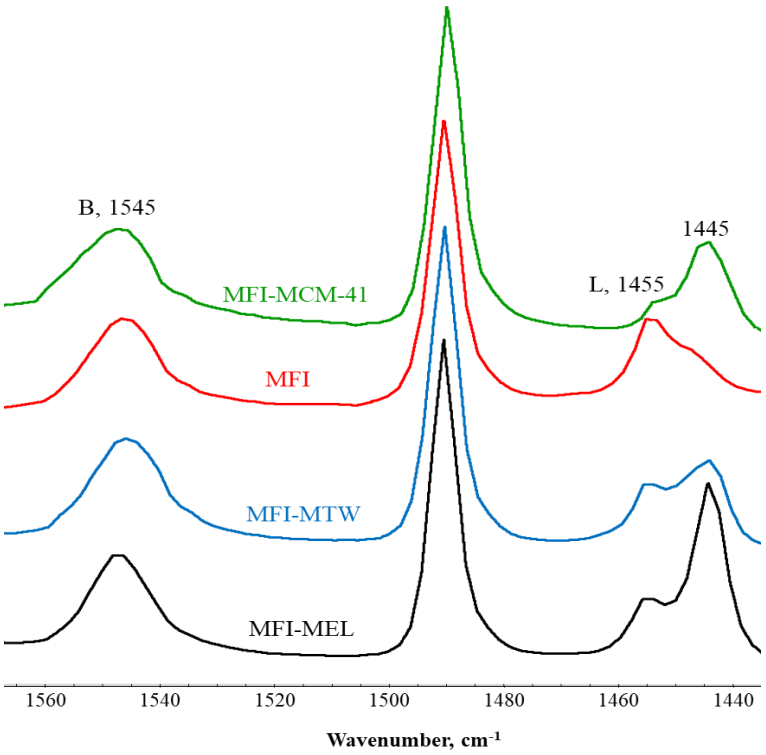

Figure S2. IR spectra of pyridine adsorbed on hybrid zeolites.

Figure S2: For all samples in the IR spectra of absorbed pyridine, the peaks at 1455 cm^{-1} and 1545 cm^{-1} indicate the adsorption of pyridine on strong Lewis and Brønsted acid sites, respectively. The peak at 1445 cm^{-1} characterizes physically adsorbed pyridine.

Table S2. Acid characteristics of hybrid zeolites.

	Number of acid sites, μmol Py/g			*B/L*
	BASs	*LASs*	*Total*	
MFI-MEL	90	25	115	3.6
MFI-MTW	110	27	137	4.1
MFI-MCM-41	85	20	105	4.3
MFI	80	35	115	2.3

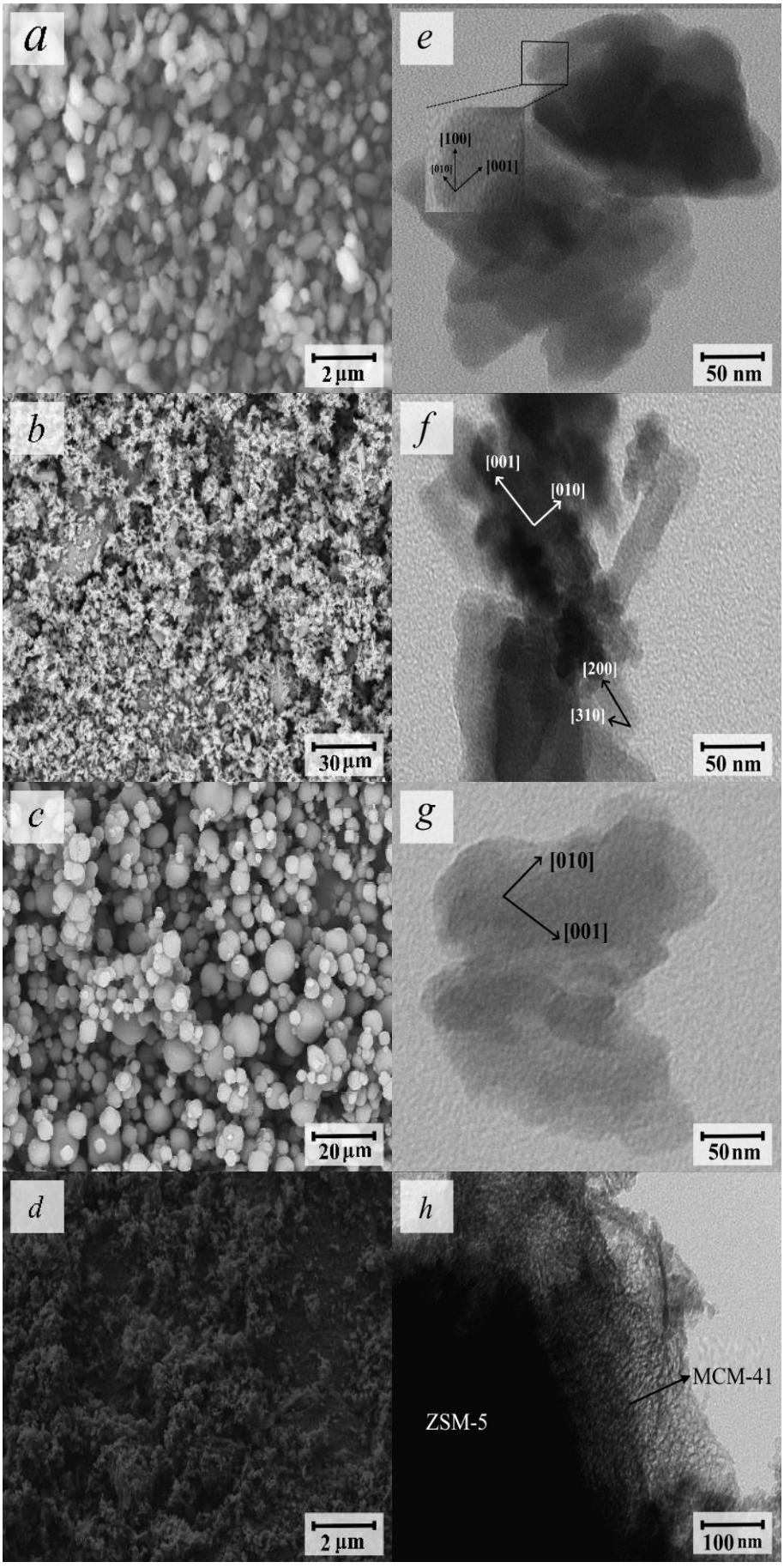

Figure S3. The micrographs of **(*a-d*)** SEM- and **(*e-h*)** TEM-hybrid zeolites: MFI-MEL **(*a,e*)**; MFI-MTW**(*b,f*)**; MFI **(*c,g*)**; MFI-MCM-41 **(*d,h*)**. Crystal planes of MFI: [010] and [001]; MEL: [010] and [100]; MTW: [200] and [310].

Figure S3: According to scanning and transmission electron microscopy ***(a,e)***, MEL-MFI co-crystallites are elongated oval crystals with an average size of 7–9 μm, which consist of agglomerated nanocrystallites. It indicates the cocrystallization nature of the material. The MFI-MTW is represented by polycrystalline agglomerates with a size of 1-5 μm, which consist of crystals with a hexagonal structure ***(b,f)***. The structure is characteristic of both MFI and MTW type zeolites. MFI structure zeolite particles have a spherical shape with an average size of 4–5 μm **(*c,g*)**. The MFI-MCM-41 composite is presented in the form of particles with a size of 1–2 μm. According to the TEM, MFI-MCM-41 is a "core-shell" composite material **(*d,h*)**.

Table S3. Additional textural properties of the catalysts.

	S_{BET}, $m^2/g(cat)$	S_{micro}, $m^2/g(cat)$	S_{meso}, $m^2/g(cat)$	$S_{external}$, $m^2/g(cat)$	d_{micro}[1], nm	d_{meso}[2], nm
MFI-MEL/Al_2O_3	361	250	95	15	0.91	5.79
MFI-MTW/Al_2O_3	179	11	101	67	0.69	19.67
MFI-MCM-41/Al_2O_3	250	42	185	24	0.63	7.58
MFI/Al_2O_3	293	181	96	14	0.90	8.27

[1] average micropore diameter (MP-plot)
[2] average mesopore diameter (BJH method)

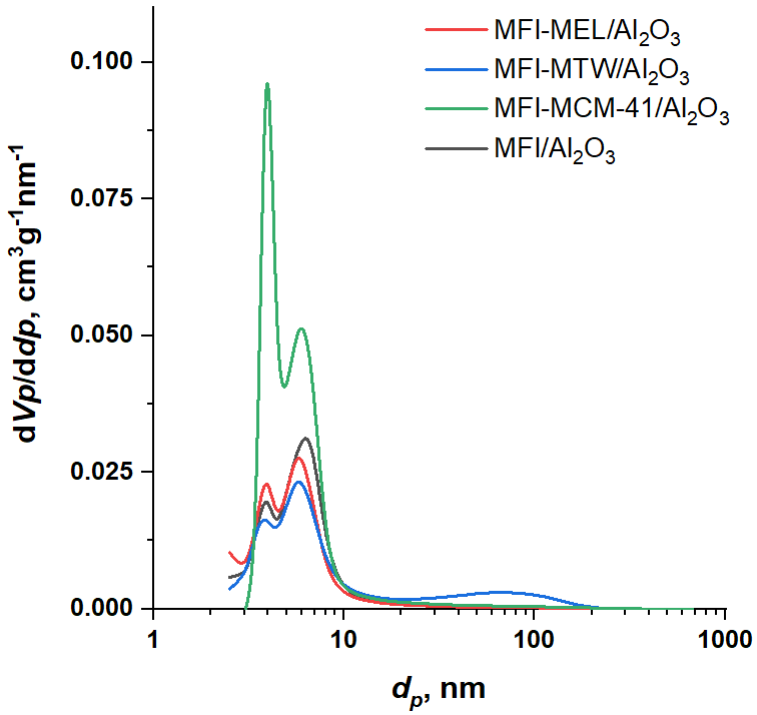

Figure S4. BJH plots (desorption curve) for the catalysts MFI-MEL/Al_2O_3, MFI-MTW/Al_2O_3, MFI-MCM-41/Al_2O_3, and MFI/Al_2O_3.

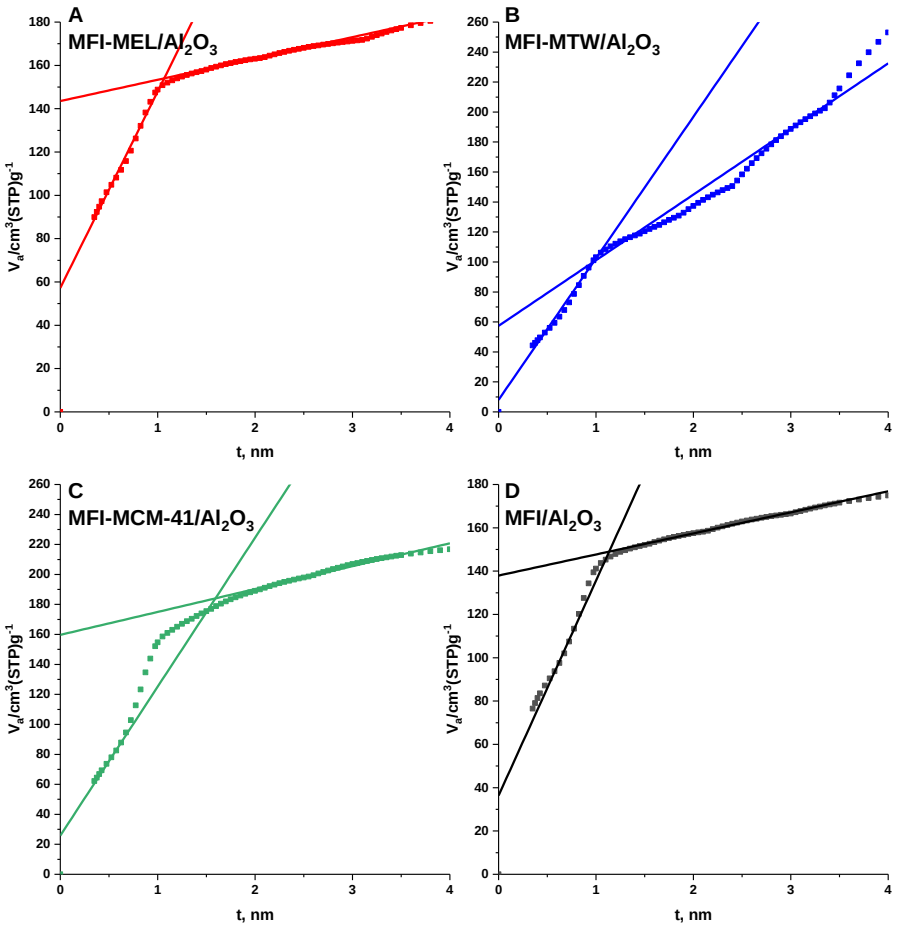

Figure S5. t-plots for nitrogen adsorbed in the catalysts MFI-MEL/Al_2O_3 **(A)**, MFI-MTW/Al_2O_3 **(B)**, MFI-MCM-41/Al_2O_3 **(C)**, and MFI/Al_2O_3 **(D)**.