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ARTICLE TYPE

Core-Shell H-ZSM-5/Silicalite-1 Composites: Brønsted Acidity and Catalyst Deactivation at the Individual Particle Level

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Supplementary Information

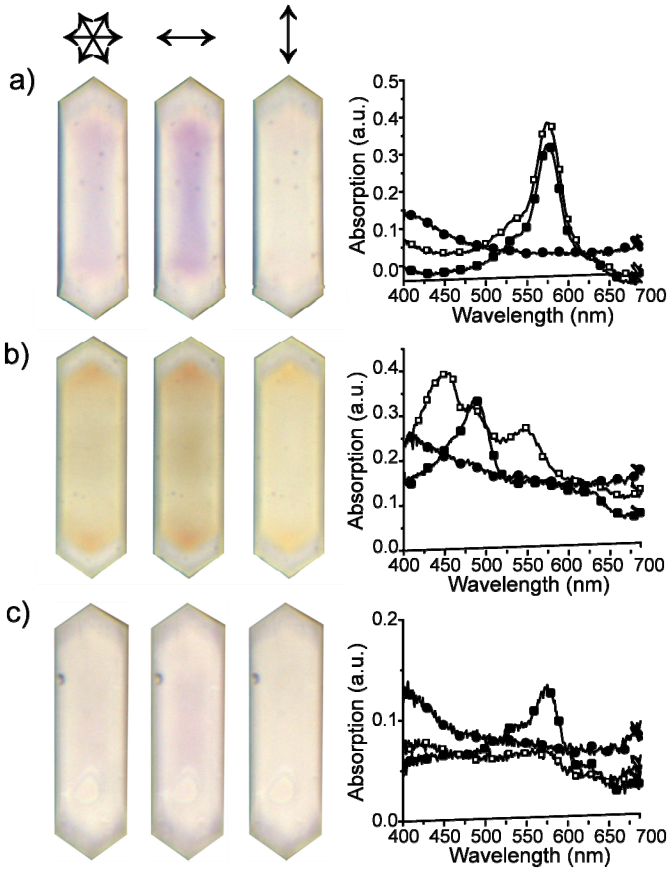

Fig. S-1 Optical microphotographs and the corresponding UV-Vis absorption spectra of the H-ZSM-5/silicalite-1 core/shell crystals after oligomerization at 373 K with a) 2-chlorostyrene, b) 3-chlorostyrene and c) 3,4-dichlorostyrene. The images were taken with and without the use of polarized light. The polarization directions are indicated with the arrows and the represented absorption spectra are measured with 0 degree polarization direction (↔) at the H-ZSM-5 core (-■-), the H-ZSM-5/silicalite-1 boundary (- -) and the silicalite-1 external shell (-●-) of the zeolite composite.

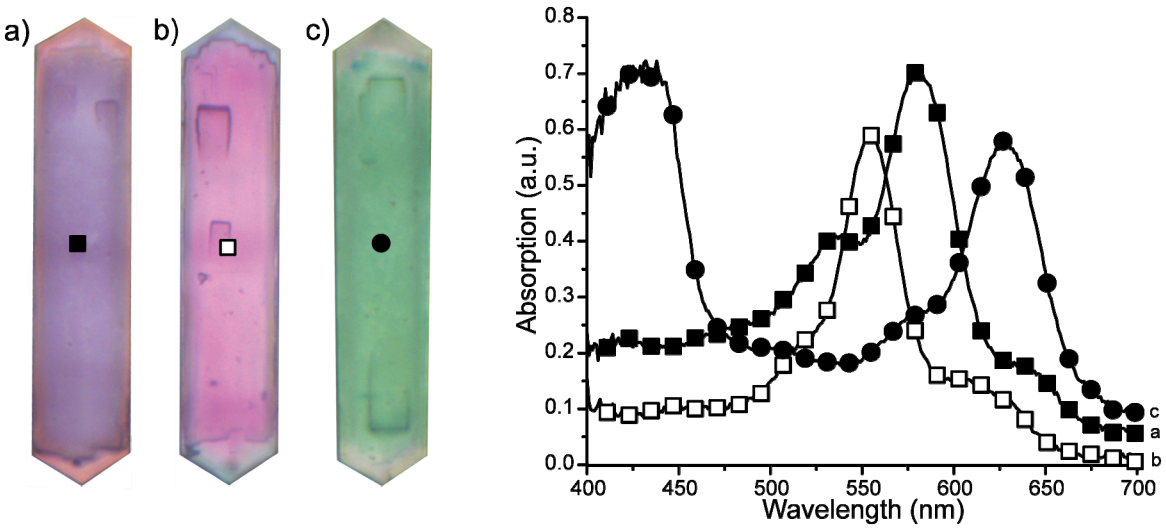

Fig. S-2 Optical microphotographs taken after oligomerization of H-ZSM-5 crystals lacking an external silicalite-1 layer at 373 K with a) 4-chlorostyrene, b) 4-fluorostyrene and c) 4-methoxystyrene. The images are taken with the 0 degree (↔) light polarization. The corresponding UV-Vis absorption spectra are measured from a 2 µm spot in the middle of the zeolite crystal.

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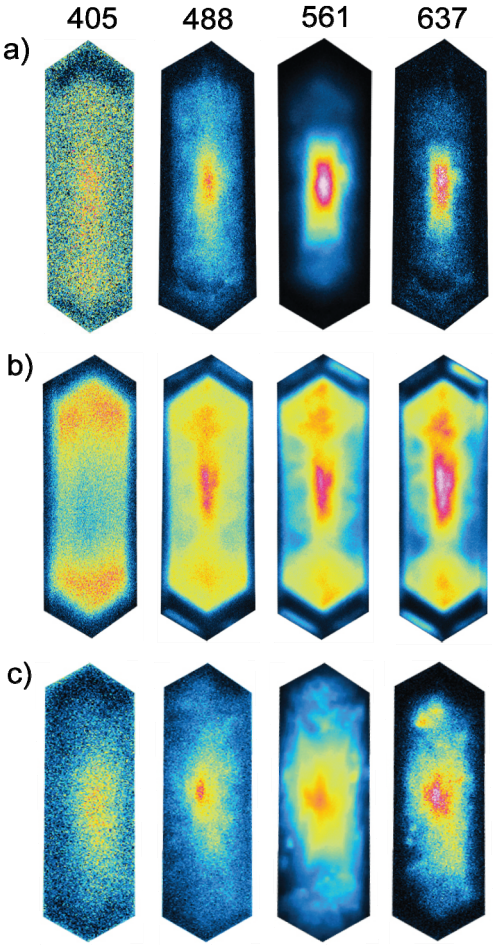

Fig. S-3 Confocal fluorescence microphotographs of the core-shell H-ZSM-5/silicalite-1 composites after oligomerization at 373 K with a) 2-chlorostyrene, b) 3-chlorostyrene and c) 3,4-chlorostyrene. The corresponding laser excitation wavelength is indicated in nm**.**

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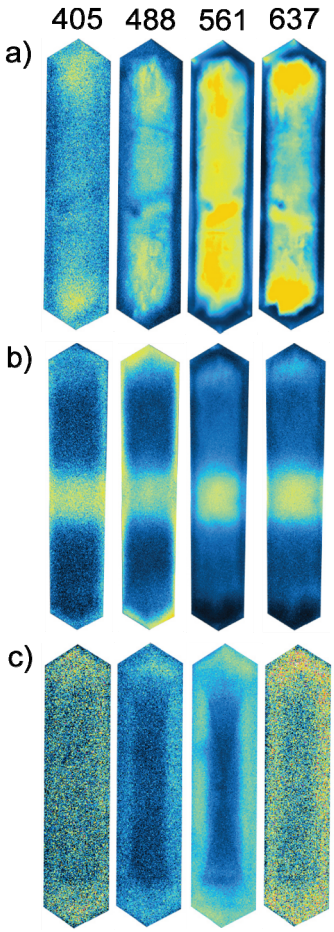

Fig. S-4 Confocal fluorescence microphotographs of H-ZSM-5 crystals lacking an external silicalite-1 layer after oligomerization at 373 K with a) 4-chlorostyrene, b) 4-fluorostyrene and c) 4-methoxystyrene. The corresponding laser excitation wavelength is indicated in nm**.**

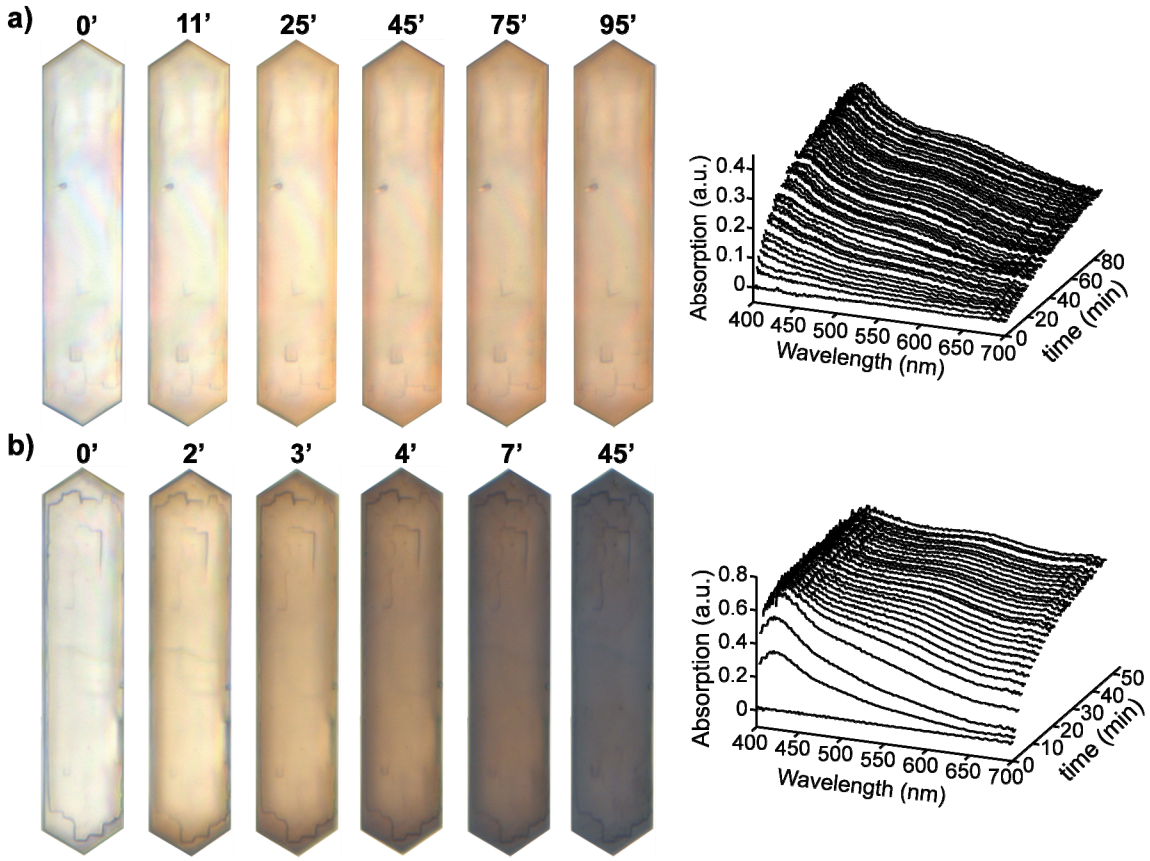

Fig. S-5 Selection of the optical microphotographs taken during the methanol-to-olefin conversion at 573 K (a) and at 773 K (b) on a H-ZSM-5 crystal without external silicalite-1 coating. The corresponding UV-Vis absorption is taken from a 2 μm spot in the middle of the zeolite crystal. (From D. Mores et al, *Chem. Eur. J.,* 2008, **14**, 11320-11327).

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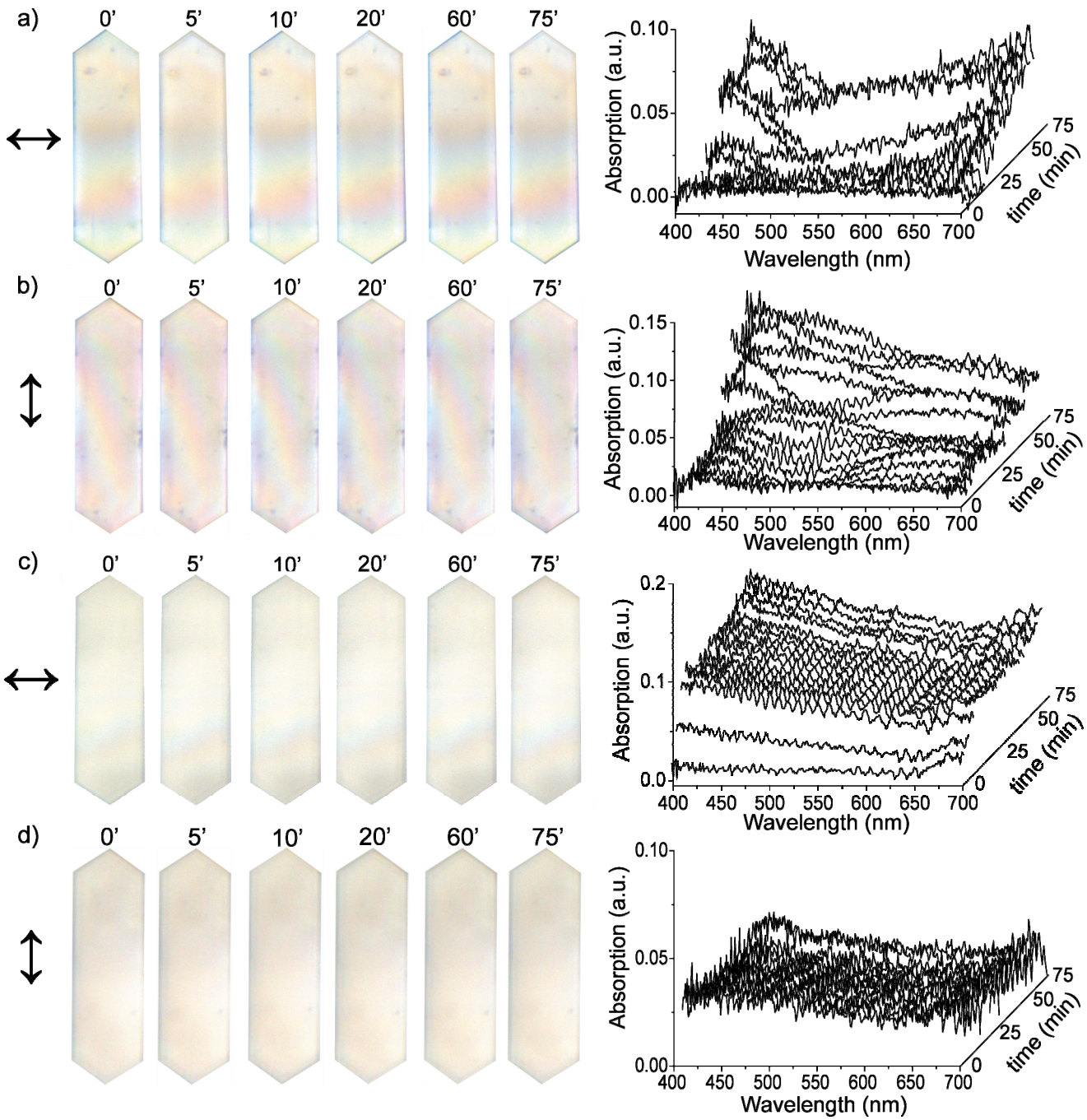

Fig. S-6 Selection of polarization dependent optical microphotographs of the H-ZSM-5/silicalite-1 zeolite crystals taken during the aromatization process at 773 K with pentane (a, b), and 2-methylpentane (c, d). The corresponding time-resolved spectra are taken from a spot in the middle of the zeolite, the polarization direction is indicated with the arrows and the time is indicated in min.

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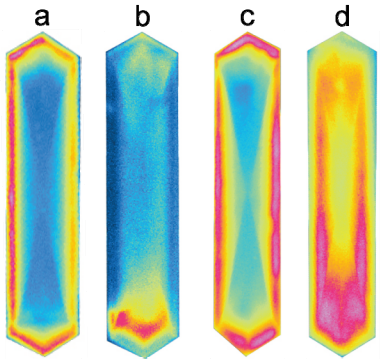

Fig. S-7 Confocal fluorescence images of H-ZSM-5 without external silicalite-1 shell after aromatization with a) pentane, b) 1-pentene, c) 2-methylpentane and d) 2-methyl-2-pentene. The images are taken from the middle plane of the zeolite with 561 nm excitation wavelength. (From Y.M. Chung et al., *Appl. Catal. A*, revised submission)

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