Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Core-shell Zeolite Y with Ant-nest like Hollow Interior Constructed by Amino Acids and Enhanced Catalytic Activity

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Supplementary Methods

Chemicals: All reagents were of analytical grade and used as purchased without further purification. L-carnitine (LC), L-lysine (lys), toluene, benzyl chloride, inorganic salts of sodium hydroxide and sodium aluminate (38%Na_2O, 50%Al_2O_3) were purchased from Adamas-beta, benzyltoluene (*m*-, *o*-, *p*-) from J&K® and Ludox (25% aqueous solution) from Qingdao Ocean Co., Ltd.

Electron tomography: Electron tomography (ET, 3D-TEM) were performed in bright-field mode with a FEI Tecnai G2 F30 electron microscope. The microscope was equipped with a TWIN objective lens, a LaB_6 electron source and was operated at 300 kV. TEM grids were prepared by dropwise addition of homogenous suspension of zeolite in ethanol, followed by drying in air at 60 °C. These grids were initially labelled with 5 nm gold particles by applying a droplet of colloidal gold suspension on a carbon-coated Quantifoil R2/1 copper grid with parallel bars in order to provide markers for alignment of tilt images. Tilt series of images were acquired using Xplore3D software over an angular range of ±70 ° with a tilt increment of 1 ° or 5 °. Images were recorded on a 2048 x 2048 pixel TVIPS CCD camera. Alignment of the acquired tilt images was performed using the inspect3D software and facilitated by tracking the gold markers. 3D reconstruction of the aligned images was done by filtered back projection using the same software. 3D visualization and analysis were performed by Amira software to better explore the pore channel structure.

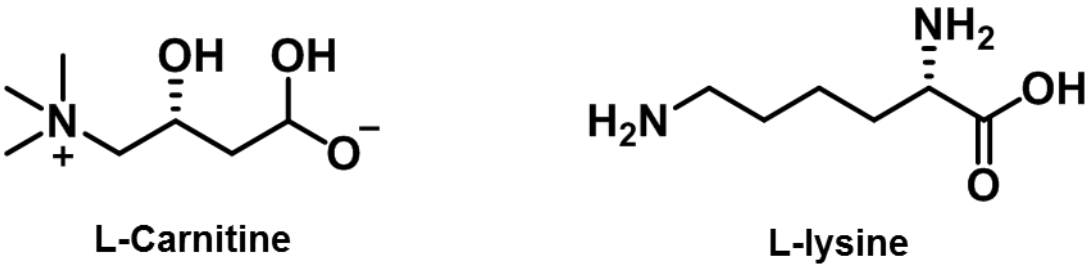


Figure S1. Chemical structural of the amino acids used in this study.

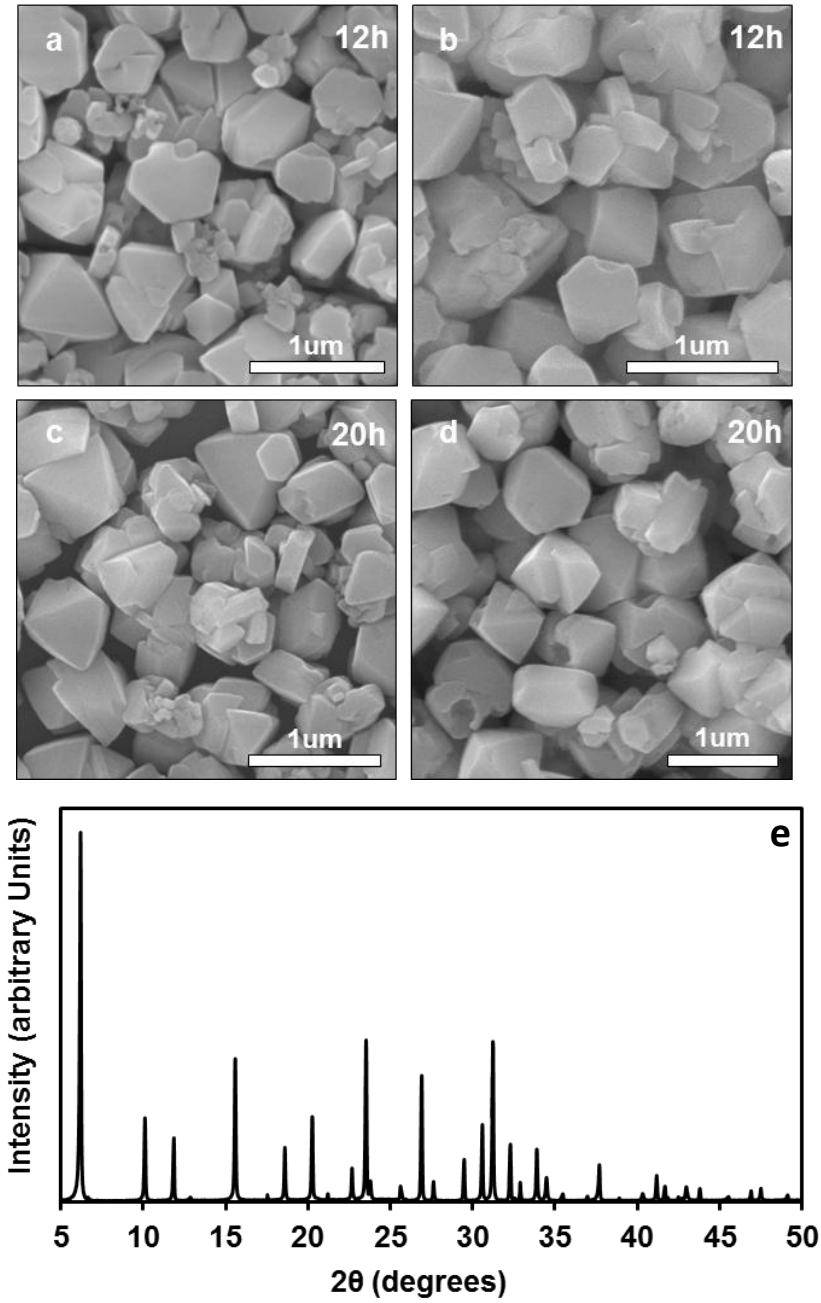


Figure S2. SEM images of conventional zeolite Y synthesized without amino acids at (a, b) 12 h and (c, d) 20 h. (e) XRD pattern of conventional zeolite Y synthesized without amino acids at 20 h.

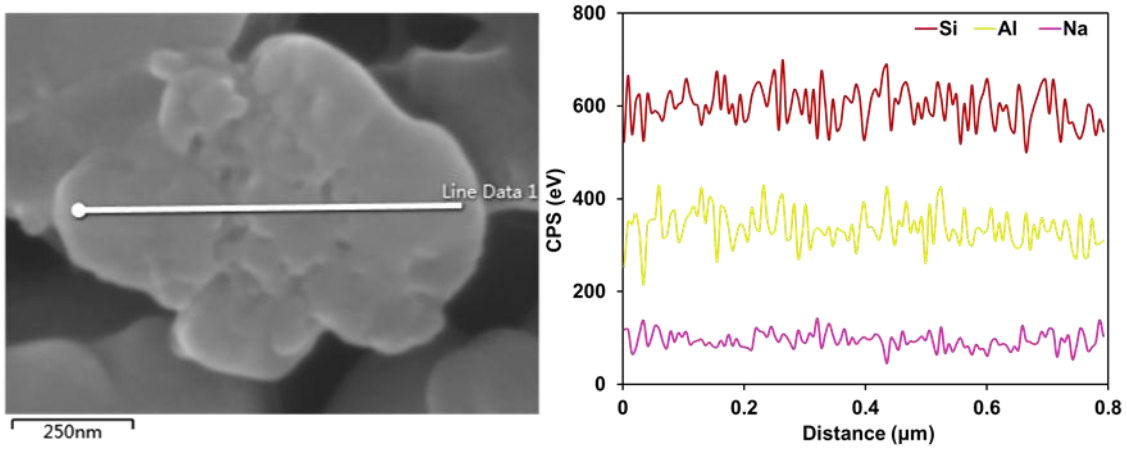


Figure S3. EDS cross-sectional scan of a single core-shelled LC-Y-20 particle showing homogenous distribution of the zeolite framework elements of Si (red line), Al (yellow line), and extra frame work Na content (purple line) on both the core and shell of the particle.

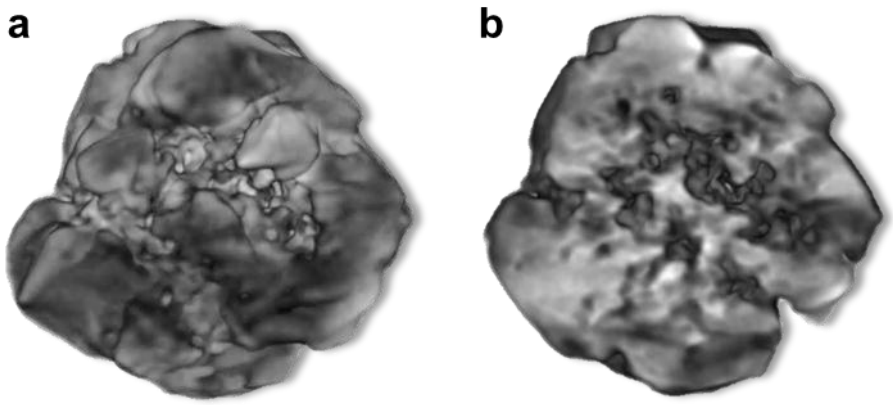


Figure S4. (a) Total volume 3D representation and (b) selected volume of the ET reconstruction with half of the crystal clipped along *xz* plane for a surface broken core-shell zeolite Y templated by L-carnitine. Animated versions of the tomograms are provided in the Supporting Information as videos.

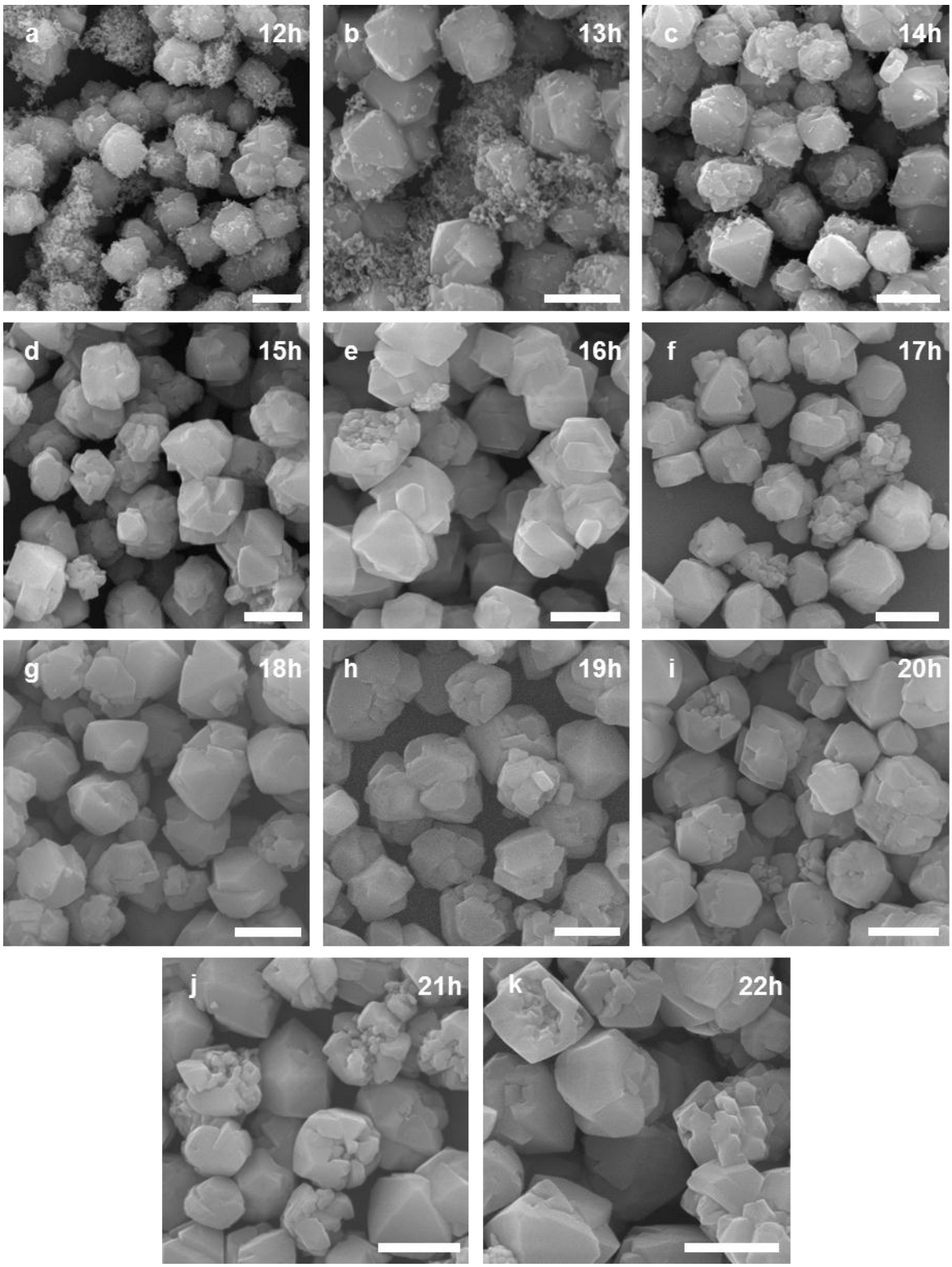
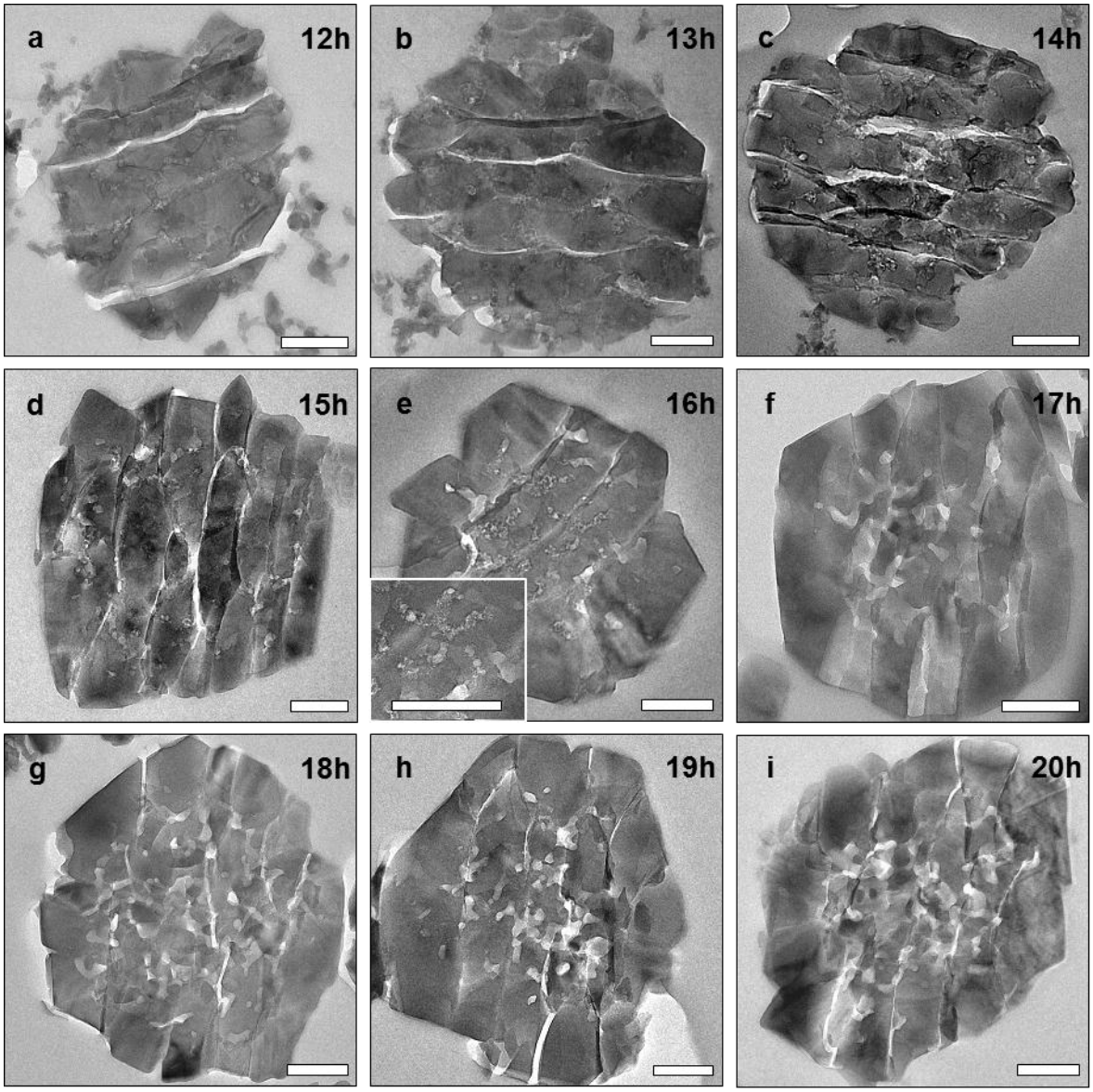


Figure S5. Time-series SEM images of the core-shell zeolite Y synthesized with L-carnitine at different hydrothermal synthesis stages from 12 h to 22 h at 1 h interval, revealing similar crystal morphology. Scale bar represents 1 μm.



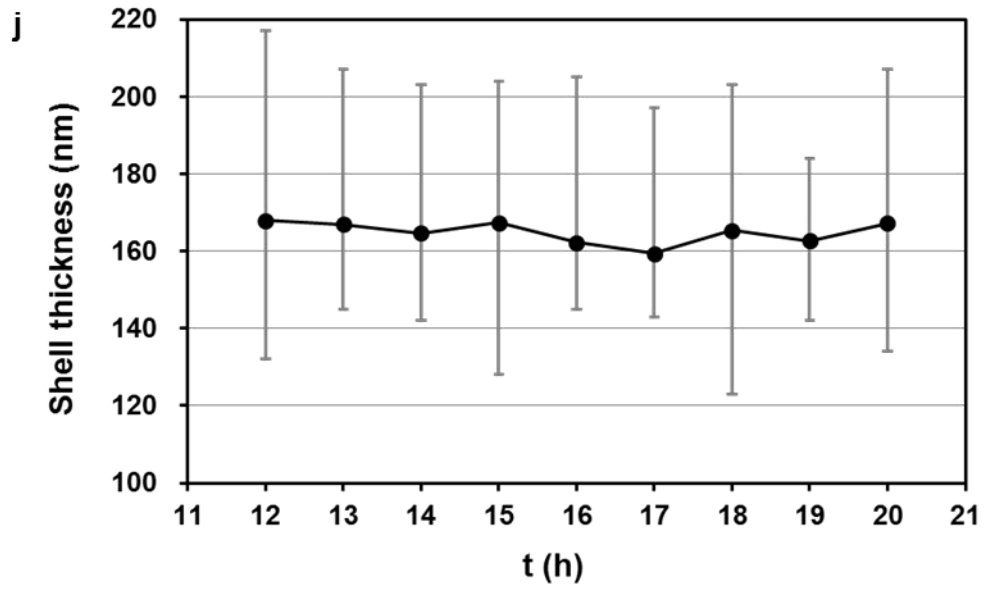


Figure S6. (a-i) Time-series cross sectional TEM images of the core-shell zeolite Y synthesized with L-carnitine at different hydrothermal synthesis stages from 12 h to 20 h at 1 h interval, revealing the

gradual development of internal mesopore channels. Scale bar represents 200 nm. (j) Measured shell thickness at different hydrothermal synthesis stages showing constant values of ~165 nm.

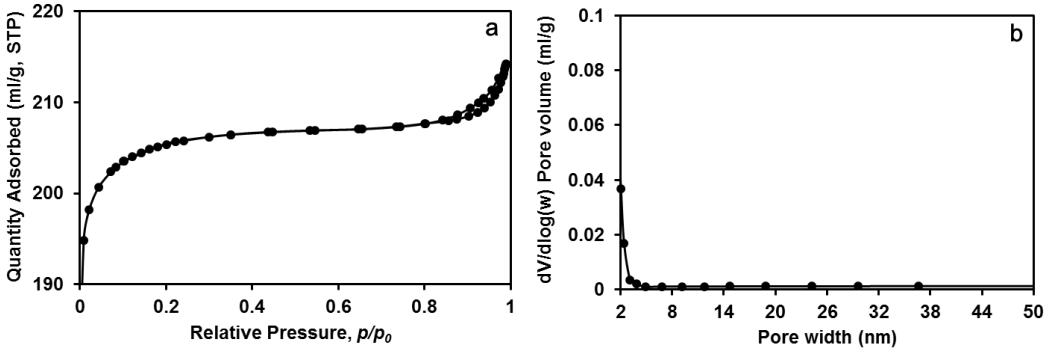
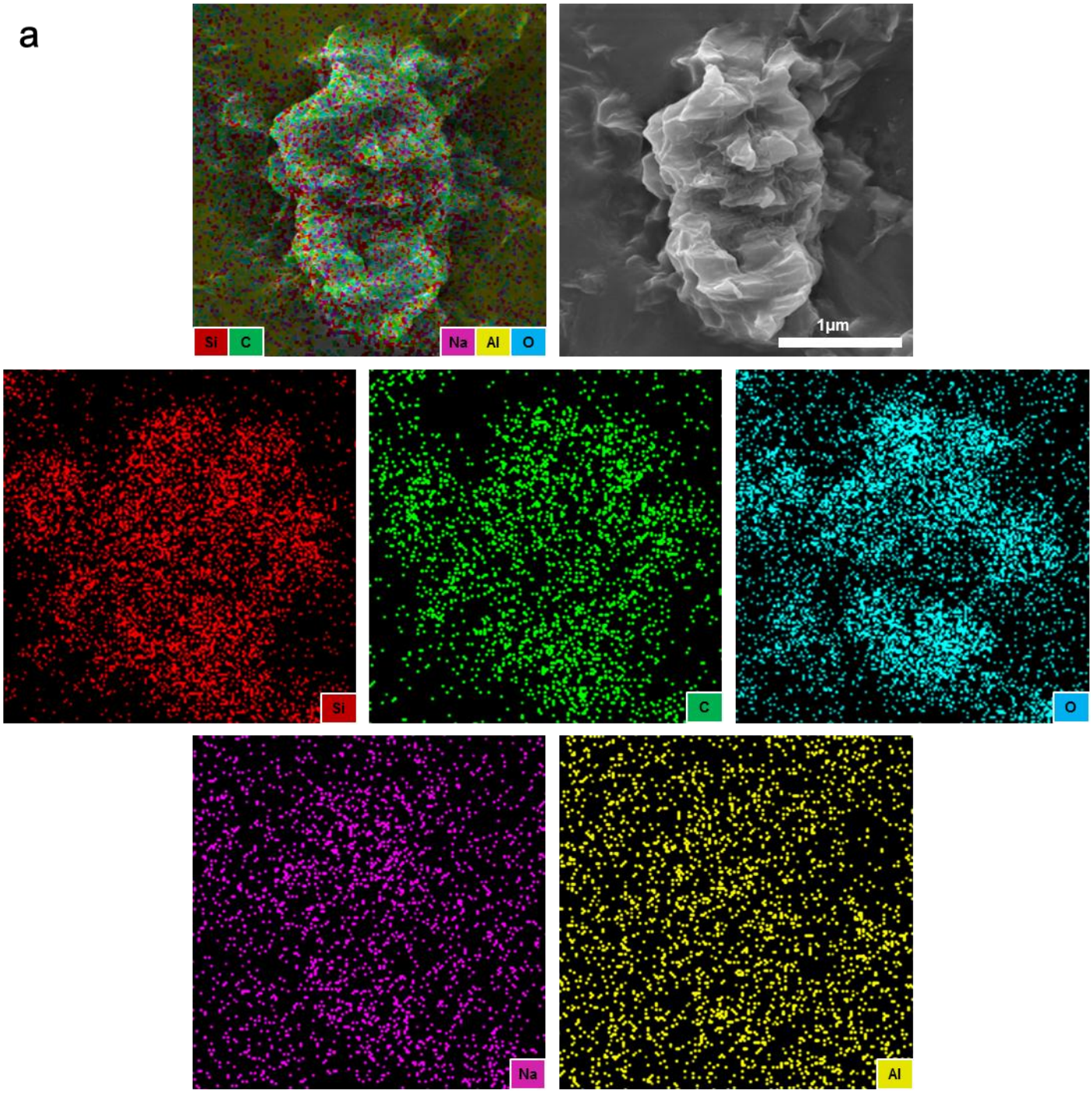
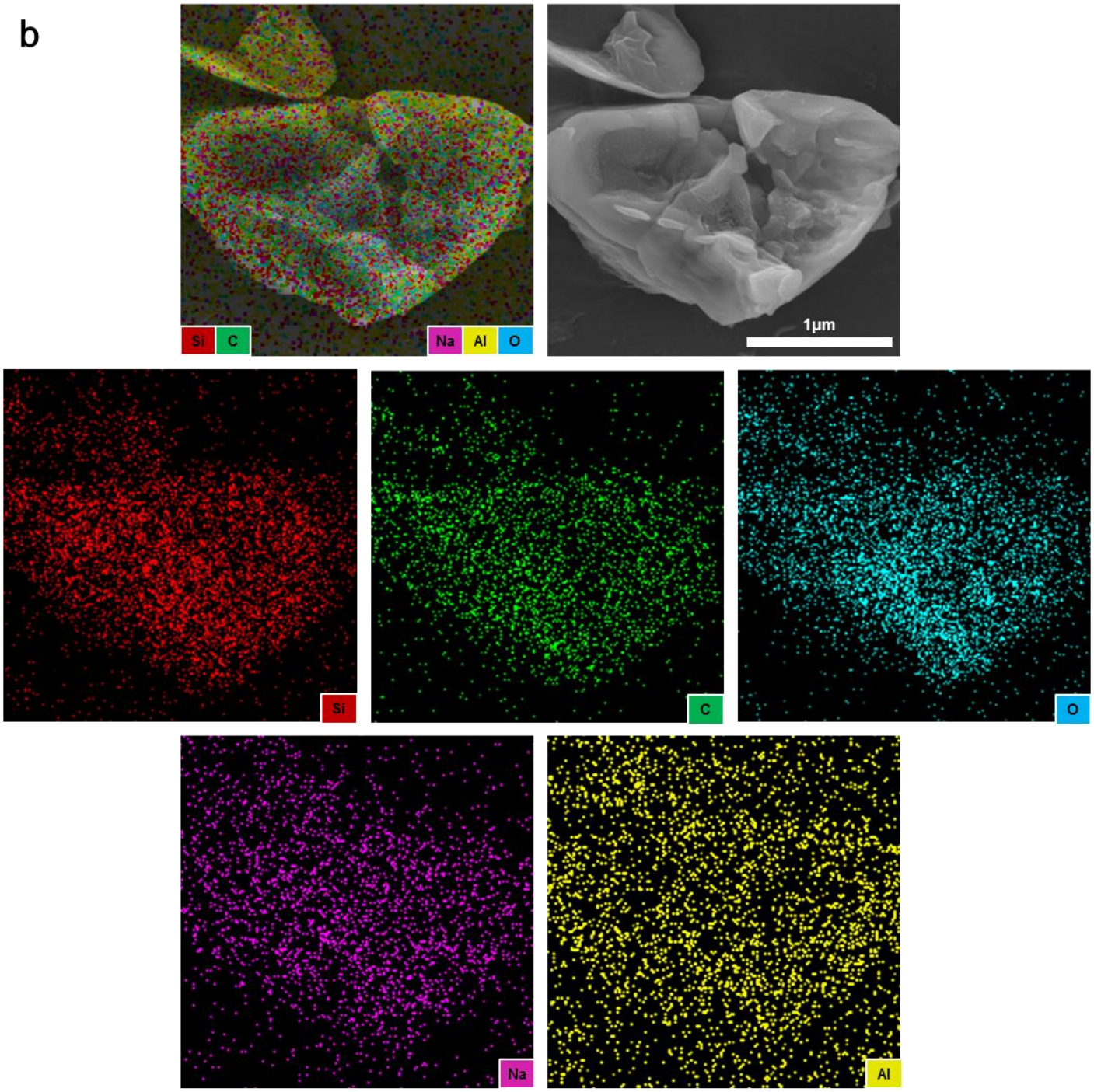


Figure S7. (a) N_2 adsorption–desorption isotherms and (b) BJH mesopore size distributions of conventional zeolite Y-20 corresponding to the desorption branch.

a



b



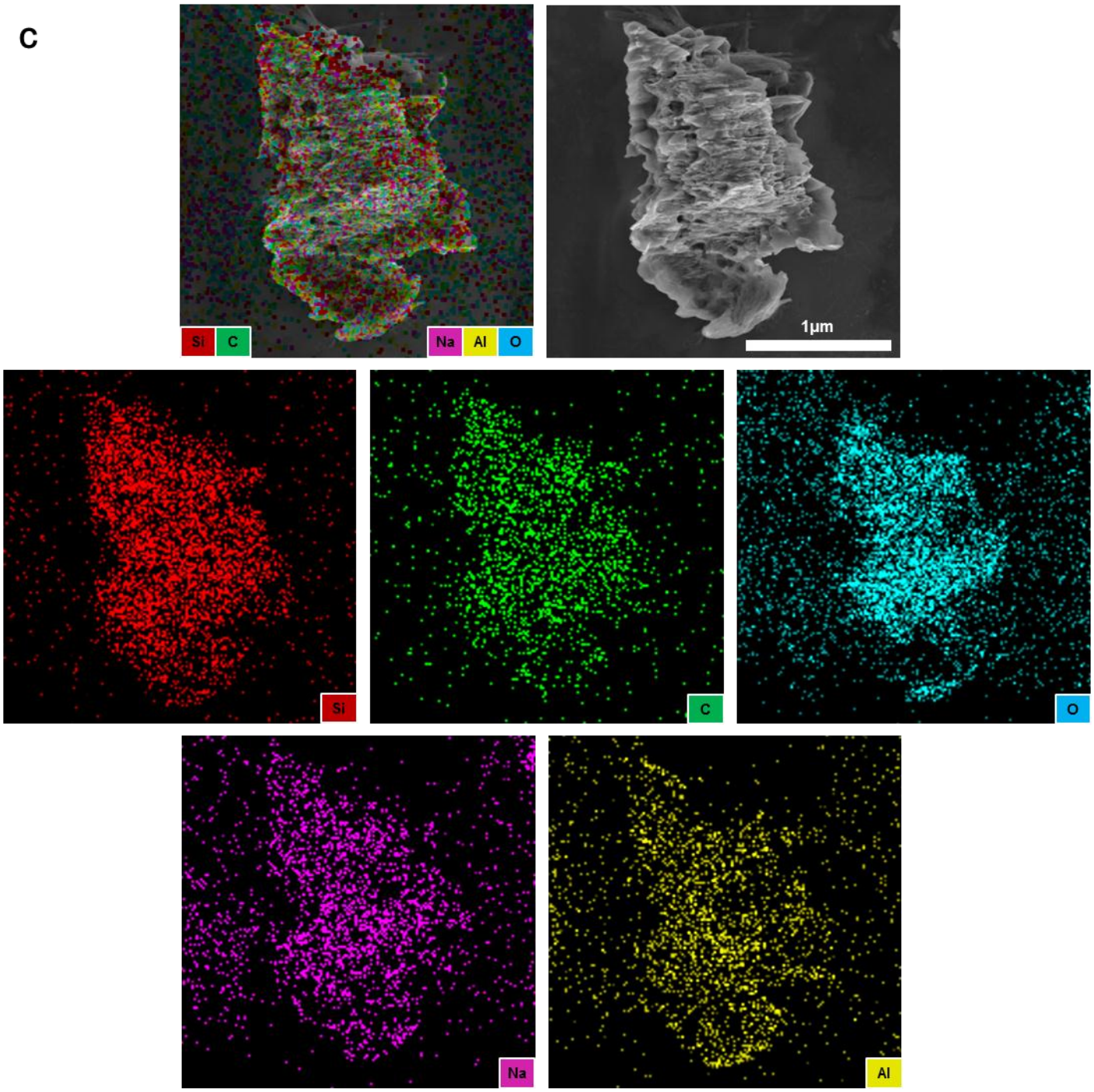


Figure S8. Time-series SEM-EDS map of a focused ion beam eroded core-shell zeolite LC-Y samples before washing with water, showing the location of silicon (Si, red), carbon (C, green), oxygen (O, blue), sodium (Na, purple), and aluminium (Al, yellow). (a) LC-Y-12; (b) LC-Y-14; (c) LC-Y-20.

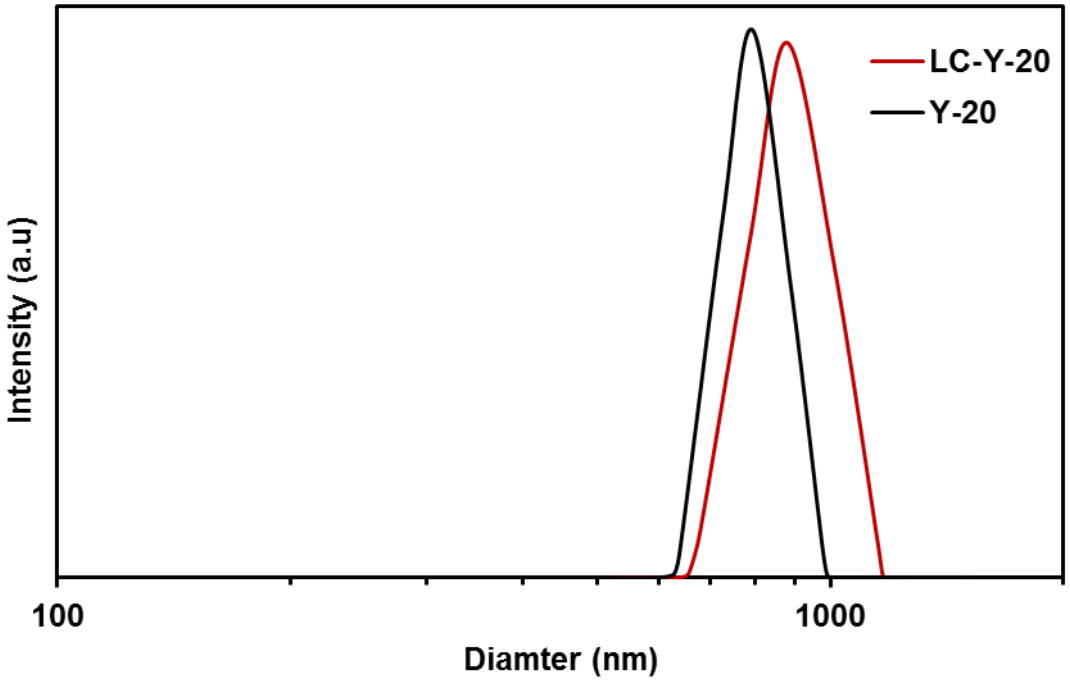


Figure S9. DLS curves of the core-shell zeolite Y (LC-Y-20) and conventional zeolite Y (Y-20).

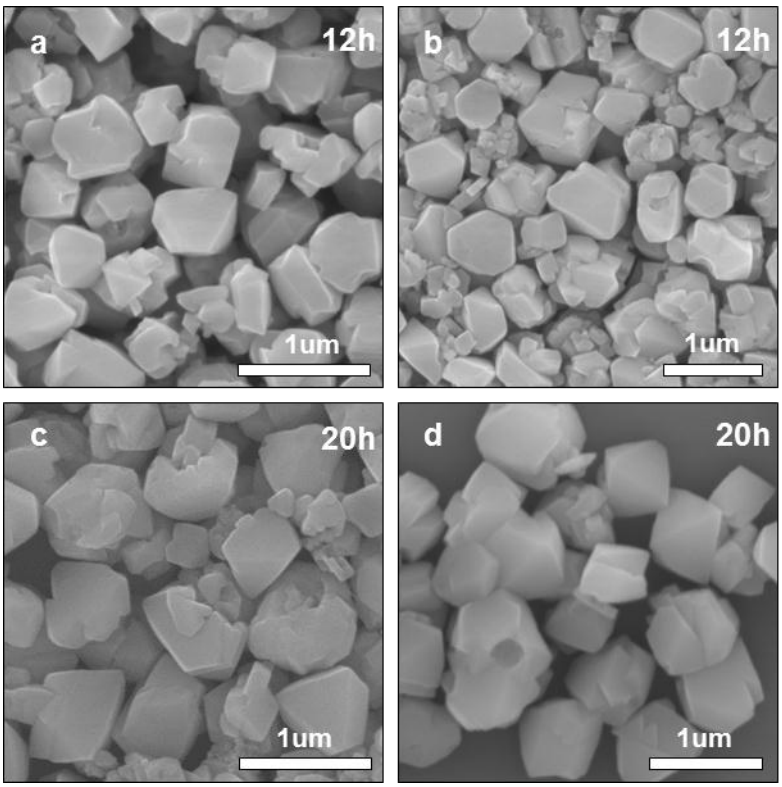


Figure S10. SEM images of (a, b) conventional zeolite Y synthesized at intermediate 12 h after post-treatment, sample Y-12-TD and (c, d) conventional zeolite Y synthesized at final 20 h after post-treatment, sample Y-20-TD.

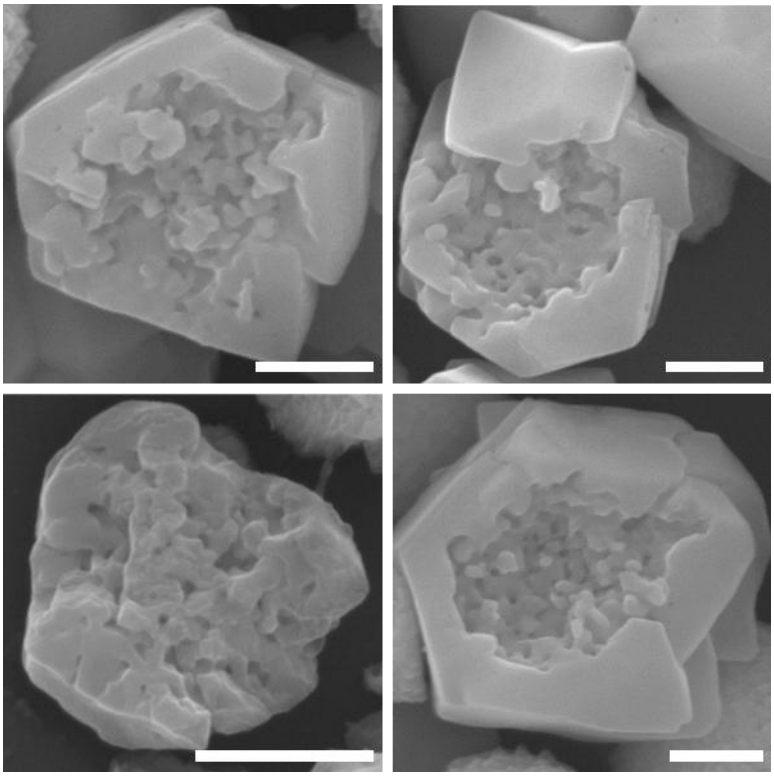


Figure S11. SEM images of surface broken core-shell zeolite Y templated by L-lysine. Scale bar represents 500 nm.

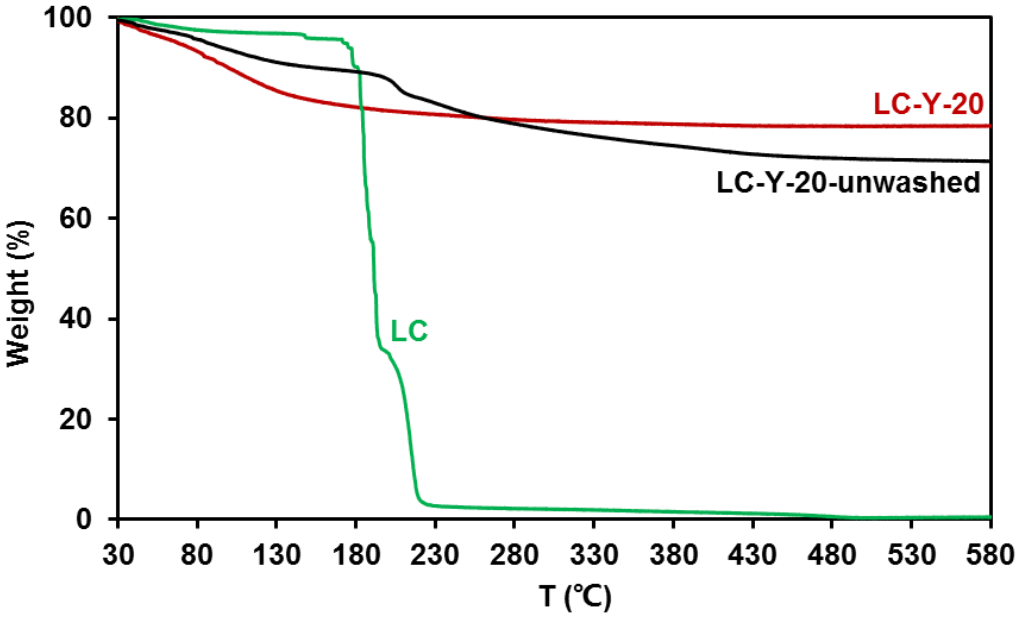


Figure S12. Relative weight loss by TGA of core-shell zeolite Y before and after washing with water, in comparison with that of free L-carnitine.

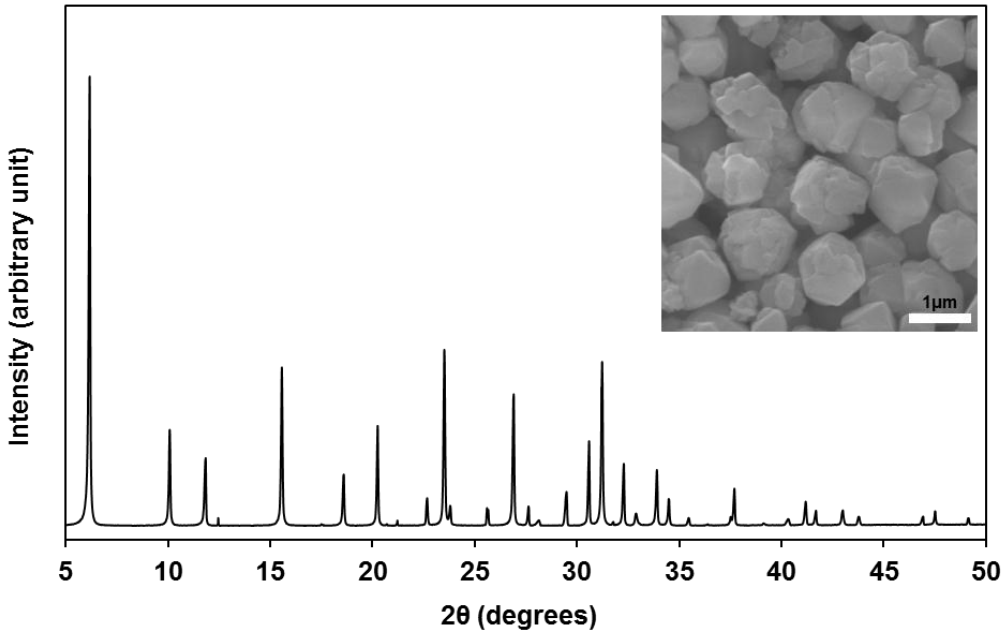


Figure S13. XRD pattern and SEM image of LC-Y-20 recycled from catalytic reaction.

Supporting videos

Supporting video 1. Total volume 3D reconstruction of a carnitine mediated zeolite Y particle LC-Y-20. This video shows intact outer shell covering the entire particle with no discernible mesopores.

Supporting video 2. Selected volume 3D reconstruction of the same carnitine mediated zeolite Y particle LC-Y-20 as shown in video 1. Upon inspecting the volume with part of the shell clipped, it is apparent that the mesopore system of this LC-Y-20 crystal is solely in the core, composed of channel-like mesopores that are three-dimensionally interconnected.

Supporting video 3. Total volume 3D reconstruction of a purposely selected surface broken particle LC-Y-20. This video shows part of the non-mesoporous shell and part of the bored though channels.

Supporting video 4. Selected volume 3D reconstruction of the same carnitine mediated zeolite Y particle LC-Y-20 as shown in video 3. Upon clipping part of the crystal along *xz* plane, the ~165 nm thick shell and the mesoporous core having meanderingly continuous channels are obvious.