Supporting Information

for

Structural and Mechanistic Differences in Mixed-Linker Zeolitic Imidazolate Framework Synthesis by Solvent Assisted Linker Exchange (SALE) and *de novo* Routes

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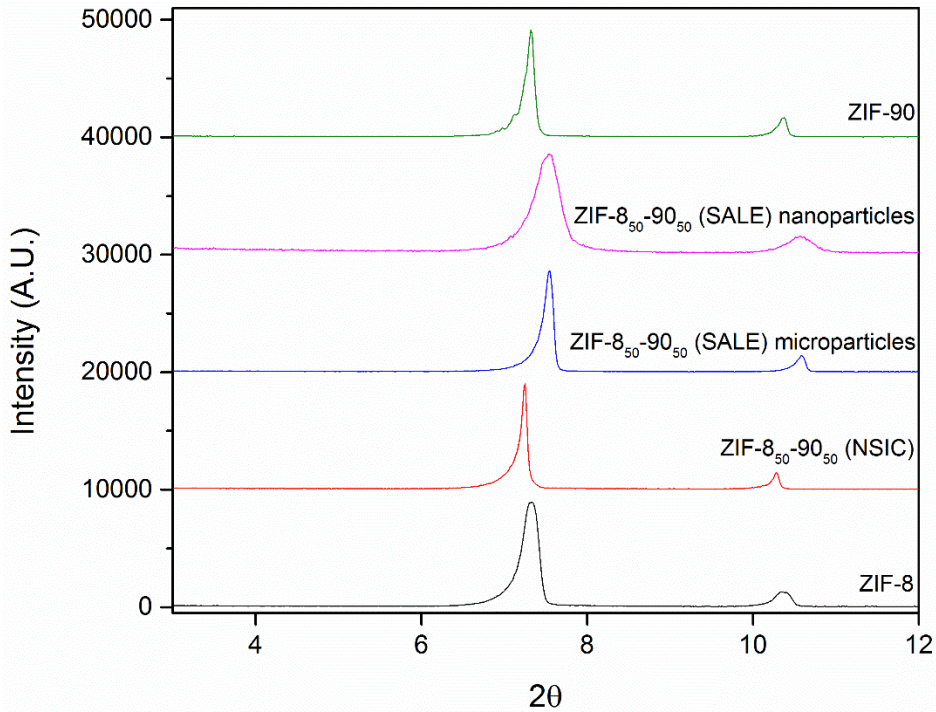

Figure S1. X-ray diffraction patterns of ZIF-8, ZIF-90, NSIC and SALE hybrids The low-angle region is plotted to highlight subtle differences in peak positions for the different structures.

Video file 'A-Pure ZIF-8.mp4'

This video shows fluorescent confocal microscopy images of a pure ZIF-8 crystal. Each frame moves the confocal plane deeper into the crystal. The blue fluorescence exhibited by this crystal is natural (i.e. no dye was required). The relative orientation of the rhombic dodecahedral crystal is shown below in Figure S2.

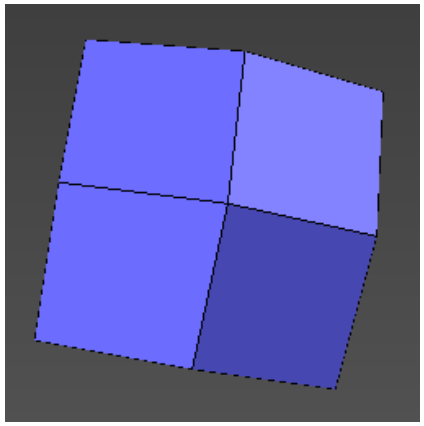

Figure S2. Orientation of the ZIF-8 crystal with respect to the objective lens - as seen in A-Pure ZIF-8.mp4

Video file 'B-Pure ZIF-90.mp4'

This video shows fluorescent confocal microscopy images of a pure ZIF-90 crystal. Each frame moves the confocal plane deeper into the crystal. The green fluorescence exhibited by this crystal is natural (i.e. no dye was required). The relative orientation of the rhombic dodecahedral crystal is shown below in Figure S3.

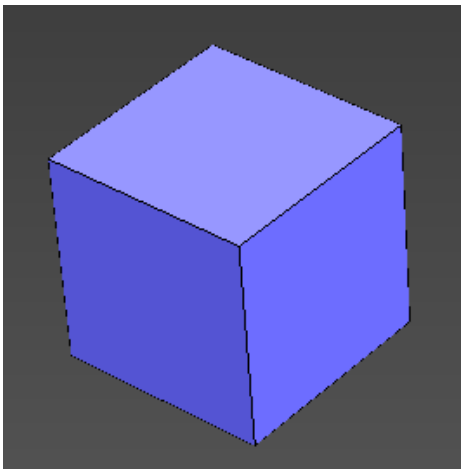

Figure S3. Orientation of the ZIF-90 crystal with respect to the objective lens - as seen in B-Pure ZIF-90.mp4

Video file 'C-ZIF-8-90 de novo hybrid.mp4'

This video shows fluorescent confocal microscopy images of two hybrid ZIF-8_{75}-90_{25} crystal made via *de novo* synthesis route. The cyan color emitted by the crystal is a combination of the blue fluorescence from ZIF-8 and green fluorescence from ZIF-90.

Video file 'D-ZIF-8-90 de novo hybrid blue only.mp4'

This video shows the isolated blue fluorescence from the confocal microscopy images of two hybrid ZIF-8_{75}-90_{25} crystal made via *de novo* synthesis route. Blue color emission is seen throughout the crystal showing the ZIF-8 distributed evenly through the framework.

Video file 'E-ZIF-8-90 de novo hybrid green only.mp4'

This video shows the isolated green fluorescence from the confocal microscopy images of two hybrid ZIF-8_{75}-90_{25} crystal made via *de novo* synthesis route. Green color emission is seen throughout the crystal showing the ZIF-90 distributed evenly through the framework.

Video file 'F-ZIF-8-90 SALE hybrid 1.mp4'

This video shows the fluorescent confocal microscopy images of hybrid ZIF-8_{90}-90_{10} crystal made via Solvent Assisted Linker Exchange route. Blue color emission is seen at the center part of the crystal and green color is seen at the outer edges of the crystal which corresponds to a core of ZIF-8 and a shell of ZIF-90. Each frame moves the confocal plane deeper into the crystal. The relative orientation of the crystal with respect to the objective lens of the microscope is shown in Figure S4.

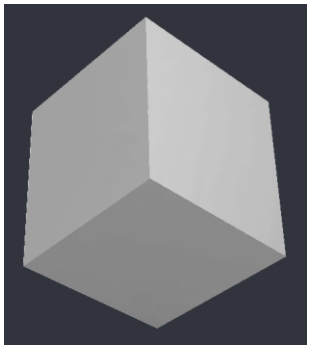

Figure S4. Orientation of the SALE hybrid ZIF-8_{90}-90_{10} crystal with respect to the objective lens - as seen in F-ZIF-8-90 SALE hybrid 1.mp4

Video file 'G-ZIF-8-90 SALE hybrid 2.mp4'

This video shows the fluorescent confocal microscopy images of two hybrid ZIF-8_{90}-90_{10} crystals made via Solvent Assisted Linker Exchange route, sitting side-by-side. Core-shell distribution of ZIF-8 and ZIF-90 can been seen from the blue and green light emitted from the center and edge of the crystals.

Video file 'H-ZIF-8-90 SALE hybrid 3.mp4'

This video shows the fluorescent confocal microscopy images of hybrid ZIF-8_{90}-90_{10} crystal made via SALE. Blue color emission is seen at the center part of the crystal and green color is seen at the outer edges of the crystal which corresponds to a core of ZIF-8 and a shell of ZIF-90. Each frame moves the confocal plane deeper into the crystal. The relative orientation of the crystal with respect to the objective lens of the microscope is shown in Figure S5.

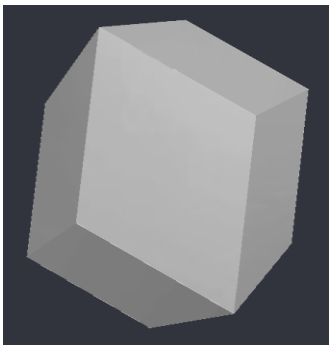

Figure S5. Orientation of the SALE hybrid ZIF-8_{90}-90_{10} crystal with respect to the objective lens - as seen in F-ZIF-8-90 SALE hybrid 3.mp4