

Supporting information:

As we know, ZSM-35 zeolite is composed of 10-membered ring (10-MR, $4.2 \times 5.4 \text{ \AA}$) straight channels and 8-MR ($3.5 \times 4.8 \text{ \AA}$) straight channels, which are perpendicularly intersected each other with 6-MR straight channels in framework. A spherical cavity with a size of 0.6–0.7 nm is formed by an intersection of the 8-MR and 6-MR channel^[51]. We have synthesized pure ZSM-35 zeolite under static hydrothermal treatment system, at 493K, using cyclohexylamine as template, Si-sol as Si source by UV resonance Raman spectroscopy (Figure S1). The building units in the precursors were identified as five- and six-membered silicate ring by UV Raman spectroscopy. The intensity of the Raman band at 450 cm^{-1} increase and the new characteristic bands of ZSM-35 zeolite at 215, 312 and 421 cm^{-1} appear during the formation of ZSM-35 zeolite, which turned out to be the aggregation of these ring species through hydrothermal treatment. The strong Raman band at 421 cm^{-1} is assigned to the bending vibration of five-membered silicate ring, and the Raman band at 312 cm^{-1} may correspond to the six-membered silicate ring of the FER framework.

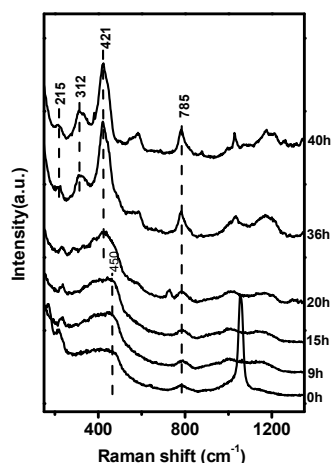


Figure S1. UV resonance Raman spectra of ZSM-35 samples for different crystallization stages ($\lambda_{\text{exc}}=244 \text{ nm}$).

Figure S2 shows UV Raman spectra of the solid phase of products with direct-synthesis method excited at 325 nm. The Raman bands at 460, 794, and 984 cm^{-1} are characteristics for all the samples and the three bands are all characteristics of vitreous SiO_2 ^[52]. The broad Raman band in the $400\text{--}500 \text{ cm}^{-1}$ region is assigned to $\nu_s(\text{Si-O-Si})$ of five- and six-membered silicate rings and comes from amorphous or vitreous silica^[53]. The presence of the 984 cm^{-1} band is related to the Si-O-Si bond next to the framework iron species or other defect site such as the surface silanol group^[45, 54-55]. The two broad Raman bands centered at 460 and 984 cm^{-1} become prominent with increasing the crystallization time, while the band at 984 cm^{-1} shifts to 990 cm^{-1} . The result suggests the amount of five- and six-membered silicate rings increases in the precursors. The product crystallized for 20 h with direct-synthesis is still in the amorphous structure as determined by X-ray diffraction pattern. Notably, a new Raman band at 1143 cm^{-1} appeared in 3 hour. This band ascribes to a totally symmetric stretching vibration of $[\text{FeO}_4]^{2-}$. The appearance of the band indicates that the iron ions are uniform and rigid tetrahedral coordination, although the framework of Fe-ZSM-35 is not formed yet. This indicates the iron ions with tightly tetrahedral coordination in the amorphous silica in the early stage. In other words, by extending the crystallization time, the FER structure will not be formed from the gel.

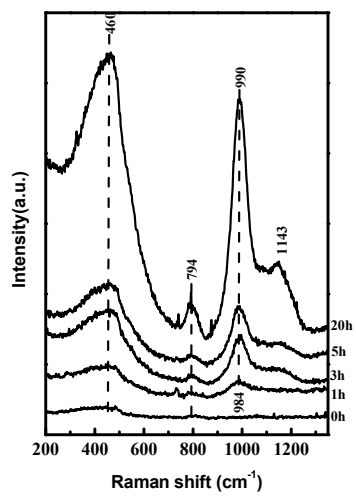


Figure S2. UV Raman spectra of Fe-ZSM-35 (Si/Fe=80) samples with the direct-synthesis method for different crystallization stages at various time of synthesis ($\lambda_{\text{ex}}=325$ nm).