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6 **Sodium carbonate-assisted synthesis of hierarchically porous single-**  
7 **crystalline nanosized zeolites**

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13 **Abstract**

14 Hierarchically porous single-crystalline nanosized zeolites as heterogeneous catalysts show  
15 great potential in fine chemistry because they offer more rich hierarchically porous channels  
16 for the mass transfer and molecular diffusion. However, the synthesis of hierarchically porous  
17 nanosized zeolites generally requires the assistance of templates acting as the mesoporegens,  
18 which limits its popularity. Herein, we report a one-pot and template-free synthesis of  
19 hierarchically porous single-crystalline nanosized zeolite beta only by introducing sodium  
20 carbonate in precursor solution. The resulted sample features the extraordinary properties,  
21 including the uniform nanocrystal (200-300 nm), high pore volume ( $0.65 \text{ cm}^3 \text{ g}^{-1}$ ) and the  
22 hierarchical pore-size distribution (e.g., 2-8 and 90-150 nm). After slicing processing, it is  
23 interestingly found that a large number of interconnected mesopores penetrate throughout  
24 whole material, which enables the hierarchically porous nanosized zeolite beta remarkably  
25 superior catalytic activity than the conventional zeolite beta in condensation of benzaldehyde  
26 with ethanol at room temperature. More importantly, this one-pot sodium carbonate-assisted  
27 synthetic strategy is highly versatile, which has also been successfully developed to  
28 synthesize hierarchically porous nanosized single-crystalline zeolites ZSM-5 and TS.

29 **Key words:** Hierarchically porous, Single-crystalline, Beta/ZSM-5/TS, Sodium carbonate,  
30 Template-free, Condensation of benzaldehyde

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