





## Formation Process of Columnar Grown (101)-Oriented Silicalite-1 Membrane and its Separation Property for Xylene Isomer

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Figure S1 shows a particle size distribution of seed crystals in slurry for dip-coating measured by a dynamic light scattering. Almost all crystals in the slurry had a diameter of 200–500 nm which is slightly larger than pore size of  $\alpha$ -alumina support of 150 nm.



Figure S1. Particle size distribution in seed slurry for dip-coating.

Figure S2 shows typical FE-SEM images of a surface of seeded support after insuccation into synthesis solution. Bare support surface can be observed in some part, suggesting that seed crystals were partially dropped by the insuccation.



**Figure S2.** Typical FE-SEM images of surface of seeded supports immersed into synthesis solution without calcination after dip-coating. (**a**) wide and (**b**) enlarged view.

Figure S3 shows a schematic diagram of evolutionally selection. In evolutionally selection mode, crystals in which the axis of highest growth rate stand perpendicular to support are finally selected and continue to grow.

Seed crystal "Evolutionally selection" Seeded support **Oriented membrane** 

Figure S3. A schematic diagram of evolutionally selection.