

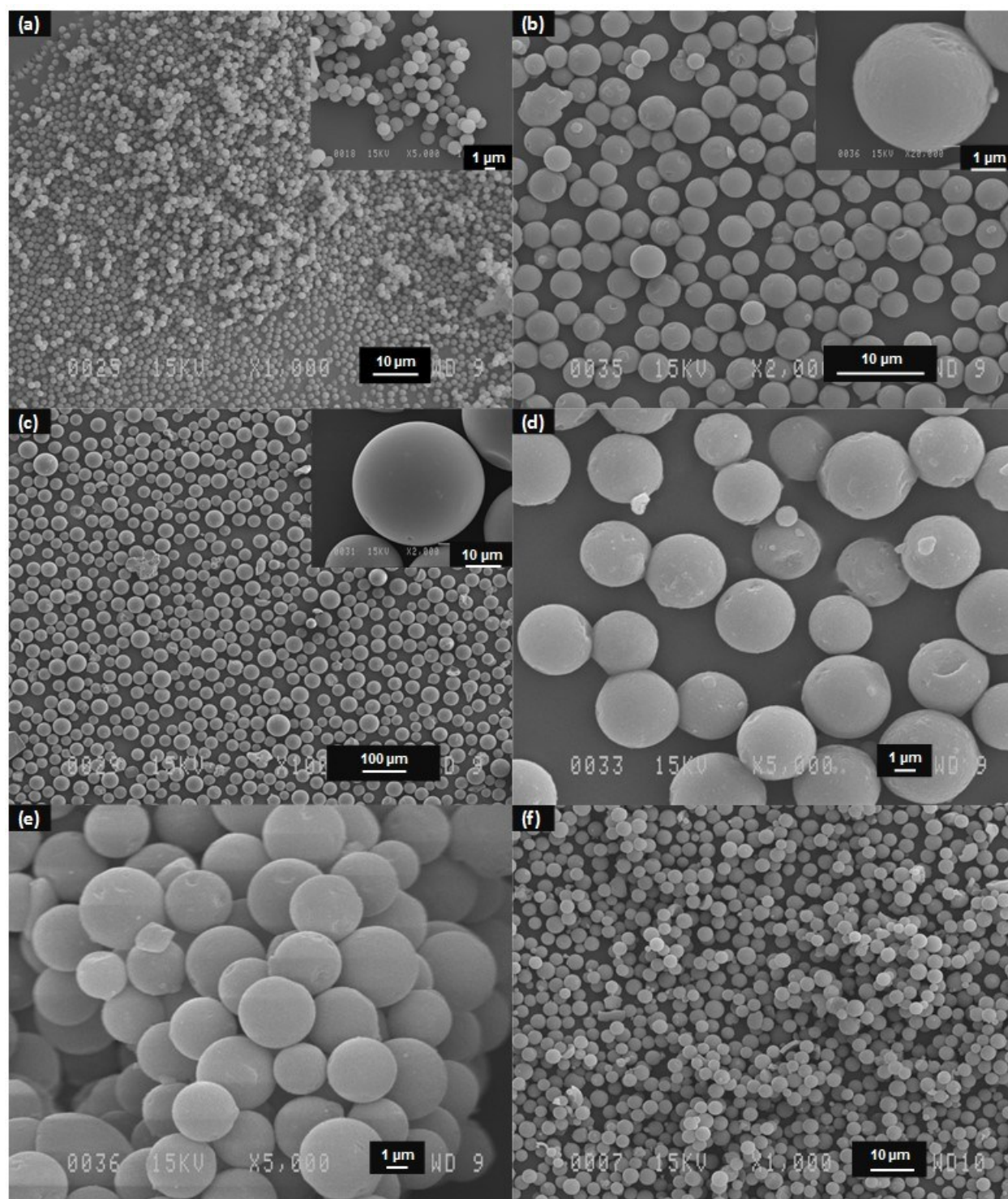


Figure S1- SEM images of (a) HMS microspheres, (b) silica gel spheres 3 μm (SG3), (c) silica gel spheres 20-45 μm (SG20) and (d-f) SG3 spheres containing (d) cobalt (Co,SG3), (e) manganese (Mn,SG3), and (f) titanium (Ti,SG3).

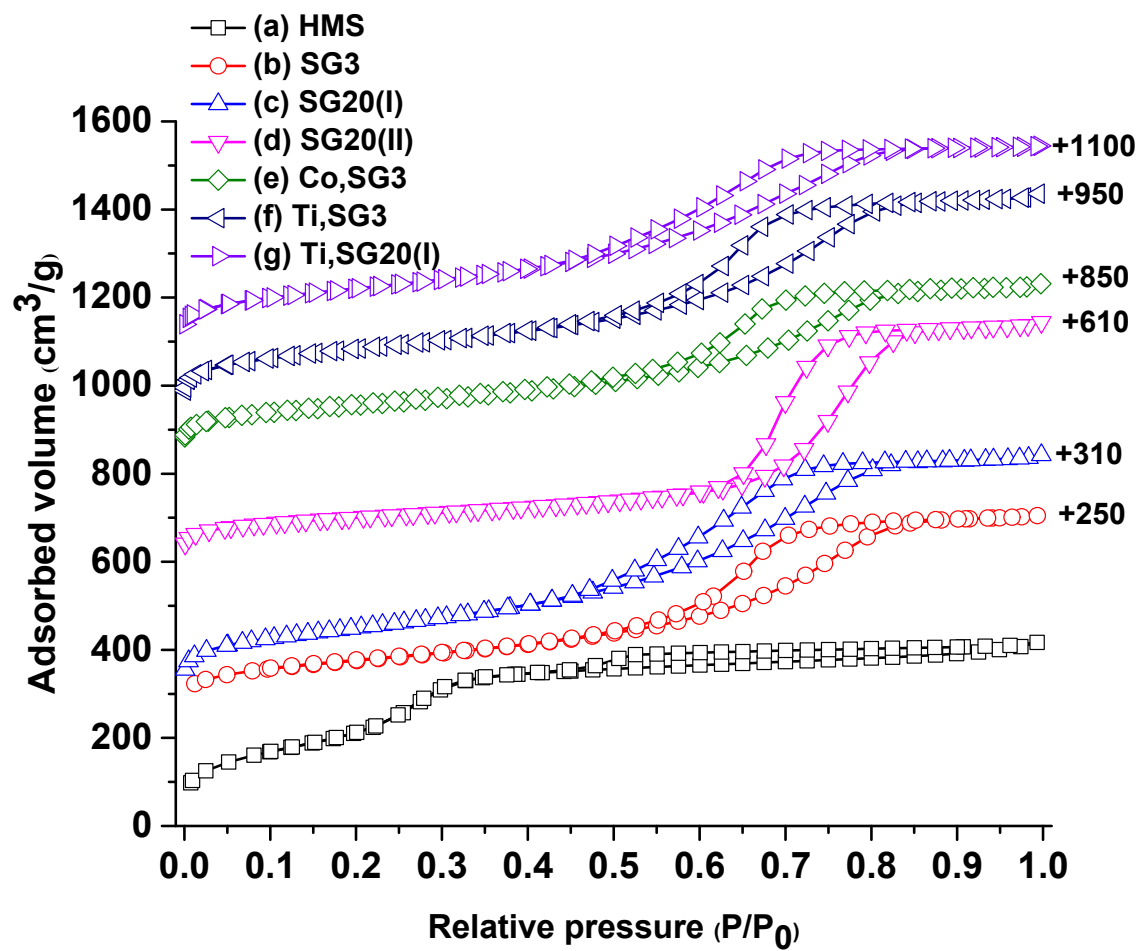


Figure S2- Nitrogen adsorption/desorption isotherms measured at -196 °C for different core particles.

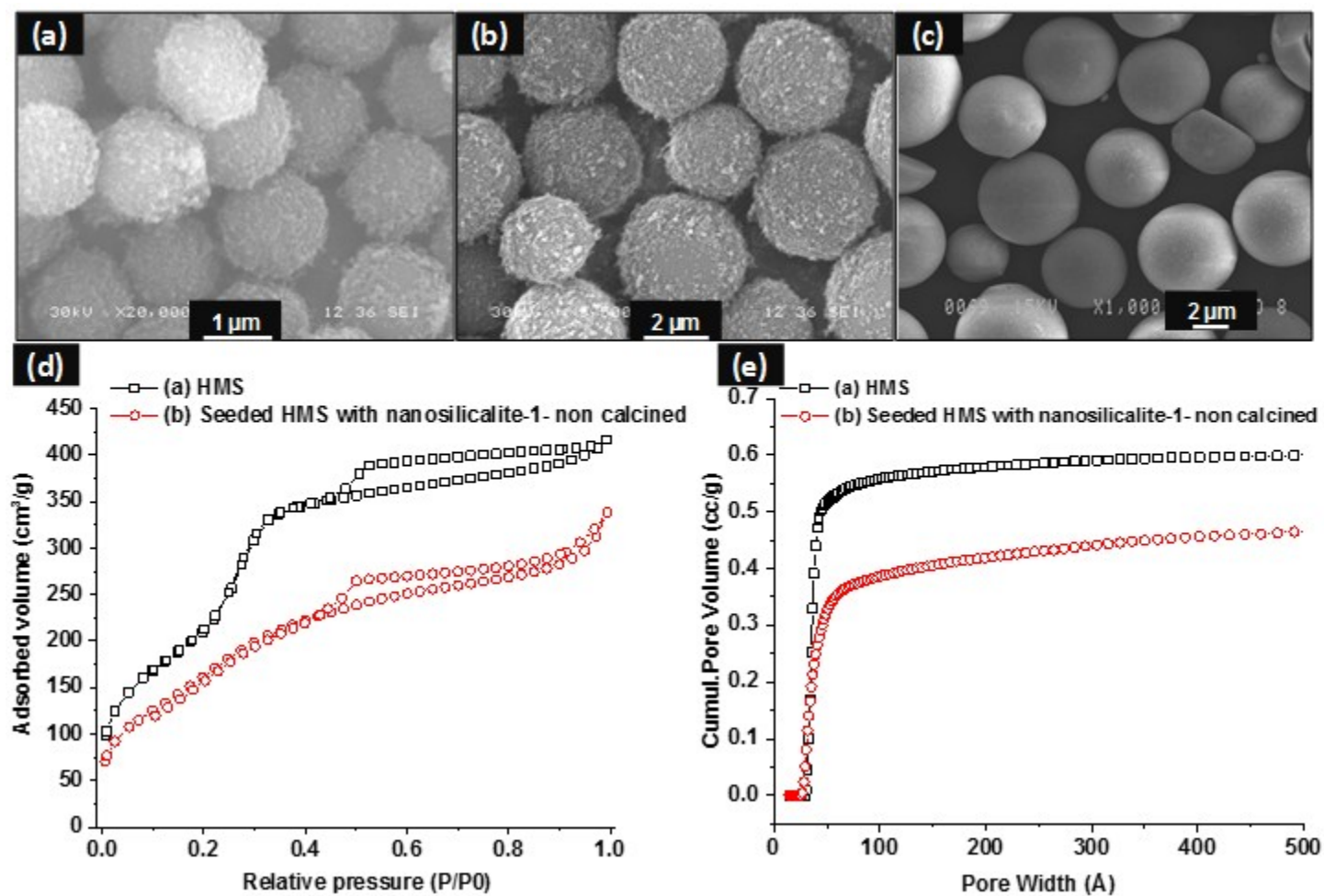


Figure S3- (a-c) SEM images of HMS, SG3 and SG20 particles, covered with silicalite-1 nanocrystals, (d) nitrogen physisorption isotherms (-196°C) and (e) NLDFT cumulative pore volume plots of pure and non-calcined seeded HMS particles.

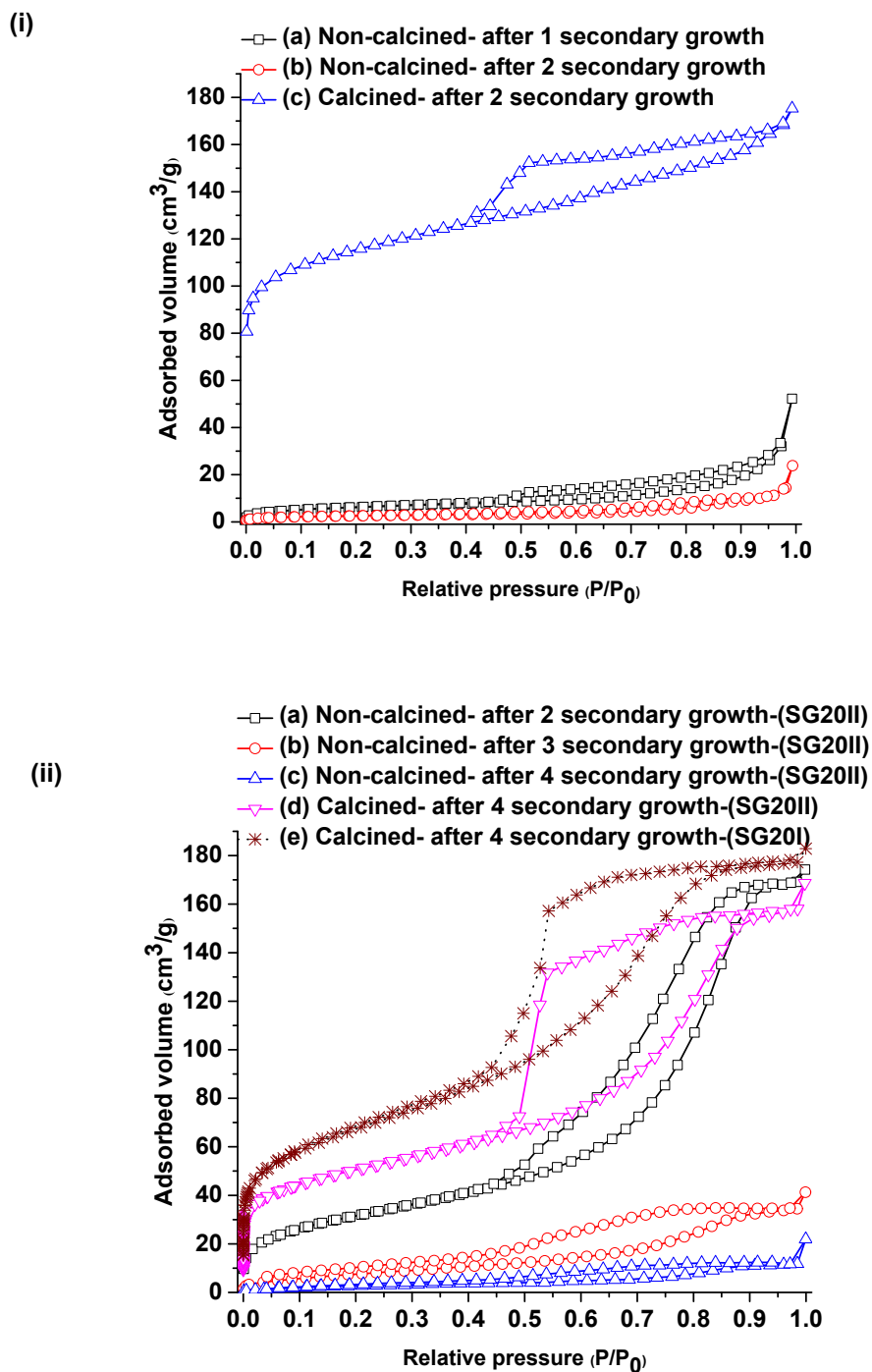


Figure S4- Nitrogen adsorption/desorption isotherms measured at -196°C (i) HMS and (ii) SG20 at different stages of the core@shell synthesis.

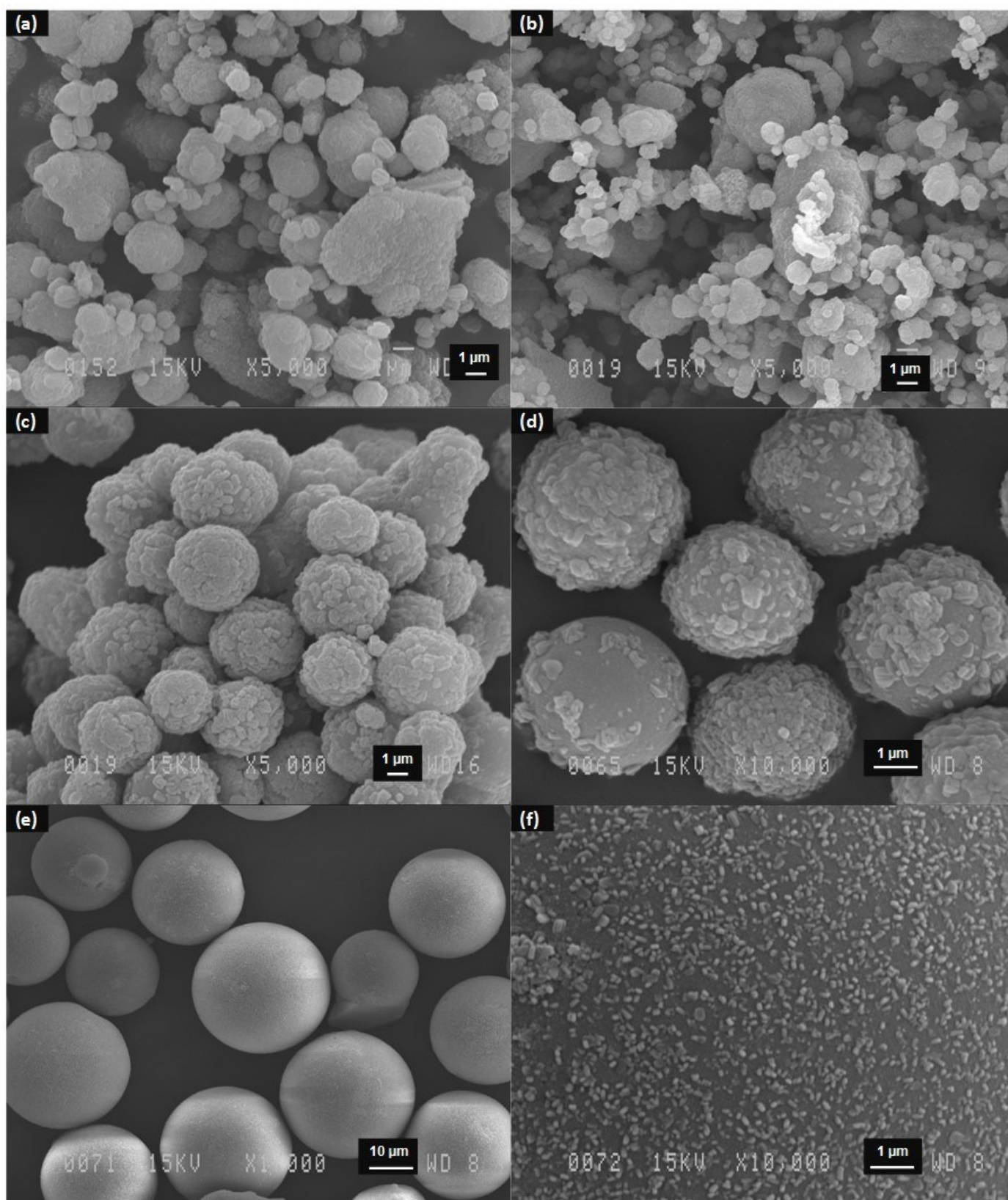


Figure S5- SEM images of (a) Co,HMS@silicalite-1, (b) Co,SG3@silicalite-1, (c) Co,SG3@silicalite-1, (d) Mn,SG3@silicalite-1, (e) Co,SG20(I)@silicalite-1, and (f) Co,SG20(I)@silicalite-1 with higher magnification.

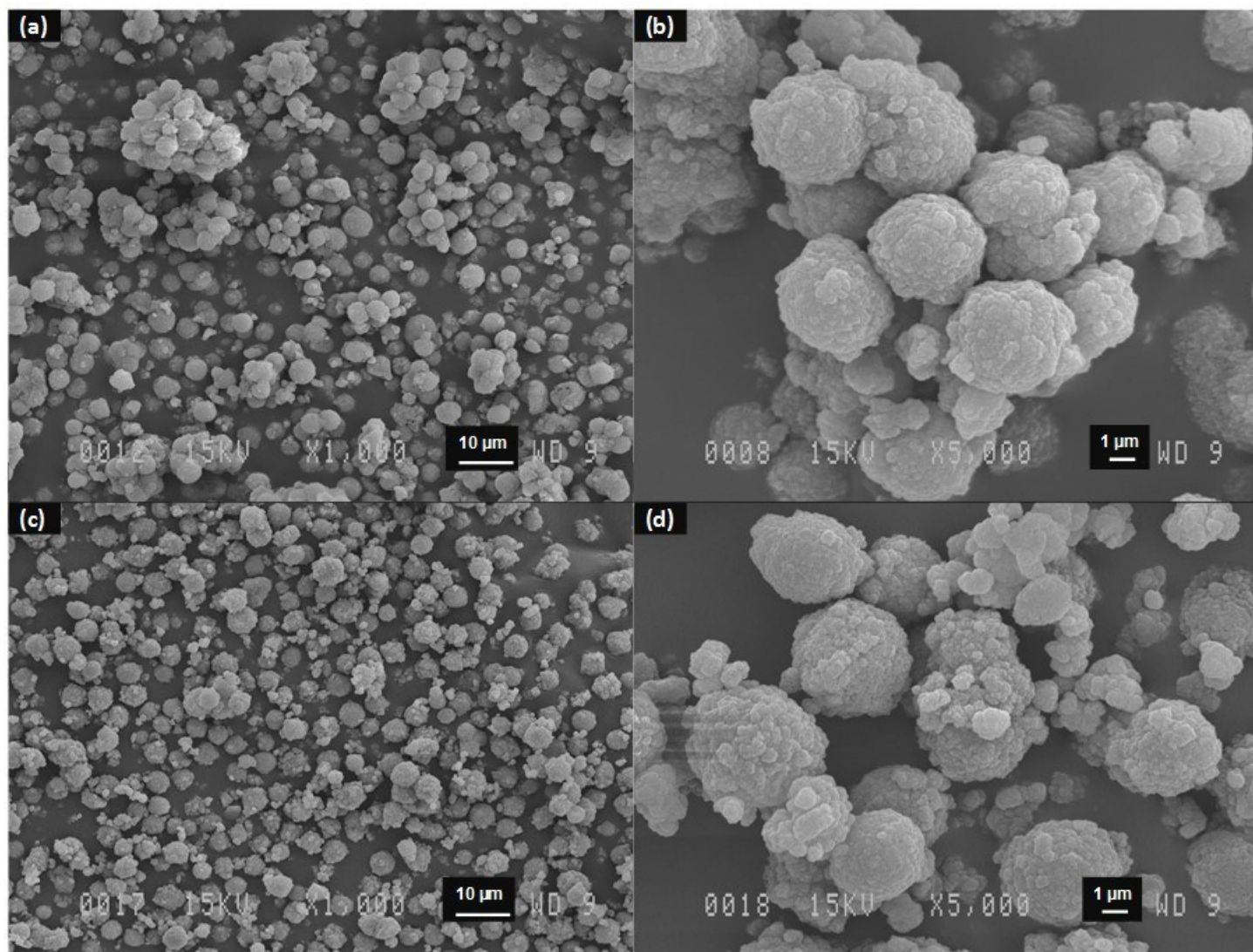


Figure S6- Effect of multilayer polymer coating (PDADMAC/PSS/PDADMAC) on (a,b) SG3@silicalite-1, and (c,d) Co, SG3@silicalite-1.

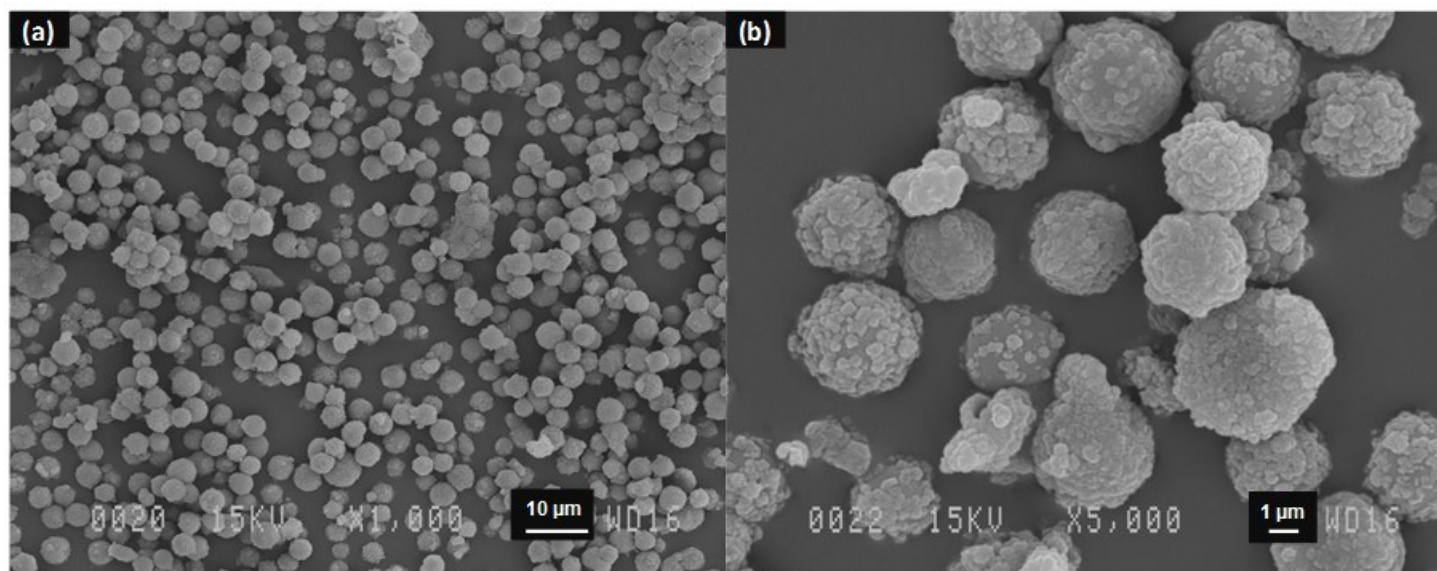


Figure S7- Effect of the sole APTES modification on Mn,SG3@silicalite-1 synthesis after three secondary growth steps.

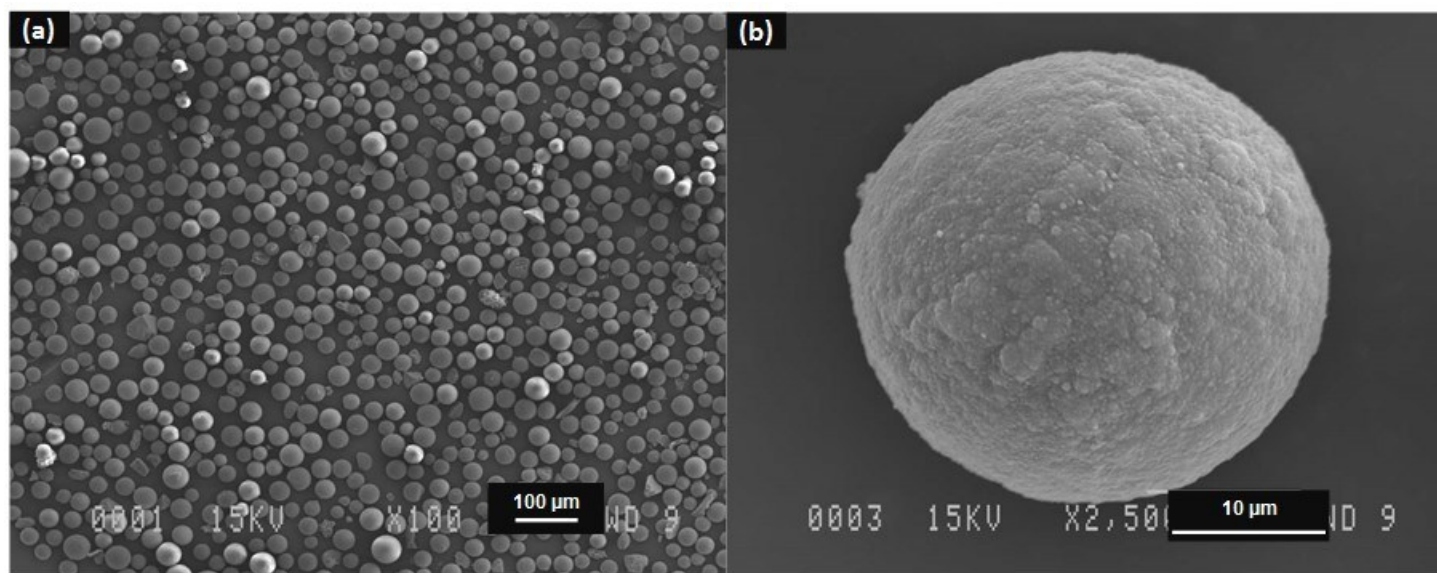


Figure S8- SEM images of Ti,SG20(I)@silicalite-1 after four secondary growth steps.

Intergrown microporous silicalite-1 shell

Mesoporous silica gel (SG20(I))

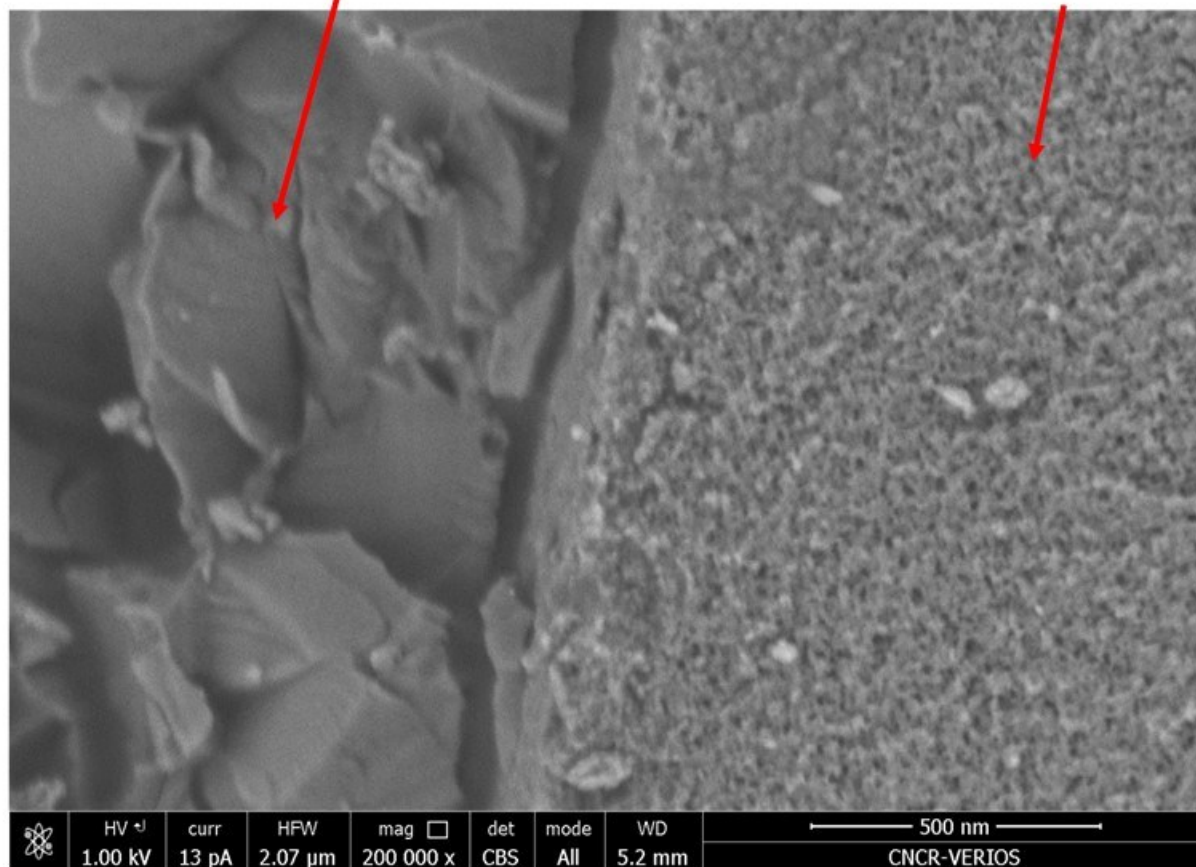


Figure S9- Higher magnification HRSEM image of Co,SG20(I)@silicalite-1 sample, broken using a mortar and pestle.

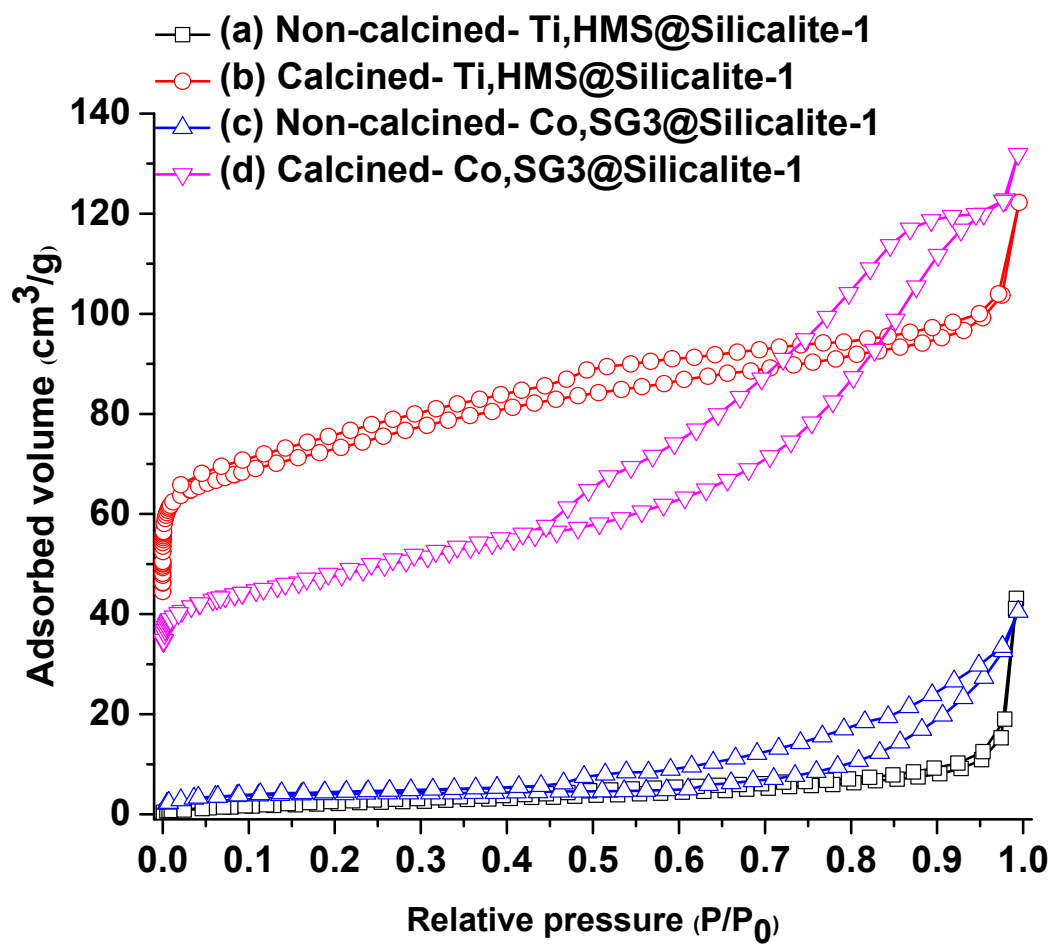


Figure S10- Nitrogen adsorption/desorption isotherms measured at -196 °C for (a,b) Ti,HMS@silicalite-1 before and after calcination, and (c,d) Co,SG3@silicalite-1 before and after calcination.

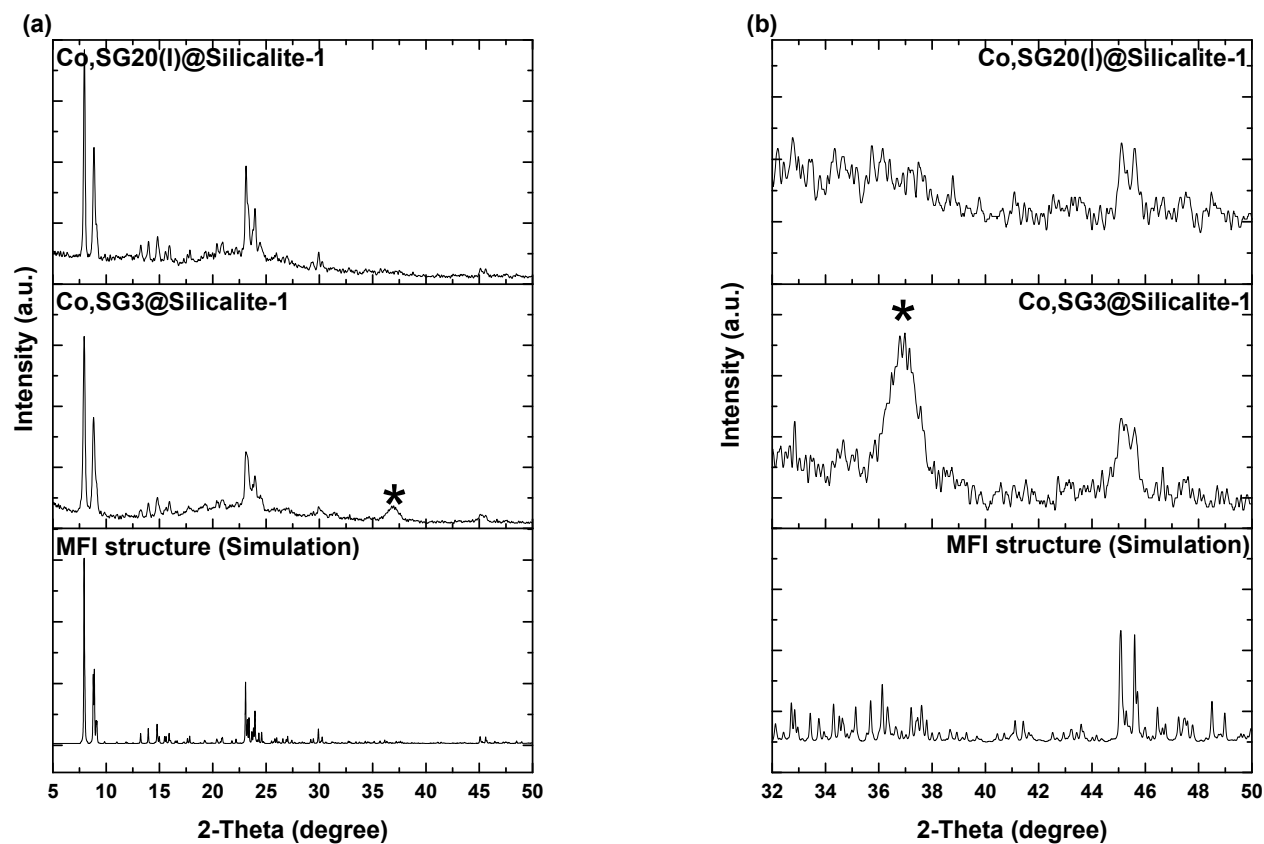


Figure S11- XRD patterns of Co,SG3@Silicalite-1 and Co,SG20(I)@Silicalite-1. Simulations for calcined MFI structure were obtained from IZA website ¹.

1. I. Webpage, <http://www.iza-structure.org/databases/>.