

**Supplementary Information:**

**Formation of Nano-structured Core-Shell Micro-  
granules by Evaporation Induced Assembly**

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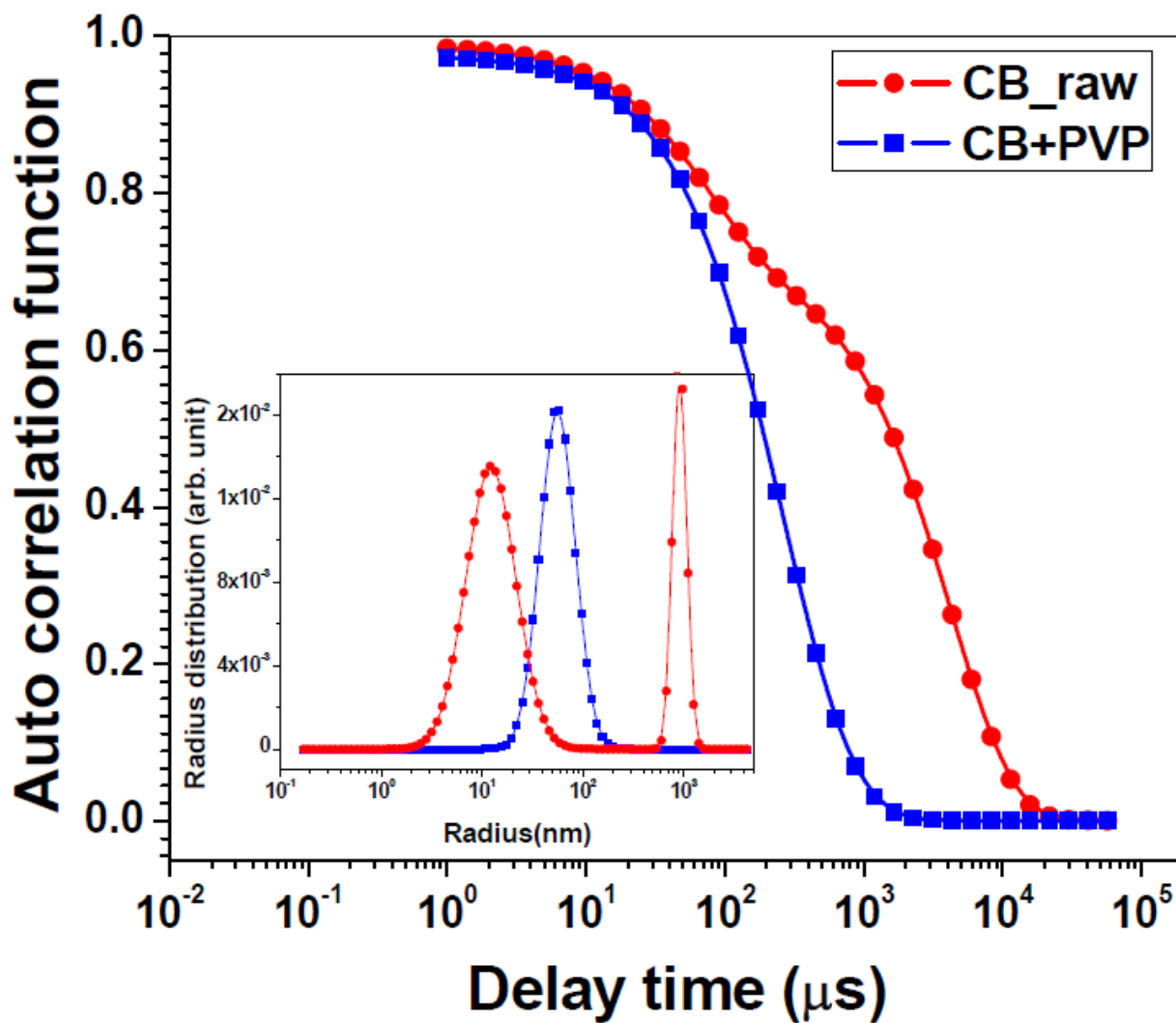
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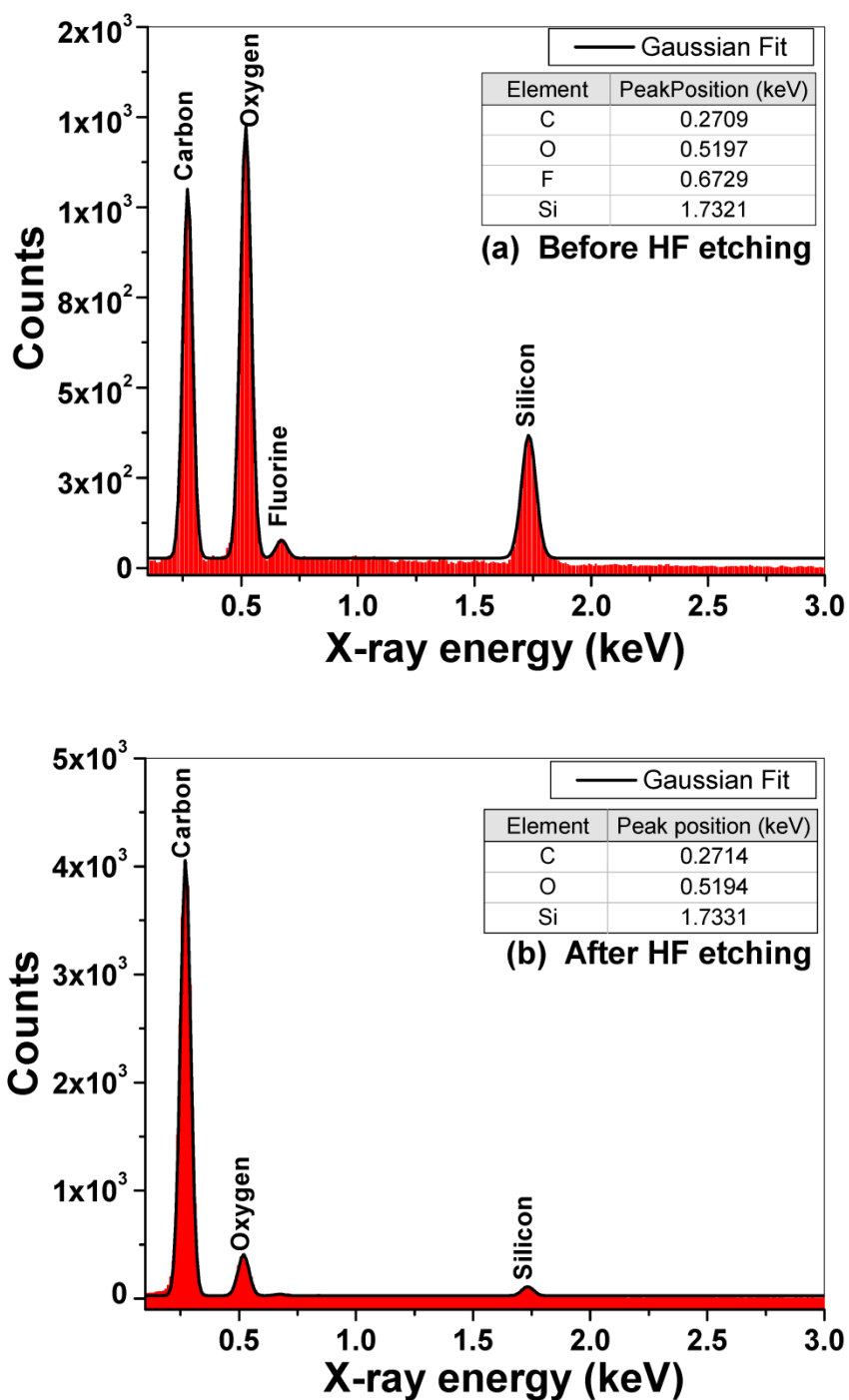
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### 1) DLS measurement:



**Fig. S1** The auto-correlation functions, obtained from DLS measurement, have been depicted. The inset shows radius distribution by fitting the auto-correlation function considering lognormal distribution function. Raw CB sample shows bi-modal distribution whereas, PVP coated CB shows mono-modal distribution.

## 2) Energy dispersive X-ray spectroscopy:



**Fig. S2** The energy dispersive spectroscopy of silica coated CB microparticles showing the presence of different elements (a) before HF etching and (b) after HF etching.

### 3) BET measurement:

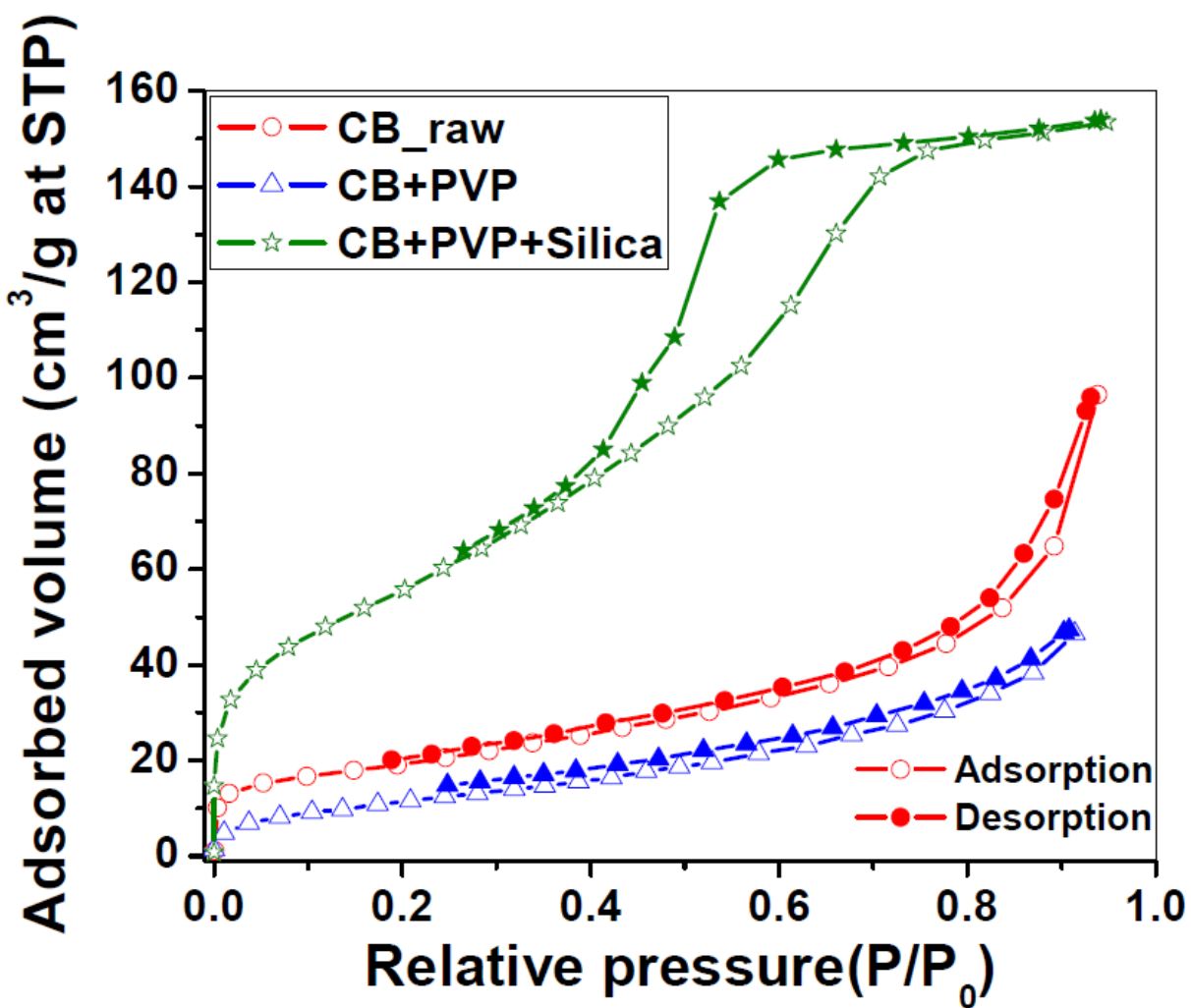
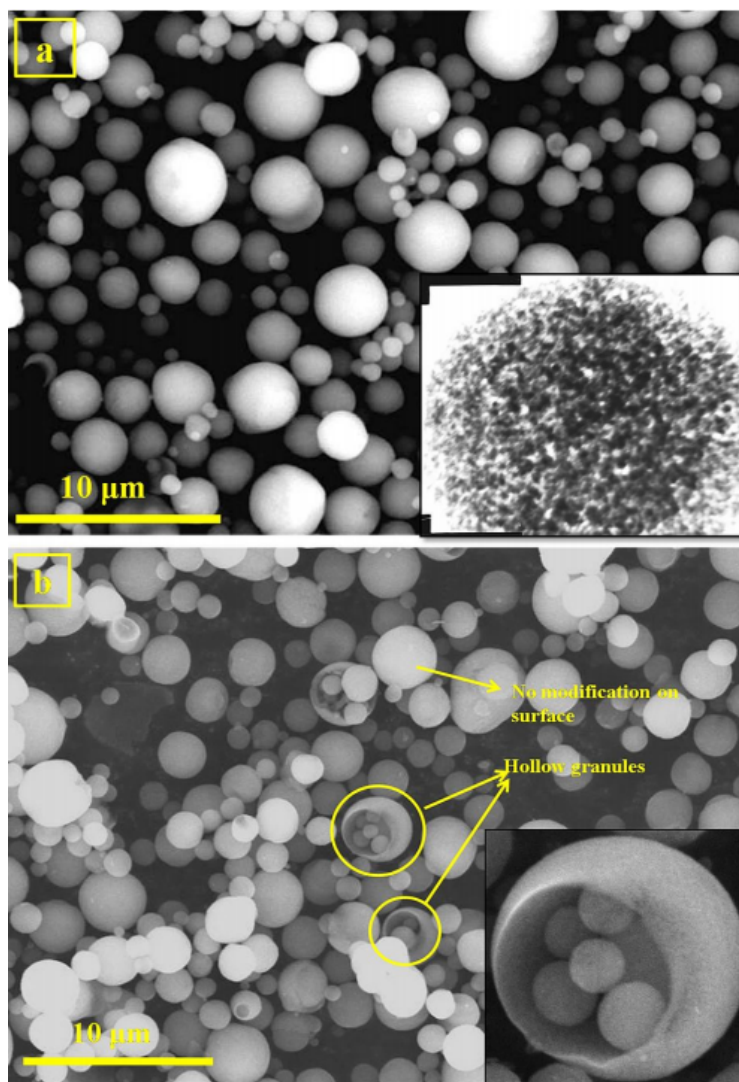


Fig. S3 Nitrogen adsorption (open motif)-desorption (solid motif) isotherms of raw carbon black powder and spray-dried powders.

#### **4) Verification of formation of core-shell micro-granules using silica and amphiphilic molecules:**

To verify the versatility of the role of interfacial interaction during evaporation induced assembly, we have spray dried a stable colloidal mixture consisting of silica nano-particles (2 wt%) and sodium dodecyl sulfate (SDS) nano-particles (2 wt%). Silica is hydrophilic whereas, SDS is comprised of hydrophilic head and hydrophobic tail. It has been observed that during drying of the colloidal suspension, evaporation of water influences the SDS nano-particles to occupy the core region of the droplet while silica nano-particles make a shell around peripheral surface. The micrograph (Figure S4(a)) depicts spherical micro-granules of silica-SDS nano-composite and the micrograph in inset reveals the core-shell morphology of the SDS-silica spray-dried micro-granule. The spray-dried granules are thoroughly washed with methanol to remove the SDS nano-particles. The micrograph of the spray-dried granules after methanol washing (Figure S4 (b)) shows no signature of templated pore on the surface of the grains, instead it reveals hollow granules. The inset micrograph in Figure S4(b) is a zoomed picture of one hollow micro-granule. This clearly demonstrates that after washing the granules with methanol, SDS leaves the granule from inside core region making the granule hollow without any modification in surface. This suggests that the SDS remains in core region while the silica forms outer shell of the granules. Thus, the competitive phenomenon of interfacial interaction during evaporation induced assembly can indeed be utilized to form core-shell micro-granules of hydrophobic and hydrophilic nano-particles for other system provided other competitive parameters are tuned properly.



**Fig. S4** Micrographs depict silica-SDS nano-composite spray-dried micro-granules (a) before methanol washing and (b) after methanol washing. The inset micrograph in (a) shows core-shell morphology of the spherical micro-granule, whereas the inset micrograph in (b) shows hollow spherical granule after methanol washing..