SANS study of Silica Aerogel as Model Material for Rocks

Author: Phung. N. H. (Henry) Vu, Tomasz Blach, Andrzej P. Radlinski, Klaus Regenauer-Lieb

Background and Motivation

- Rise in interest for CO_2 sequestration in sandstone
- Current lack of understanding in sandstone's sorption behaviour
- Rocks have wide range of pore size and the sorption capacity is different for different pores
- Possible formation of adsorbed layer which can block accessibility to nanopores
- Possibly modified density of confined fluid
- Accurate estimation of overall sorption capacity

Silica Aerogel

- Man-made material, homogeneous, total open porosity
- Has board pore size distribution from 8-100 nm
- Three populations of pores at around 0.3, 4 and 10 nm in diameter
- Suitable for studying behaviour of methane at nano-scale confinement of various extend

Experiment Methodology

- Silica aerogel sample was crushed and deposited in a sample holder of 1mm inner thickness
- For contrast matching SANS, the sample was subjected to CD4 pressure from 0-1000 bar
- SANS experiment was done at ILL using D11, covering pore Q-range from 2x10^{-3} to 0.6 A^{-1}

Methods for SANS Analysis

- Debye formula:

$$\frac{d\Sigma}{dQ}(Q) = 4\pi(\rho_1^* - \rho_2^*)^2\phi_1\phi_2\int_0^\infty r^2\gamma(r)\frac{\sin(Qr)}{Qr}dr$$

- Mass fractal

$$\frac{d\Sigma}{d\Omega}(Q) \sim Q^{-D_m}$$

- Kirste-Porod formula

$$\frac{d\Sigma}{d\Omega}(Q) = 2\pi(\rho_1^* - \rho_2^*) * \frac{S}{V}Q^{-4}\left(1 + \frac{1}{R}Q^{-2}\right)$$

- Porod Invariant

$$\int_0^\infty Q^2\frac{d\Sigma}{d\Omega}(Q)dQ = 2\pi^2(\rho_1^* - \rho_2^*)^2\phi_1(1-\phi_1)$$

- Guinier formula

$$\frac{d\Sigma}{d\Omega}(Q) = NV_p^2(\rho_1^* - \rho_2^*)^2\exp\left(-\frac{Q^2R_g^2}{3}\right)$$

$$\frac{d\Sigma}{d\Omega}(0) = NV_p^2(\rho_1^* - \rho_2^*)^2$$

Discussion

- The intensity of SANS consists of 4 components: fractal scattering from the silica skeleton and monodispersed cluster of 10nm, 4nm and sub-nm in size
- As CD_4 pressure increases from vacuum to 400 bar, the power slope of the fractal region decreases from -2.1 to -4 and remains unchanged from 400 bar to 1000 bar. This indicates a nanostructure transition from mass fractal to a scattering object with smooth surface
- In the G4 and G10 scattering region, the intensity decreases down to near zero at 400-500 bar, providing the SLD of 3.3x10^{10} cm^{-2} for the aerogel
- At the nanopore region (Q >0.1A^{-1}), the intensity increases by a factor of ~30 going from vacuum to 200 bar and remains almost constant as pressure rise to 1000 bar
- After exposure to CD_4, the plateau in the Porod plot becomes lower due to the residue CD_4 in the sub-nanopore

Results

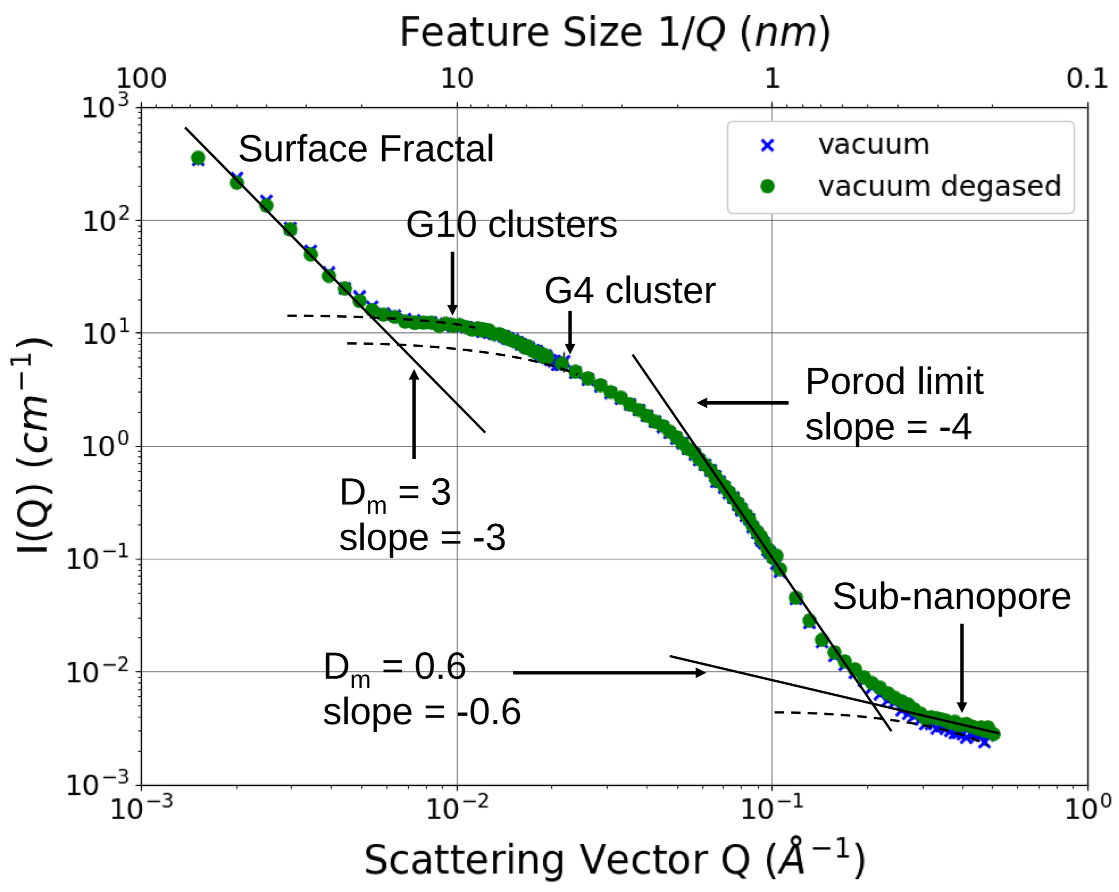

Figure 1. SANS profile at vacuum before and after gas injection

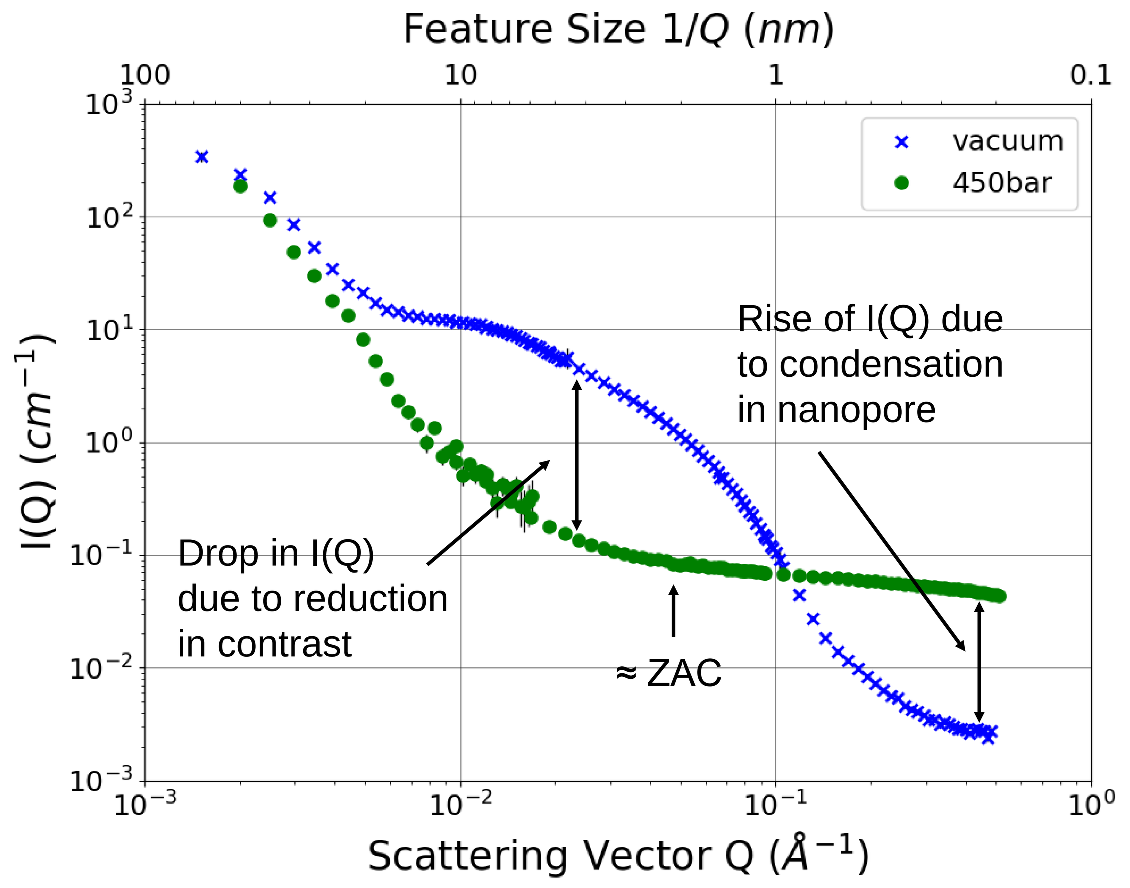

Figure 2. SANS profile at vacuum and ZAC point

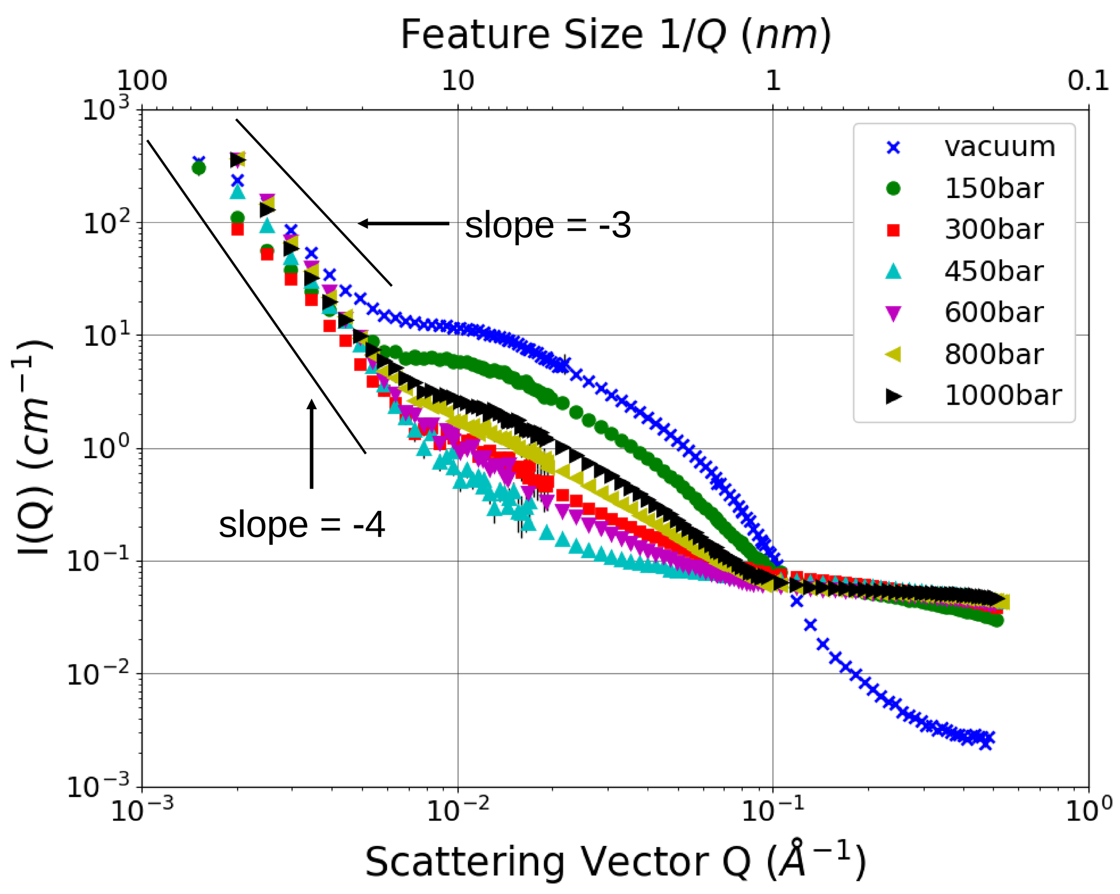

Figure 3. SANS profile variation from vacuum to 1kbar of CD_4

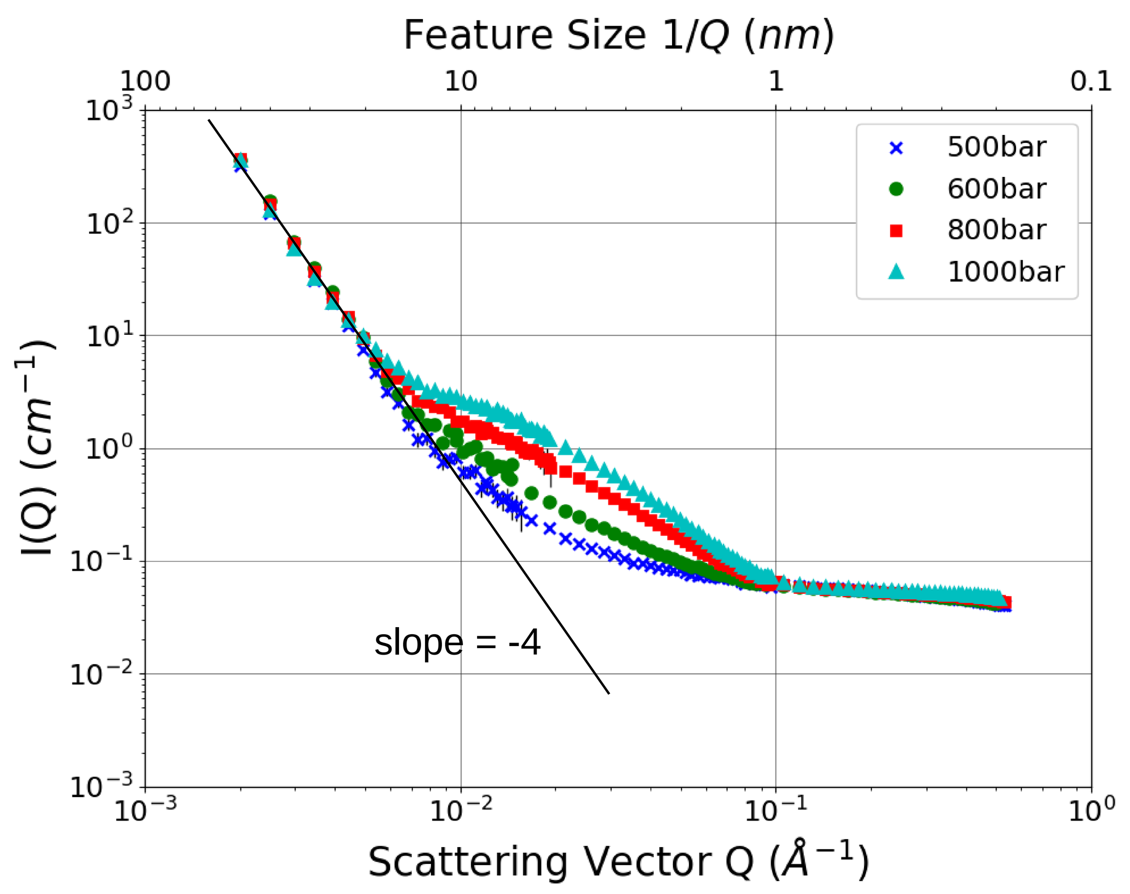

Figure 4. SANS profile variation from 500 bar to 1 kbar of CD_4

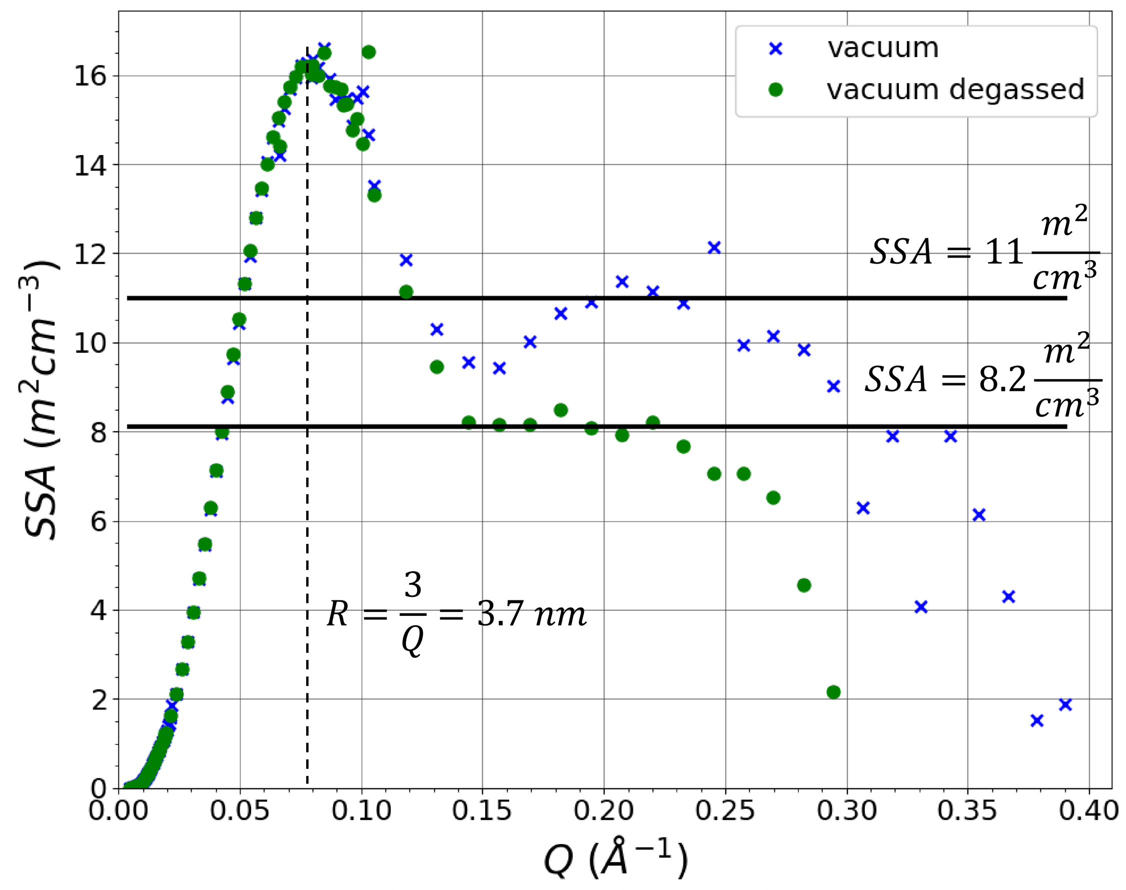

Figure 5. Porod plot at vacuum before and after CD_4 injection

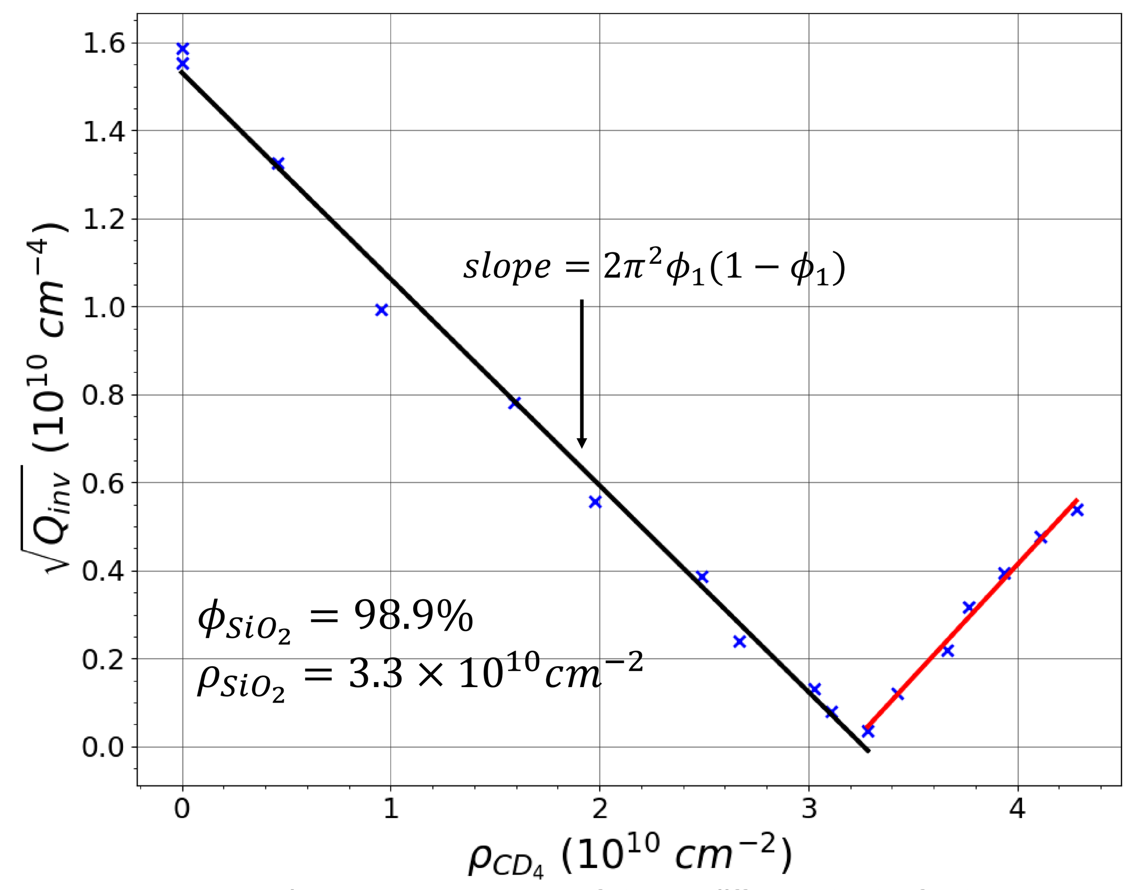

Figure 6. Square root of Q_{inv} at different SLD of CD_4

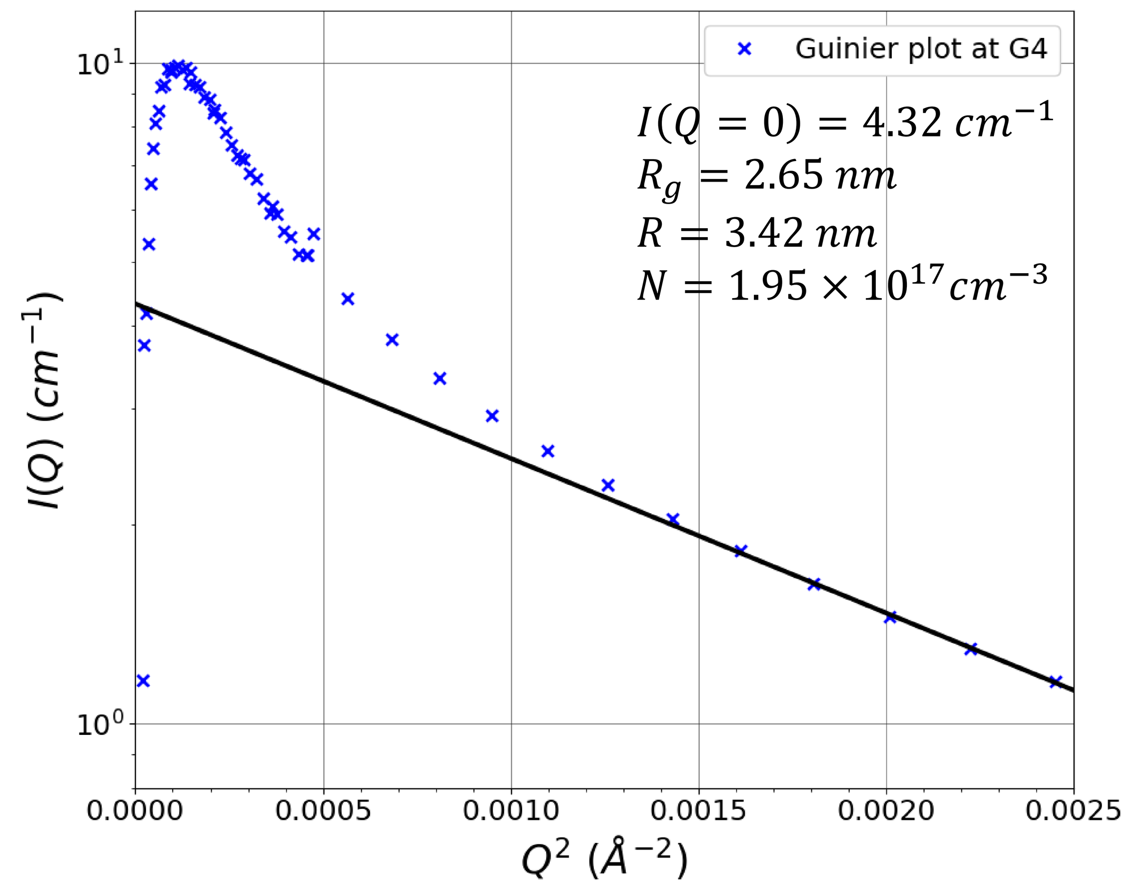

Figure 7. Guinier plot for G4 region at vacuum

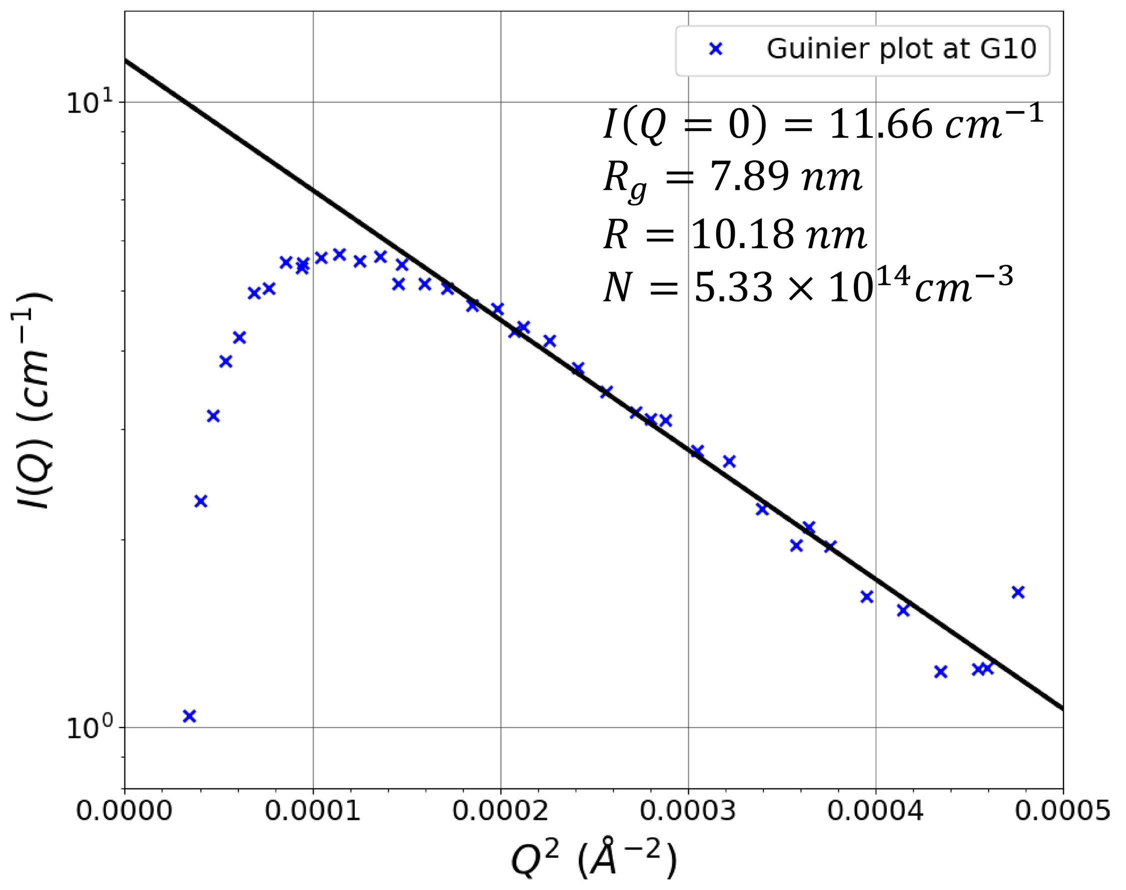

Figure 8. Guinier plot for G10 region at vacuum

Conclusions

- The experiment has shown that the silica consists of multiple scattering regimes: fractal and monodisperse of multiple sizes
- The sorption behaviour of CD_4 on the silica aerogel varied with scale, with the SLD of CD_4 exhibits a linear relationship with bulk density in G4 and G10 region compared to the condensation behaviour of CD_4 in the sub-nanometer pore
- Damage to the structure and residue CD_4 is observed on the sub-nanopore region of the aerogel after the injection process

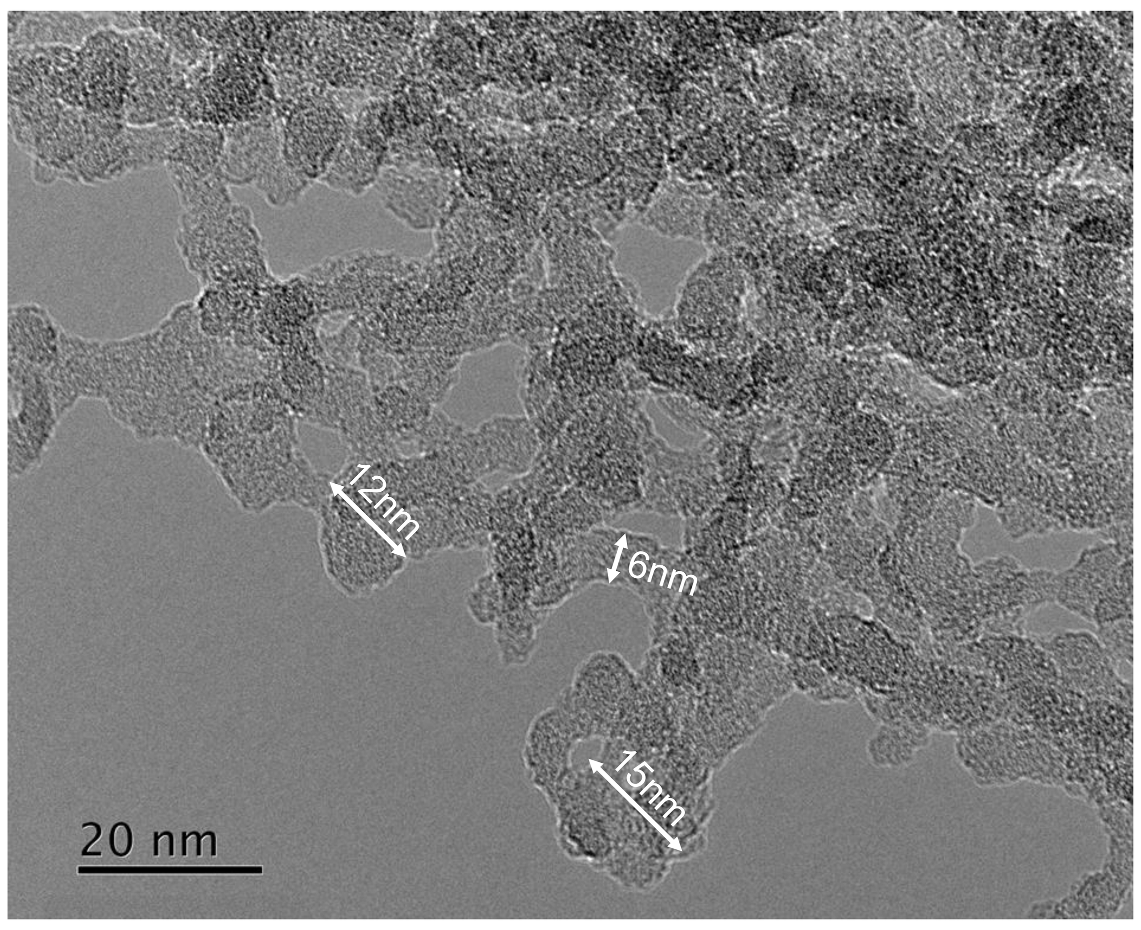

Figure 9. TEM image of the silica aerogel