Supplementary Information

Magnetically induced structural anisotropy in binary colloidal gels and its effect on diffusion and pressure driven permeability

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Derivation of Equation 3 and equations to calculate the bond water layer thickness on the silica particle spheres

The observed self-diffusion coefficient of water, D_{obs}, is a population average of the free water fraction p_{free} with self-diffusion coefficient D_{free} and the fraction of water bound to the silica surface p_{bound} with self-diffusion coefficient D_{bound}[1]. The exchange between these two domains is considered to be fast compared to the diffusion time of the NMR experiment. The diffusion of bound water follows the silica particle and the self-diffusion coefficient of silica can be considered negligible. We then have the following equations:

$$D_{obs} = p_{free} D_{free} + p_{bound} D_{bound}$$

$$p_{free} = 1 - p_{bound}$$

$$D_{bound} = D_{particle} \approx 0$$

$$D_{obs} = (1 - p_{bound}) D_{free}$$

We define the volume ratio of pure silica particles in the system as ϕ. From the volume of silica $V_{spheres}$ and the total volume V_{tot}.

$$\phi \equiv \frac{V_{spheres}}{V_{tot}}$$

The obstruction effect of water diffusion due to particles can be described by the following well-known model[2] where D_0 is the self-diffusion coefficient of free water in the absence of silica.

$$D_{free} = \frac{1}{1 + \frac{\phi}{2}} D_0$$

The water fraction that is bound to the silica surface, p_{bound}, is given by the water volume ratio that can be written as,

$$p_{bound} = \frac{V_{bound}^{H_2O}}{V_{tot}^{H_2O}} = \frac{V_{bound}^{H_2O}}{(1 - \phi) V_{tot}}$$

where $V_{bond}^{H_2o}$ is the volume of surface bond water and $V_{\mathrm{tot}}^{\mathrm{H_2o}}$ the total volume of water.

We can now define a volume fraction that is not accessible to free water diffusion as ϕ_{real}. We then have the following four equations:

$$\phi_{real} \equiv \frac{V_{spheres} + V_{bound}^{H_2O}}{V_{tot}} = \phi + \frac{V_{bound}^{H_2O}}{V_{tot}}$$

$$V_{bound}^{H_2O} = (\phi_{real} - \phi)V_{tot}$$

$$p_{bound} = \frac{\phi_{real} - \phi}{1-\phi}$$

$$D_{obs} = \frac{1}{1+\frac{\phi}{2}}\left(1 - \frac{\phi_{real} - \phi}{1-\phi}\right)D_0 = \frac{1}{1+\frac{\phi}{2}}\left(\frac{1-\phi_{real}}{1-\phi}\right)D_0$$

From the last equation above, which is Equation 3 in the main article, ϕ_{real} can be derived.

$$\phi_{real} = 1 - \frac{D_{obs}}{D_0}\left(1 - \frac{\phi}{2} - \frac{\phi^2}{2}\right)$$

We can now express the bound water layer thickness ($l_{bond}^{H_2o}$) at the silica sphere surface.

$$\frac{\phi_{real}}{\phi} = \frac{\frac{V_{spheres} + V_{bound}^{H_2O}}{V_{tot}}}{\frac{V_{spheres}}{V_{tot}}} = \frac{V_{spheres} + V_{bound}^{H_2O}}{V_{spheres}} = \frac{n_{spheres}\left(V_{sphere} + V_{bound,sphere}^{H_2O}\right)}{n_{spheres}V_{sphere}} = \frac{\left(r_{sphere} + l_{bound}^{H_2O}\right)^3}{r_{sphere}^3}$$

$$l_{bound}^{H_2O} = r_{sphere}\left(\left(\frac{\phi_{real}}{\phi}\right)^{\frac{1}{3}} - 1\right) = r_{sphere}\left(\left(\frac{1 - \frac{D_{obs}}{D_0}\left(1 - \frac{\phi}{2} - \frac{\phi^2}{2}\right)}{\phi}\right)^{\frac{1}{3}} - 1\right)$$

where n_{sphere} and r_{sphere} is the number and radius of the silica spheres, respectively.

References

1. P. G. Nilsson and B. Lindman, *The Journal of Physical Chemistry*, 1983, **87**, 4756-4761.
2. G. Bell, *Transactions of the Faraday Society*, 1964, **60**, 1752-1759.