SUPPLEMENTARY INFORMATION

On the dual behaviour of water in octanol-rich aqueous n-octanol mixtures: an x-ray scattering and computer simulation study

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1) Diffraction pre-peak analysis

Fig.SI-1 and Fig.SI-2 show the pre-peak intensities and positions, respectively, as obtained from a direct analysis of Fig.2 and Fig.3 of the main manuscript.

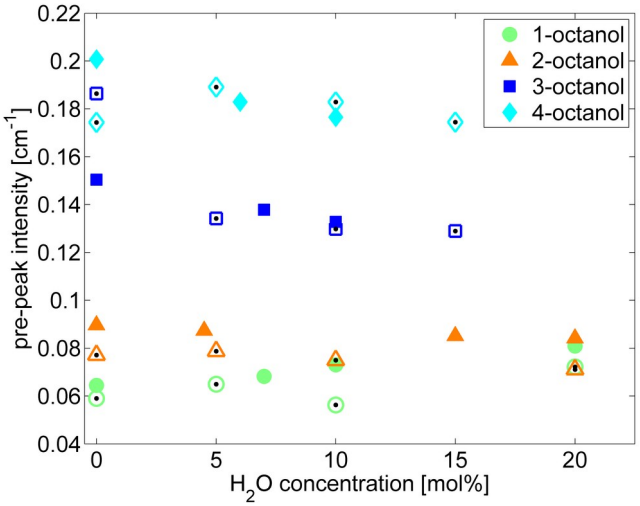

Fig.SI-1: Pre-peak intensities of the experimental (closed symbols) and simulation (open symbols) data from Fig.2 and Fig.3.

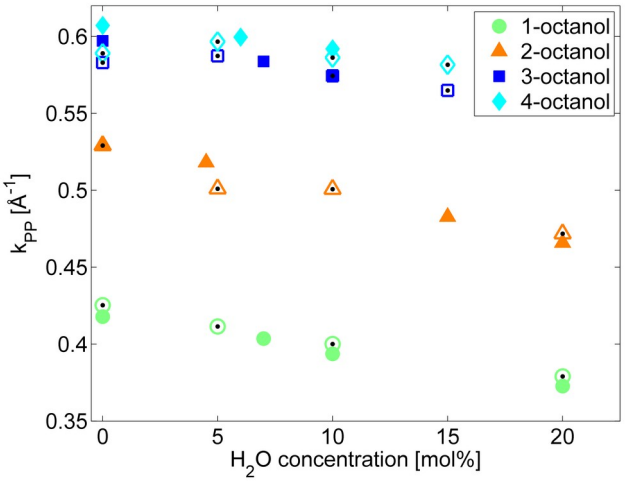

Fig.SI-2: Pre-peak positions (symbols as in Fig.SI-1)

These 2 figures show the near excellent agreement between the experimental and simulated data,

taking into account that the cluster formation should, in principle, be very model dependent. This can be seen through the main peaks positions in Figs.2 and 3, which reflect the mean atom sizes. The type of agreement observed in Fig.SI-1 and 2 suggest that the aggregates have little model dependence, despite differences in sizes between the real and model atoms.

2) Structure factors for 1-octanol

The following figure shows all the structure factors (Eq.3 of main text) for 1-octanol (inset) with typical of them highlighted in the main panel. The main purpose is to illustrate the various types of contributions to the pre-peak and main in the scattering intensity Eq.(1) in the main text. These concern positive pre-peak and negative anti-peak contributions around $k \approx 0.45\ \text{Å}^{-1}$ and the positive main peak contributions around $k \approx 1.5\ \text{Å}^{-1}$

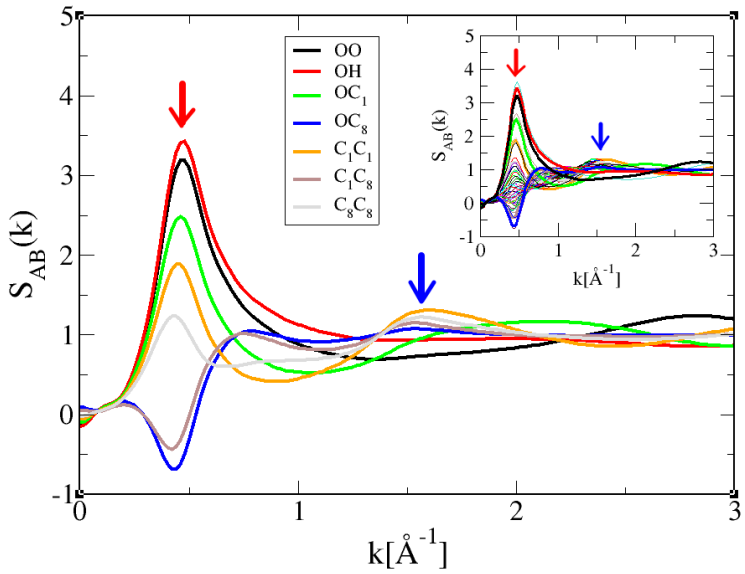

Fig.SI-3: Typical structure factors $S_{AB}(k)$ contributing to positive pre-peak and negative anti-peaks (marked by the red arrow) and positive main main peak (marked by the blue arrow)

1) Cluster distributions

The cluster distributions for 2-octanol and 3-octanol, equivalent of those for 1-octanol (Fig.5) and 4-octanol (Fig.6) in the main text, are shown in the figures below, Fig.SI-4 and Fig.SI-5, respectively.

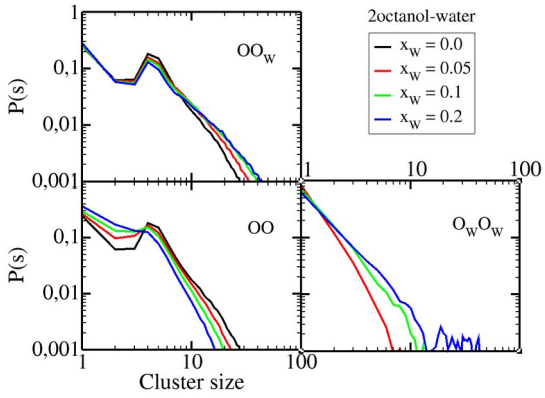

Fig.SI-4: Oxygen atom clusters size distributions for 2-octanol and for different water content

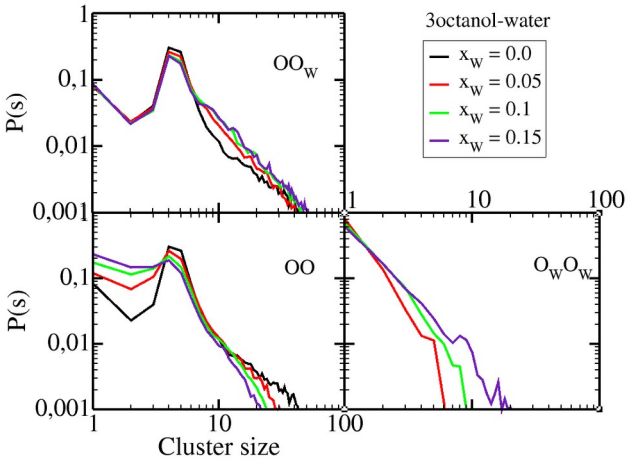

Fig.SI-5: Oxygen atom clusters size distributions for 3-octanol and for different water content

2) Pair correlations and structure factors

The various oxygen-oxygen atom pair correlation functions and corresponding structure factors for 2-octanol are shown below in Fig.SI-6 to Fig.SI-8.

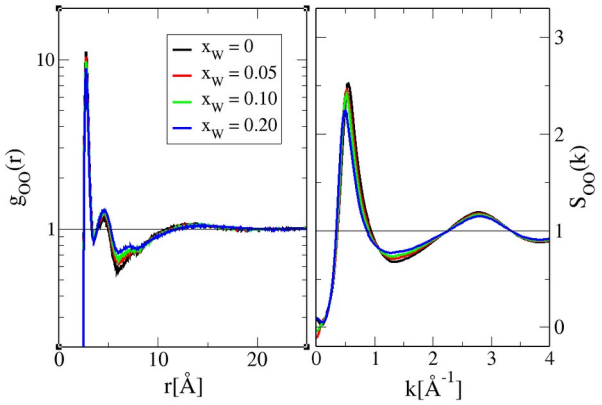

Fig.SI-6: Octanol oxygen-oxygen pair correlation function g_{OO} (r) for 2-octanol (left) and structure factor S_{OO}(k) (right), and for various water mole fractions as shown in the legend panels. The pure octanol curves (x_W = 0) are shown in black. Note that the vertical scale for the g_{OO} (r) is logarithmic.

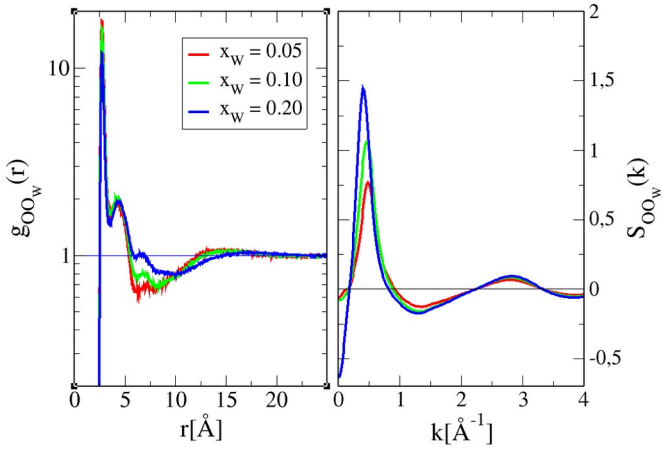

Fig.SI-7: same as Fig.SI-4, but for the cross OO_W octanol and water oxygen atoms.

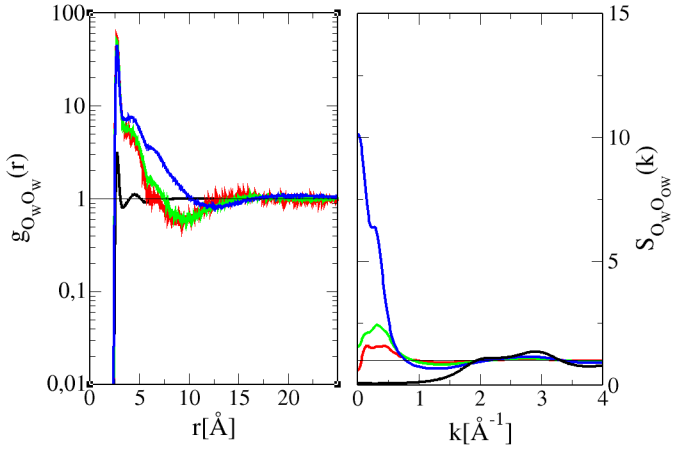

Fig.SI-8: same as Fig.SI-4, but for the O_WO_W water oxygen atoms.

The various oxygen-oxygen atom pair correlation functions and corresponding structure factors for 3-octanol are shown below in Fig.SI-9 to Fig.SI-11.

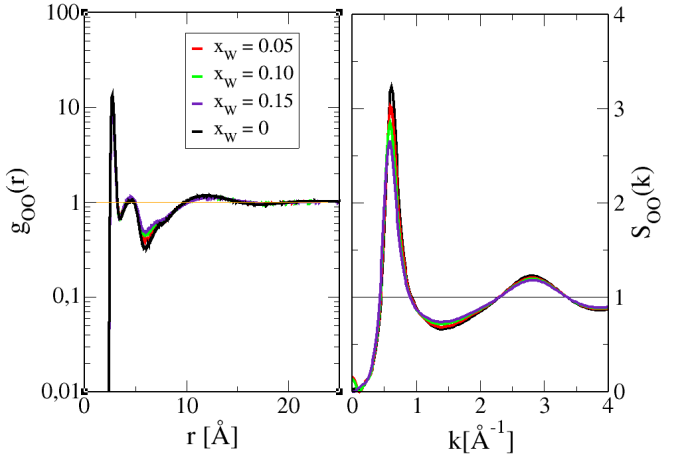

Fig.SI-9: Octanol oxygen-oxygen pair correlation function g_{OO} (r) for 3-octanol (left) and structure factor S_{OO}(k) (right), and for various water mole fractions as shown in the legend panels. The pure octanol curves (x_W = 0) are shown in black. Note that the vertical scale for the g_{OO} (r) is logarithmic.

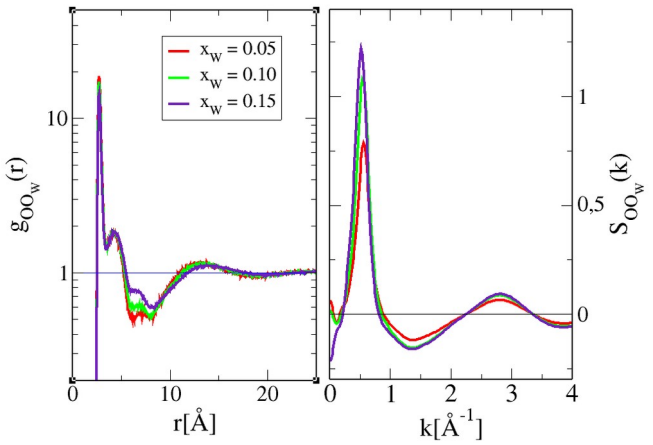

Fig.SI-10: same as Fig.SI-4, but for the cross OO_W octanol and water oxygen atoms.

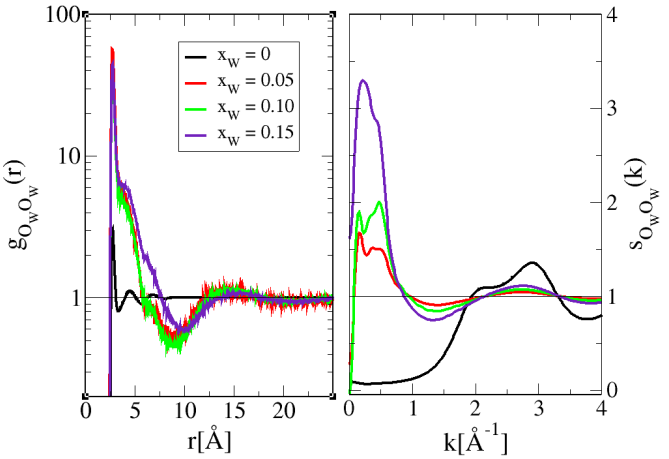

Fig.SI-11: same as Fig.SI-3, but for the O_WO_W water oxygen atoms.