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# High-q, High Intensity Small Angle Neutron Scattering to Probe Formaldehyde-Methanol-Water Mixtures

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Methanol-water mixtures are known for their unusual thermodynamic behaviour. On varying mixture composition, the thermodynamic properties do not vary linearly. This is attributed to the formation of structures at a molecular length scale, called as micro-phase. When formaldehyde is solvated in methanol-water mixtures, its chemical and physical behaviour is very much dependent on its micro-phase environment. Recently, liquid phase heterogeneous catalytic routs for the production of formaldehyde and its higher order oligomers are being developed1,2. The liquid phase (generally, methanol-water mixture) increases formaldehyde's yield after its desorption from the catalytic surface1. Therefore, the study of formaldehyde's solvation in methanol-water mixtures may be crucial to further develop these liquid phase catalytic reaction pathways. However, the understanding of the structure of formaldehyde–methanol-water mixtures at molecular length scales is a challenge to the contemporary experimental techniques due to their dynamical and chemical nature. We use molecular dynamics simulations and the Small Angle Neutron Scattering (SANS) measurements to predict the molecular clustering in these mixtures.

Classical Molecular Dynamics (MD) simulations were performed using GROMACS software package3 and the OPLS-AA forcefield parameters were used to describe bonded and non-bonded interactions. The radial pair distribution function g(r) and the coordination number were used to estimate the cluster composition and to compose backgrounds for these ternary mixtures. The Neutron Scattering data was collected at the Quokka beamline of the Australian Nuclear Science and Technology Organisation (ANSTO). The data modelling program SASview was used to model the scattering data and five different curve-fitting models were used, namely, the Guinier model, sphere model, sticky-hardsphere (SHS) sphere model, and SHS ellipsoid model. The sticky-hardsphere model fitting parameters were derived from the Potential of Mean Force (PME), calculated by the MD simulations.

We observe a hydrophobic clustering of methanol around methoxymethanol molecule (i.e., the metastable solvated form of formaldehyde) at formaldehyde–methanol-water mixtures where 1 mole-percent formaldehyde is dissolved in $x_m \leq 0.3$ methanol-water mixture. The SHS-sphere model results in a sphere of 4.29 Å radius, which, when drawn from the centroid of a molecular cluster obtained via MD data, perfectly encapsulates it. On further increasing the methanol concentration, we do not observe any molecular clusters for $x_m > 0.5$. In summary, we formulate a framework of analysing the dynamic ternary liquid mixtures for molecular clustering using SANS measurements and MD simulations and report hydrophobic clustering in formaldehyde-methanol-water ternary mixtures at low methanol composition.

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## Speakers Gender

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## Level of Expertise

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## Do you wish to take part in the poster slam

Yes

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