Experimental report

23/01/2025

Proposal: 6-02-647 **Council:** 10/2023

Title: The structure of liquid cyrene andcyrene aqueous solutions.

Research area: Physics

This proposal is a new proposal

Main proposer: Simone DE PANFILIS

Experimental team:

Local contacts: Anne STUNAULT

Samples: Dihydrolevoglucosenone + water (C6H8O3 + H2O)

Instrument	Requested days	Allocated days	From	To
D3	6	3	22/11/2023	24/11/2023
			07/12/2023	08/12/2023

Abstract:

Dihydrolevoglucosenone, commercially known as Cyrene, is a novel polar aprotic solvent with excellent environmental properties: it is non-toxic, non-mutagenic and "green" both during production and after disposal. It is soluble in water and it is seen to substitute more hazardous solvents (such as NMP, DMF, sulfane, etc) in several industrially-relevant applications, like in pharmaceutics and agrochemistry. Still, very little if nothing is known of its microscopic structure in the liquid state and in aqueous mixtures. This is particularly relevant to assess if and how it interacts with water in biological systems. To shed light on these fundamental properties, we intend to collect high quality neutron diffraction measurements of pure dihydrolevoglucosenone and of its aqueous solutions. As deuterated dihydrolevoglucosenone is not available, we will exploit the polarized detection capabilities of D3 to separate the incoherent and coherent scattering and measure the static structure factor even for fully hydrogenated samples.

ILL EXPERIMENTAL REPORT

Proposal: **6-02-647**

Title: **The structure of liquid Cyrene and Cyrene aqueous solutions.**

Experiment team: Simone DE PANFILIS, Miguel Angel GONZALEZ, Ferdinando FORMISANO

Local contact: Anne STUNAULT

Instrument: D3

Dihydrolevoglucosenone, commercially known as Cyrene, is a novel polar aprotic solvent with excellent environmental properties: it is non-toxic, non-mutagenic and "green" both during production and after disposal. It is soluble in water and it is seen to substitute more hazardous solvents (such as NMP, DMF, sulfane, etc.) in several industrially-relevant applications, like in pharmaceutic agrochemistry. Still, very little if nothing is known of its microscopic structure in the liquid state and in aqueous mixtures. This is particularly relevant to assess if and how it interacts with water in biological systems. To shed light on these fundamental properties, we collected high quality neutron diffraction measurements of pure dihydrolevoglucosenone and of its aqueous solutions. As deuterated dihydrolevoglucosenone is not available, we exploited the polarized detection capabilities of D3 to separate incoherent and coherent scattering and measure the static structure factor even for fully hydrogenated samples.

Spin-flip and non-spin-flip angle resolved scattering intensity were collected at ambient conditions. Samples were loaded in annular vanadium sealed cells optimized for the D3 instrument. A single crystal was mounted on top of the sample container and cyclically inserted into the neutron beam to monitor the efficiency of the ^{3}He polarized spin filters during the measurements. In order to collect data over a rather large exchanged momentum Q range (0.6 to 20 Å^{-1}), we chose to probe each sample at two different neutron wavelength of 0.5 and 0.83 Å. Each single measurement was repeated twice, both to increase statistics and to check for any possible systematic error due to data collection and processing. In fig. 1 we show raw spin-flip and non-spin-flip data (left panel) and calculated coherent and incoherent scattering data (right panel) for pure H_2O.

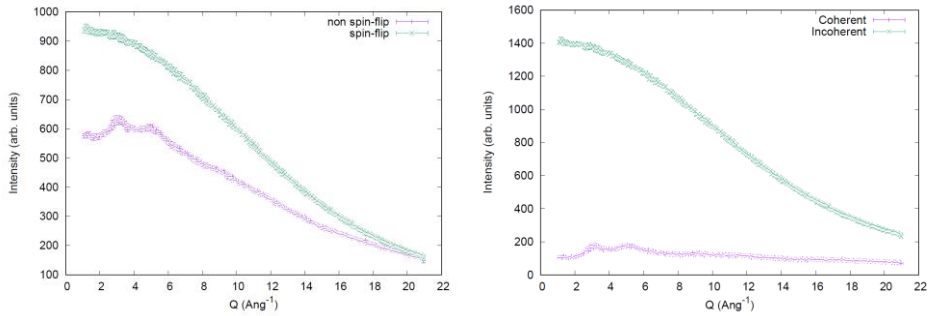

Fig. 1: Intensity of elastic scattered neutrons at 0.5 Å from pure water as a function of the exchanged momentum *Q*. Left panel, raw spin-flip and non-spin-flip channels data. Right panel, coherent and incoherent scattering intensities as derived from the raw data.

Raw data were corrected and reduced to coherent and incoherent scattering intensity through an established procedure [1,2]. The resulting coherent scattering intensities for pure Cyrene and fully hydrogenated Cyrene-water mixtures at selected concentrations are shown in Fig. 2. A continuous trend of the main features can be easily observed. Further analysis of these datasets will provide detailed information on the microscopic structure in Cyrene-water mixtures.

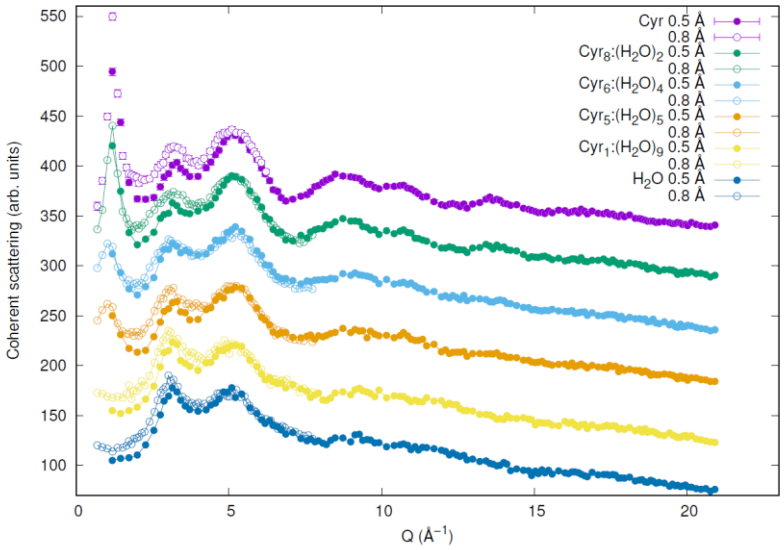

Fig. 2: Coherent scattering signals for fully hydrogenated Cyrene-water mixtures and pure moieties measured on D3, at both neutron wavelengths.

The possibility to extract the pure coherent contribution from fully hydrogenated liquids is a unique capability of the D3 instrument, and these results motivate us in further probing the static structure of hydrogenated systems, possibly in a wider range of the phase diagram.

[1] A. Stunault, S. Vial, D. Jullien and G.J. Cuello, *Physica B* 373-376, **551** (2018).
[2] L. Temleitner, L. Pusztai, G.J. Cuello and A. Stunault, *J. Mol. Liq.* 118535, **350** (2022).