

MID-IR INVESTIGATION OF SOLVENT CLUSTERING AROUND SOLUTES IN CO₂-EXPANDED ORGANIC SOLUTIONS

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CO₂-expanded solvents can be used to generate a continuum of media offering endless opportunities for chemical reaction and material processing design ^{1,2}.

Here we show the feasibility of exploring the solvating properties of CO₂-expanded solvents from infrared spectral shifts measurements. Mid-IR spectroscopy has been used to understand at the molecular level the difference in the solubility behaviour of acetaminophen in “CO₂-expanded acetone” and in “CO₂-expanded ethanol” and to assess the higher anti-solvent character of CO₂ over the system acetaminophen-acetone than in acetaminophen-ethanol.

The evolution of some acetaminophen vibrational modes frequency with the nature and composition of the CO₂ expanded solvent has been found to provide a valuable insight about solvent-solute interactions and clustering phenomena. These experimental results have been interpreted from ab-initio calculations and an analytical molecular solvation model to assess the nature of the intermolecular interactions between acetaminophen and the solvent molecules.

We have proved that the dependence of the solubility in a CO₂-expanded solvent of a given compound with the solvent composition can be qualitatively and quantitatively predicted from mid-IR spectroscopic measurements in diluted solutions.

Finally, infrared studies of CO₂-acetone and CO₂-ethanol mixtures have also been done in order to characterise the solvent-solvent and solvent-CO₂ interactions responsible of the variation of the solvent media cohesiveness on going from the pure organic solvent to the neat CO₂.

¹ P.M.Gallagher, M.P. Coffey, V.J. Krukonis, N. Klasutis, *American Chemical Symposium Series 406*, American Chemical Society: Washington, D.C., **1989**.

²M. Wei, G.T. Musie, D.H. Busch, B. Subramaniam, *J.Am.Chem.Soc.* **2002**, *124*, 2513-2517.