

実験報告書様式 (一般利用課題・成果公開利用)

(※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。)

 	承認日 Date of Approval 2015/1/4 承認者 Approver Jun-ichi Suzuki 提出日 Date of Report 2014/5/13
課題番号 Project No. 2014A0003 実験課題名 Title of experiment Aggregation and micro-heterogeneities in alcohol-water mixtures 実験責任者名 Name of principal investigator PUSZTAI, László 所属 Affiliation Wigner Research Centre, Hungarian Academy of Sciences	装置責任者 Name of Instrument scientist TAKATA, Shin-ichi 装置名 Name of Instrument/(BL No.) TAIKAN (BL-15) 実施日 Date of Experiment 2014.05.09-2014.05.12

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)
 Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Deuterated methanol-heavy water mixtures, (CD₃OD)_x : (D₂O)_{100-x}; x: 0,10,20,40,60,70,80,100
 Deuterated ethanol-heavy water mixtures, (C₂D₅OD)_x : (D₂O)_{100-x}; x: 20,40,60,80,100
 Deuterated 2-propanol-heavy water mixtures, (2-C₃D₇OD)_x : (D₂O)_{100-x}; x: 20,40,60,80,100
 Deuterated 1-propanol-heavy water mixtures, (1-C₃D₇OD)_x : (D₂O)_{100-x}; x: 18,100
 Deuterated t-butanol-heavy water mixtures, (C₄D₉OD)_x : (D₂O)_{100-x}; x: 20,50,80,100
 All samples were liquids.

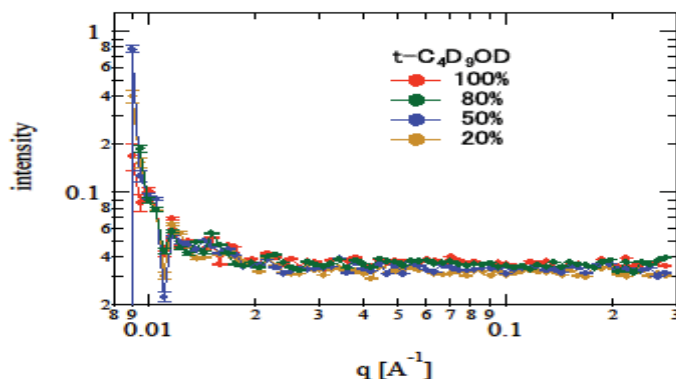
2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。)

Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

The liquid samples were contained in 2 mm thick quartz sample holders ('cuvettes'). We have used the TAIKAN instrument (installed on BL-15) for measuring the total scattering signal over a wide momentum transfer (Q-)range, including much of the small angle scattering region (down to about 0.01 Å⁻¹). Auxiliary measurements on the empty sample holder, the empty instrument and on a standard sample have also been carried out, for facilitating data correction. Each sample received 2 hours of beamtime on average, which proved to be providing reasonable statistical accuracy up to at least 15 inverse angstroms. Below some examples of the corrected total scattering signals are provided. Since the present experiment was focused on the small angle scattering part of the signal, only representative examples for that region are shown.

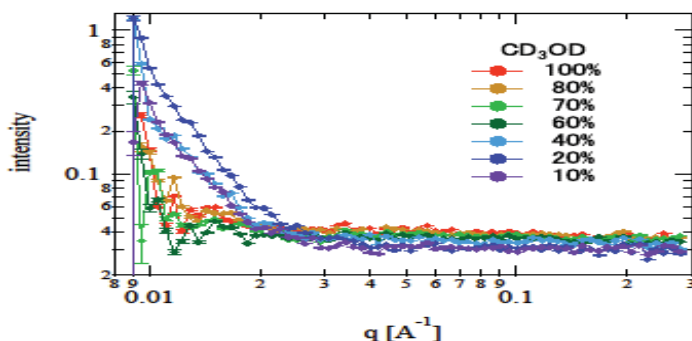
The picture below shows the small angle scattering region, between about 0.01 and 0.4 Å⁻¹, of (fully deuterated) tertier-butanol/(heavy) water mixtures, as measured by the small angle detector bank. The intensity at the smallest Q values should be considered as residual scattering from the direct beam; that is, no SANS signal could be detected for these samples.

2. 実験方法及び結果(つづき) Experimental method and results (continued)



This is somewhat surprising in the light of earlier synchrotron X-ray diffraction results [1], which indicated that for the 50–50 mixture, there could be a strong SAS signal. Note, however, that the X-ray data were only reliable down to about $(0.2\text{--}0.3) \text{\AA}^{-1}$, so no real SAS could be measured there.

On the other hand, the figure below shows the same information for (deuterated) methanol/(heavy) water mixtures. Clear SANS contributions are apparent on samples containing 10, 20 and 40 molar % of methanol. The SANS seems to have a maximum, as a function of the composition, at 20 % methanol content. These findings are most intriguing and call for further, more detailed investigations (a) for the precise determination of the composition of the SANS maximum, and (b) for a temperature dependent study over the entire composition range to see whether lowering T would result in an enhanced level of aggregation (as might be expected).



In summary we have determined the total scattering structure factors, including substantial portions of the small angle scattering region, of 20+ alcohol–water mixtures in the liquid state. Only very few of the datasets show clear signs of larger–scale structural features; notably, these all are methanol–water mixtures on the water–rich side of the composition map. This finding is fundamentally different from earlier results, e.g. of Dixit et al. [2], who reported the presence of micro–heterogeneities on the alcohol–rich side. The data measured on TAIKAN (at J-PARC) are planned to be complemented by similar information from X-ray diffraction/small angle scattering.

[1] Temleitner L, private communication on his SPring-8 experiments (2013)

[2] Dixit S, Crain J, Poon WCK, Finney JL, Soper AK. Nature 416, 829 (2002)