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。 ）

CROSS TOKAI Experimental Report J-PARC	承認日Date of Approval 2013/11/23 承認者Approver Jun-ichi SUZUKI 提出日Date of Report 2013/02/28
課題番号 Project No. 2012B0080 実験課題名 Title of experiment Effects of solvents on hierarchical structure of poly(vinyl alcohol) gels 実験責任者名 Name of principal investigator Toshiji Kanaya 所属 Affiliation Kyoto University	装置責任者 Name of responsible person Jun-ichi Suzuki 装置名 Name of Instrument/(BL No.) TAKAN 実施日 Date of Experiment 2012/12/21-25

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.

poly(vinyl alcohol)/heavy water /deuterated dimethy sulphoxide

-(CH2CH(OH))n-/D2O/(CD3)2SO (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.

We have investigated the PVA gels in mixtures of DMSO and water for a long time to reveal the relationship between the hierarchic structure and the properties using various scattering methods in a wide q range. In the previous studies we focused on the hierarchic structure of PVA gels in a mixture of DMSO and water (60/40), but not as a function of the solvent composition because it took too long time to perform the measurements in a wide q range using various scattering methods and impossible to do the time-resolved measurements. This is the present status of the work on the PVA gels. In addition, it was hard to see the structure of the PVA gels and the aggregates of DMSO and water simultaneously. This study is very important because we can revel the effects of solvent on the hierarchic structure of PVA gels. Now the research situation has changed because TIKAN (BL15) is available in J-PARC/MLF. We can now study the PVA gel structure and the structure of aggregates of DMSO and water simultaneously by a time-resolved measurement.

In the present study we did time-resolved measurements on poly(vinyl alcohol) in mixtures of deterated DMSO and heavy water with various compositions. The sample solution was prepared in a quartz cell at ~100°C to obtain a homogeneous solution before the measurements. Then the solution was cooled down to room temperature (25 °C) and quickly transferred to a sample changer in TAIKAN.

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

Similar sample preparation (preparation of homogeneous solution and quenching to RT) was repeated for five PVA solutions with different DMSO/water compositions (40/60, 50/50, 60/40, 70/30, 76/24). The gelation times of the samples depend on the solvent compositions, and the PVA solution in 76/24 composition has the longest gelation time (~3 days) and hence this sample was prepared first of all. The samples thus prepared were transferred to the sample changer in TAIKAN, and then time-resolved measurements were started on the samples. Solvent scattering, empty cell and background measurements were done after the sample measurements.

The time evolution of scattering intensity of the solution with the DMSO/water composition of 60/40 is shown in Figure 1.

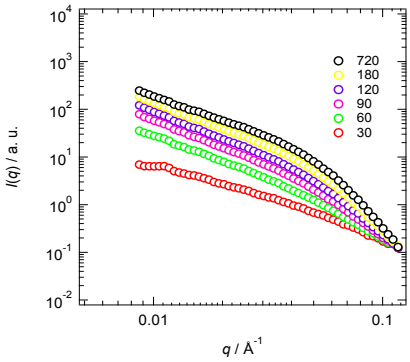

Figure 1. Time evolution of the scattering intensity of the PVA solution in a mixture of DMSO/water (60/40). Numbers in the figure are the annealing times.

These data were corrected for the solvent, empty cell and neutron transmittance. It was found that the data could be well described by the OZ formula in a q range below 0.04 Å^{-1}, then we have evaluated the correlation length ξ of the solution. The time evaluation of the correlation length is shown in Figure 2 for the five PVA solutions with different DMSO/water compositions (40/60, 50/50, 60/40, 70/30, 76/24) with the gelation times t_g. As seen in the Figure 2, the correlation length shows a maximum at around the gelation time and decreases with the annealing time to reach a final value in the gel. In the gel, it was revealed in a previous study, the cross-linking points were crystallites. The correlation length in the gel must be governed by the correlation between the crystallites because the correlation is very strong. In Figure 3, the final correlation length in the gel is shown as a function of the DMSO fraction. It decreases with increasing with the DMSO fraction. It is interesting to point out that the this DMSO fraction dependence agrees with the tensile strength of films prepared from the PVA gel, suggesting that the distance between the crystallites in the gels is one of the important factors to determine the strength of the films prepared from gels.

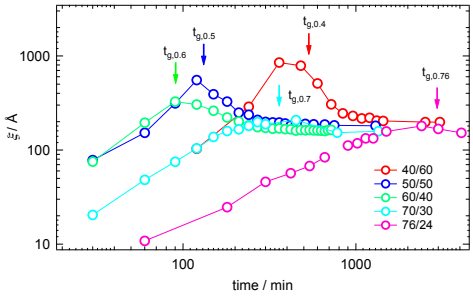

Figure 2. Time evaluation of the correlation length for the five PVA solutions with different DMSO/water compositions (40/60, 50/50, 70/30, 76/24) with the gelation times t_g.

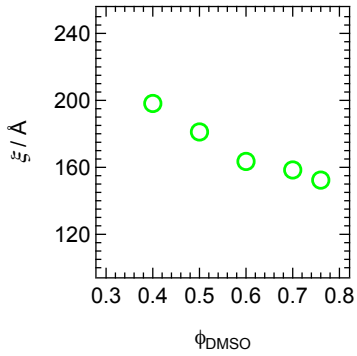

Figure 3. Correlation length as a function of DMSO fraction.