（ ※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。 ）

MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2013B0016 実験課題名 Title of experiment Studies on Dynamcs of Highly Crosslinked Rubber by μSR 実験責任者名 Name of principal investigator Toshiji Kanaya 所属 Affiliation Institute for Chemical Research, Kyoto University	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) D1 実施日 Date of Experiment 2014/3/3 10:00 ～ 2014/3/5 10:00

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.

Materials and sample preparation

The details of materials used in the present study are summarized in Table 1. We used polybutadiene (PB) as a matrix rubber. Zinc diacrylate (ZDA) and dicumyl peroxide (DCP) were used as a crosslinking agent and a polymerization initiator of butadiene, respectively. The components were mixed on a 6-inch two-roll mill, and they were molded into sheet 1 mm thick at 170 ℃ for 20 min. The glass transition temperature (T_g) of Sample A and B are 168.6 K and 168.1 K, respectively. The Elastic modulus at 293 K of Samples A and B are 4.0 MPa and 104.6 MPa, respectively.

Table 1.: Detail of components of samples used in present study.

	Sample A	Sample B
Polybutadiene / vol%	99.2	83.6
Zinc diacrylate / vol%		15.7
Dicumyl peroxide / vol%	0.8	0.7
Elastic modulus / MPa	4.0	104.6

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.

Measurements

The Longitudinal field (LF) μSR experiment was performed using a beamline D1 instrument installed at J-PARC in the temperature range between 30 and 270 K. External magnetic field is 100G. Each sample is set onto the sample holder. The obtained raw asymmetry data were corrected for the instrumental constant and for the backgrounds from the sample holder.

Results

The LF-μSR spectra of Samples A and B are shown in Fig. 1 and 2, respectively. We used the simple stretched exponential fitting function (1) as first trial.

$$f_{(t)} = A\exp\left(-(t/\tau)^{\beta}\right) + B \qquad (1)$$

where τ is relaxation time, β is the stretch parameter, A and B are the fitting parameters. Fig. 1 and 2 show the observed and calculated spectra agree fairly well.

The obtained relaxation rate ($1/\tau$) of Samples by fitting is shown in Fig. 3. The relaxation rates of both Samples are almost constant and same value below T_g and rise monotonically with increasing temperature above T_g. This result is almost same as previous study for non-crosslinked PB[2]. The dynamics of muon, therefore, is thought to correspond to the dynamics of crosslinked polymer because the behavior of the relaxation rate of Samples corresponds to the behavior of crosslinked polymer glass transition. The reason why the relaxation rates of Samples are constant and same below T_g is thought that the micro-Brownian motion of polymer chain is frozen below T_g. On the other hand, the relaxation rates rise monotonically with increasing temperature above T_g, suggesting that the frozen micro-Brownian motion of polymer chain is thawed and the polymer chain move actively with increasing temperature. Moreover, the relaxation rate of Sample A is higher than that of Sample B above T_g, relating to the elastic modulus of Sample A is lower than that of Sample B. So that means in the case of Sample B, the polymer chain motion is limited because of crosslinking with strong crosslinker ZDA, and the relaxation rate is lower than that of Sample A crosslinked without ZDA.

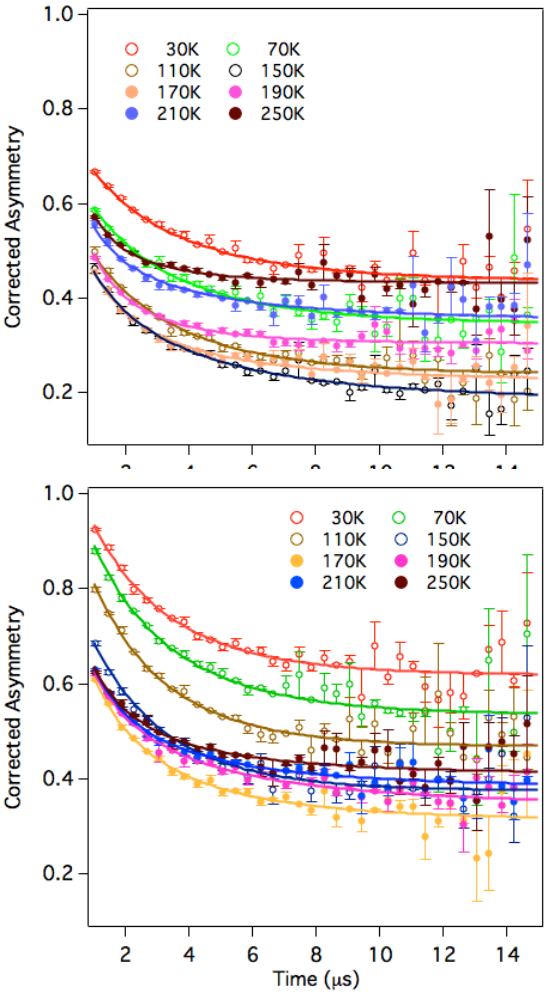

Fig. 2.: LF-μSR spectra of Sample B.

[1] R. Mashita *et al.*, Polym. J. 45(2013)57.

[2] F. L. Pratt *et al.*, Physica B 289-290(2000)625.

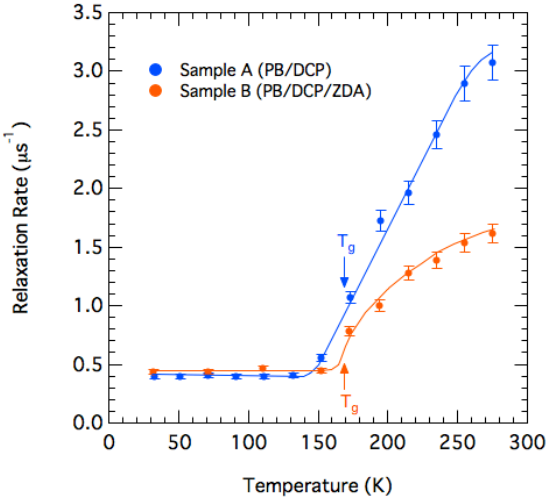

Fig. 3.: Temperature dependence of the relaxation rate ($1/\tau$) of Sample A and B.