MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2015A0107 実験課題名 Title of experiment Systematic studies on hierarchic structure and dynamics in model rubber materials: heterogeneous local dynamics in rubbers 実験責任者名 Name of principal investigator Toshiji Kanaya 所属 Affiliation High Energy Accelerator Research Organization (KEK)	装置責任者 Name of responsible person Kenji Nakajima 装置名 Name of Instrument/（BL No.） BL-14 実施日 Date of Experiment 2016/3/28 ～ 2016/4/1

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.
Polybutadiene rubbers (-$(C_4H_6)_n$-) including silica filler (SiO_2) (volume fraction of 0, 0.2) were used for sample and the thickness es of prepared samples were at around 0.2mm.

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.

Neutron scattering studies are now looking towards realistic materials (or industrial materials) in addition to traditional scientific researches on simple model materials. In soft matter, intensive studies are now performed on industrial rubber materials such as mobile tire and rubber base-isolated structure. However, generally speaking, the realistic rubber materials have complicated structure and dynamics in wide spatial and time scales, which are dominated by many factors such as fillers, aggregations of fillers, crosslinks between polymers, coupling agents between filler and polymer. Hence, it is not easy to interpret the complicated results or extract useful information for industries from the results.

To understand the data on the realistic (industrial) complicated rubber materials and to fill up the gap of studies between the realistic (industrial) and ideal materials we propose systematic studies on some model rubber materials by means of various kinds of quantum beams. In this experiment, we focus on polybutadiene (PB) including silica powder fillers with no crosslinks using BL14 at J-PARC to see the local heterogeneous dynamics.

Dynamic scattering laws $S(Q, \omega)$ in the range Q = 1.8 Å^{-1} at 300 K are shown in Fig.1. The narrowing of $S(Q, \omega)$ by an addition of silica can be observed with an energy resolution of 0.35 meV, suggesting that the motion of PB is slowed down by an addition of silica. To understand this result in detail, we adopted the Kohlrausch-Williams-Watts (KWW) function to describe the observed $S(Q, \omega)$, and the following model

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

function was proposed.

$$S(Q,\ \omega) = A\left((1-EISF)\mathcal{F}\left\{\exp\left[-(t/\tau)^{\beta}\right]\right\} + EISF\,\delta(\omega)\right) \otimes R(Q,\ \omega) \quad (1)$$

where *A*, *EISF*, τ, and β are the amplitude of the relaxation function, the elastic incoherent structure factor, the relaxation time, and the distribution of the relaxation function, respectively. It should be noted that the KWW function is utilized in the time domain, necessitating that we adopt the Fourier-transformed KWW function ($\mathcal{F}$(KWW)) in the frequency region. $R(Q, \omega)$ indicates the instrumental resolution function. Fig. 1 shows the results of the fits to $S(Q, \omega)$ with Eq. (1) at Q = 1.8 Å^{-1} and 300 K. The $S(Q, \omega)$ from both samples are fitted fairly well by Eq. (1), confirming the appropriate model function selection.

The dependence values of the average relaxation time $<\tau>$ and the relaxation time distribution β at 300 K in the range Q = 1.8 Å^{-1} on the volume fraction of silica are shown in Fig. 2. $<\tau>$ was calculated using Eq. (2)[1]: where Γ is a gamma function.

$$\langle\tau\rangle = \left(\frac{\tau}{\beta}\right)\Gamma\left(\frac{1}{\beta}\right) \quad (2)$$

The increase of $<\tau>$ and *EISF* in the silica volume fraction were evaluated, suggesting that the motion of mobile component is slowed down and the fraction of immobile component is increased with increases in the silica volume fraction. The detailed analysis is still on progress.

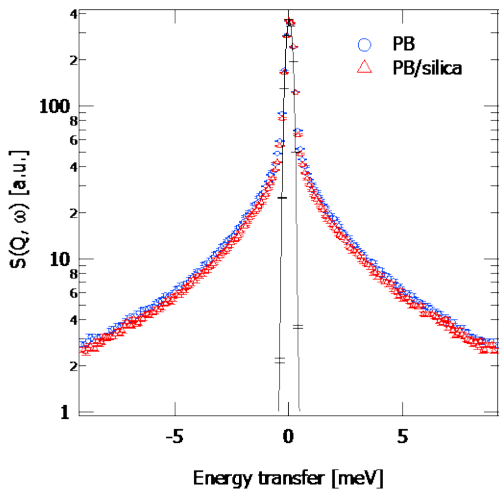

Fig. 1 Dynamic scattering laws $S(Q, \omega)$ normalized at the peak top at 300 K in the range Q = 1.8 Å^{-1}. The solid line is the instrumental resolution function.

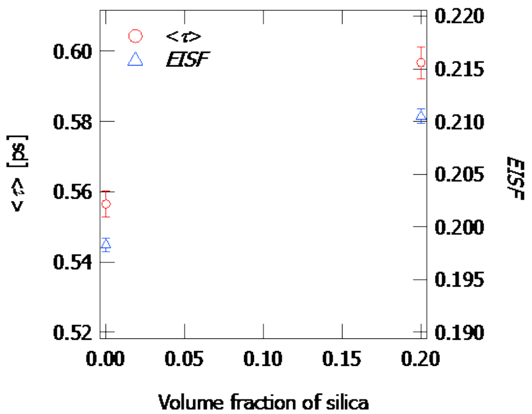

Fig. 2 Volume fraction of silica dependence of $<\tau>$ and *EISF* at 300 K in the range Q = 1. 8 Å^{-1}.

Reference

[1] F. Alvarez et al., J. *Phys. Rev. B* **44**, 7306 (1991).