MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2014B0012 実験課題名 Title of experiment Density Profiles of Porous Diamond-like Carbon 実験責任者名 Name of principal investigator 藤井 義久 所属 Affiliation (国)物質・材料研究機構	装置責任者 Name of responsible person 山田悟史 装置名 Name of Instrument/(BL No.) SOFIA / BL16 実施日 Date of Experiment 2014/12/18 - 2016/03/31

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.

Name of samples: porous diamond-like carbon (DLC)

Chemical formula: CxHy

Figure 1 shows the structural image of our porous diamond-like carbon (DLC).

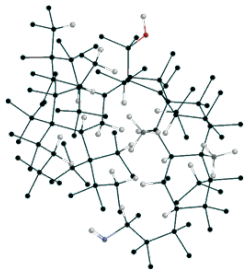

Figure 1. Structural image of DLC.

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.

Experimental method

Diamond-like carbon DLC membranes are prepared from various precursor gases onto thick silicon blocks with a size of 40 mm × 40 mm × 7 mm by CVD method. The thickness of the membranes in a dried state is controlled to be approximately 70 nm which is appropriate for NR measurements. To evaluate the intrinsic density profile of these porous DLC, membranes are aged in deuterium oxide (D_2O) or deuterated ethanol (C_2D_6O) for a given time, which is sufficiently enough to induce contrast change by sorption of deuterated liquids. Density profiles of the films along the direction normal to the surface in contact with deuterated liquids are examined by NR measurements. The neutron beams are guided into the film from the silicon side, and the reflected beam is detected under the specular condition.

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

Results

Figure 1a shows the reflectivity curves as a function of the scattering vector ($q = (4\pi/\lambda)\sin\theta$, where λ and θ are the wave length and the incident angles of the neutron beams, respectively) for graded composition and uniform composition. For clarity, the data set for the uniform composition film is offset by two decade. The open symbols denote the experimental data, and the solid curves were fit curves calculated from the scattering length density (b/V) profiles in Figure 1b and 1c. Since the solid curves in Figure 1a are in good agreement with the experimental data, it is most likely that the model (b/V) profiles drawn in Figure 1b and 1c would reflect the density profiles of the films. The (b/V) values for silicon (Si), and native oxide layer (SiO_x) were 2.21×10^{-4} and 4.18×10^{-4}, respectively. In case of uniform composition film (Fig.1c), the (b/V) values was constant over the entire region of the film. On the other hands, in the graded composition film (Fig. 1b), the (b/V) values was gradually increased from the silicon substrate to the outermost surface. This indicates that film density is increased with increasing film thickness. Another possibility of this profile might come from the composition gradient of the hydrogen atom which has negative neutron scattering length. To clarify the origin of the gradient (b/V) profile, we also carried out the Time-of-Flight Secondary Mass Spectrometry (TOF-SIMS) analysis to the same film. Figure 2 shows the TOF-SIMS profile of the carbon film. Panel a ~ c of Figure 2 indicates that the "H" contents increased with increasing the depth even so the "C" content is constant over the entire region of the film.

These structural difference affect the filtration properties. The details will be discussed with flux and rejection properties.

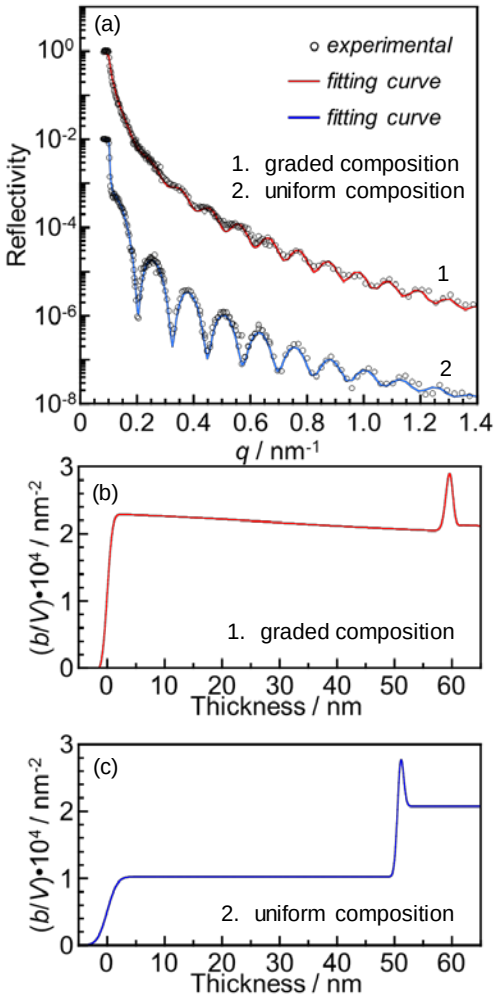

Figure 1. Neutron reflectivity for films. Open symbols depict experimental data, and solid lines represent the reflectivity calculated on the basis of the scat-tering length density (b/V) profiles.

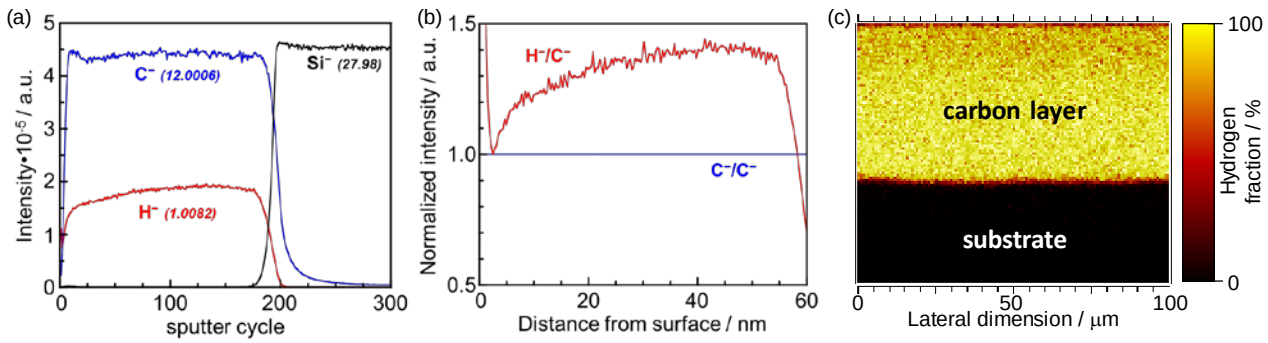

Figure 2. TOF-SIMS profile of the carbon film.