MLF Experimental Report	提出日 Date of Report 2010/7/23
課題番号 Project No. 2009B0011 実験課題名 Title of experiment Measurement of deuterium and hydrogen content o f DLC film by neutron reflectivity. 実験責任者名 Name of principal investigator Kazuhide Ozeki 所属 Affiliation Ibaraki University	装置責任者 Name of responsible person Hideki Seto 装置名 Name of Instrument/（BL No.） ARISA-II / BL 16 実施日 Date of Experiment 2010/5/28-2010/5/31

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.

Following DLC samples were measured by BL16. (Chemical formula: resource gas ratio). Two samples at a coating condition.

・Carbon hydrogen DLC film DLC (CxHy): (x=5, y=5: 50CH_4, 50H_2), (x=2, y=8: 20CH_4, 80H_2), (x=7, y=3: 70CH_4, 30H_2), (x=10, y=10: 100CH4)

・Carbon deuterated DLC film (CxDy): (x=2, y=8: 20CD_4, 80D_2), (x=5, y=5: 50CD_4, 50D_2), (x=7, y=3: 70CD_4, 30D_2), (x=10, y=10: 100CD_4)

・Carbon hydrogen deuterated DLC film (CxHyDz): (x=2, y=2, z=8: 20CH_4, 80D_2), (x=5, y=5, z=5: 50CH_4, 50D_2), (x=7, y=7, z=3: 70CH_4, 30D_2), (x=2, y=8, z=2: 20CD_4, 80D_2), (x=5, y=5, z=5: 50CD_4, 50H_2), (x=7, y=3, z=7: 70CD_4, 30H_2)

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.

1. Experimental method

1.1 Sample preparation

DLC deposition on the Si substrate (30mm x 30mm) was performed using a PED-301 RF CVD system. In this experiment, we focused on the contribution of H_2 gas source to the H content in the DLC film in CH_4+H_2 gas flow system, which helps us for understanding the complex coating mechanism of DLC film. We have chosen D_2 gas instead of H_2 gas or CD_4 gas instead of CH_4 gas for the DLC coating.

CH_4, H_2, CD_4 and D_2 gases were introduced into the chamber. The coating carried out at 13.3 Pa and a discharge power of 250W. Four types of flow gas combination were selected as CH_4+H_2, CD_4+H_2, CH_4+D_2 and CD_4+D_2. At each flow condition, the partial flow rate of H_2 or D_2 was varied from 20% to 80%. The film thickness of all samples was around 200nm.

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

1.2 Measurement

The neutron reflectivity was performed using a BL16 reflectometer, and neutron reflectivities were taken from 0.007 to 0.18 in qz ($Å^{-1}$). In addition to the reflectivity measurement, Rutherford backscattering spectroscopy (RBS) and elastic recoil detection analysis (ERDA) were performed for analysis of average H or D content of the DLC film.

2. Results

Figures 1 and 2 show an example of neutron reflectivity and the SLD of the DLC film ($50CD_4$, $50H_2$) from fitting simulation, respectively. From fig.2, the SLD at the film surface reached 7.0E-6 / $Å^2$. The layer thickness is around 50 Å, which can be considered to be an oxide layer. The SLD gradually decreased from 6.8 E-6 / $Å^2$ to 4.6 E-6 /A^2 at the range of 760 Å - 1390 Å. This indicates the decrease of the mass density or the increase of hydrogen. In addition, third layer (SLD = 3.0 7.0E-6 / $Å^2$) was also observed at the interface of the DLC/Si substrate. The layer thickness was around 20 Å, which can be an intermediate layer of Si, or SiO. The layer has not observed yet in the neutron reflectivity measurement. BL16 can provide the reflectivity at high Q range, and this high Q range measurement contributes to find this thin layer. The intermediate layer was observed in all measured samples.

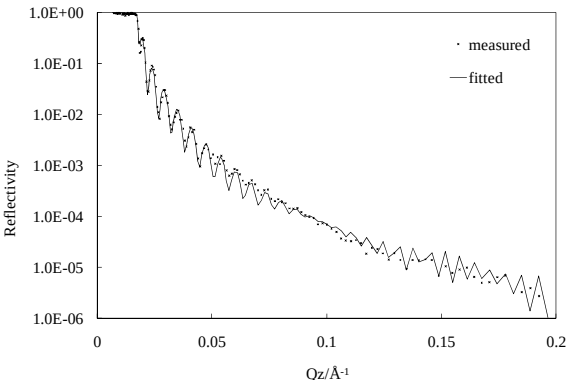

Fig.1 Neutron reflectivity for DLC ($50CD_4$,$50H_2$) film.

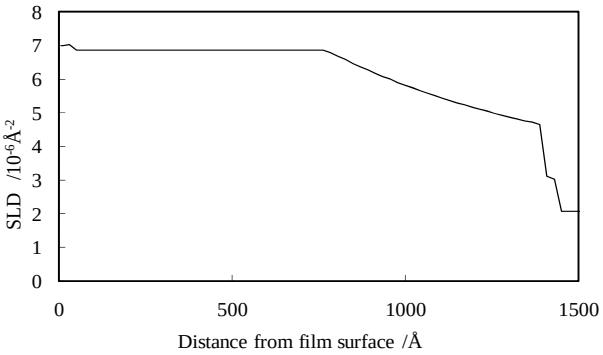

Fig.2 Scattering length density for the DLC ($50CD_4$,$50H_2$) film.

Table 1 shows SLD and D/H content of the DLC films coated under various conditions. The average SLD increased with increase of D/H ratio of the film. Even if the $H_2/(H_2+CH_4)$ ratio was changed from 0 to 0.8 in the sample No.8-10 and 14, the measured hydrogen content varied only from 31% to 38%, which may impose the small contribution of H from H_2 gas source. In the sample No.11-13, H and D contents widely changed from 6% to 26%. This might indicate that H from CH_4 gas and H from H_2 gas evenly contribute to H content in the DLC film.

Table 1 SLD and H/ D content of the DLC film.

No.	Samples	Average SLD (10^{-6} /A^2)	ERDA	
			H (%)	D (%)
1	$20CD_4,80H_2$	4.5	22	17
2	$50CD_4,50H_2$	6.4	11	35
3	$70CD_4,30H_2$	6.9	4	38
4	$20CD_4,80\ D_2$	6.1	4	32
5	$50\ CD_4,50\ D_2$	7.1	2	39
6	$70CD_4,30\ D_2$	7.0	11	25
7	$100CD_4$	7.4	2	37
8	$20CH_4,80H_2$	3.3	38	0
9	$50CH_4,50H_2$	3.5	34	0
10	$70CH_4,30H_2$	3.9	32	0
11	$20CH_4,80D_2$	5.5	11	21
12	$50CH_4,50D_2$	6.5	15	24
13	$70CH_4,30D_2$	4.2	26	6
14	$100CH_4$	4.1	31	0