MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2015A0071 実験課題名 Title of experiment Influence of neutron radiation on Ge-S (Se) films and dual film structures containing Ge-S (Se) /Ag films 実験責任者名 Name of principal investigator Yoshifumi Sakaguchi 所属 Affiliation CROSS	装置責任者 Name of responsible person Kenichi Oikawa 装置名 Name of Instrument/(BL No.) NOBORU (BL10) 実施日 Date of Experiment March 15, 2016

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.

Ge_xS_{100-x} film, Ge_xSe_{100-x} film, Ge_xS_{100-x}/ Ag film, Ge_xSe_{100-x}/ Ag film (x=20, 33, 40)

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 prepared 12 sample packages which are made of aluminum coated polythene sheet (110mm x 110mm) and in which 12 sets of different films are contained. Fig.1 shows the photograph of the inside of the sample package. One sample package was set on the beam path in order that all of the films in the package were exposed to the neutron beam with 100mm x 100mm beam size. Also, one other sample package was set on out of the neutron beam path as a control. The sample packages were exposed to the neutron beam with two different filter conditions (without filter and with bismuth filter) and for three different times (10 min, 1 hour, and more than 8 hours) as summarized in Table 1. One day beam time was used for the neutron beam irradiation. After cooling down of the irradiated samples, the samples were shipped to Boise State University (U.S.A.) and the effect of the neutron irradiation was investigated by Raman spectrometer.

Fig.1

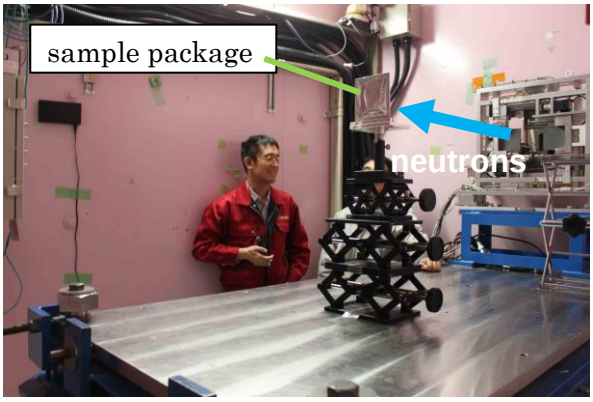

Fig.2

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

The Raman studies revealed that the studied samples have the specific features, characteristic for the particular Ge–S(Se) compositions. Example is the Raman curve for the $Ge_{30}Se_{70}$ composition. It represents the corner–sharing (CS), edge–sharing (ES) and Se chains (Se–Se) structural units building this composition after the films have been irradiated for 1 hour and 8 hours – Fig. 3. It is obvious that there are some, even though small changes in the films structure after different dose of irradiation. To make it better visible, we presented the area of the peaks as a function of the irradiation dose in Fig.4.

No filter	10min	1 hour	9.5 hours
Bi filter	10min	1 hour	10 hours

Table 1

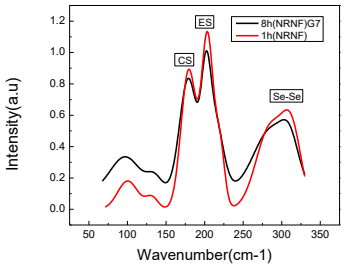

Fig. 3

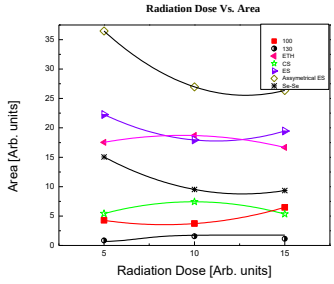

Fig. 4.

One can discern from these results that the structure condenses through formation of a higher number of ES structures but on the other hand these structures are connected with longer Se chains which decreases the density of the films. Comparing these results to our previous studies related to proton irradiation it comes out that the influence of neutron flux irradiating the samples through a gamma ray filter is lower compared to this of protons. This is especially true for the bi–layered structures containing a film of silver. The first indication for this is coming from the study of the Raman spectra of these bi–layered structures. In case of proton irradiation silver surface deposition occurs which completely wipes off the signal due to formation of a medium with a quite high conductivity. So, in the case of proton irradiation no Raman spectra of the films could be read. This was not the case at neutron irradiation. We suggest that the charge related to the protons is a reason for their stronger influence over the chalcogenide films.

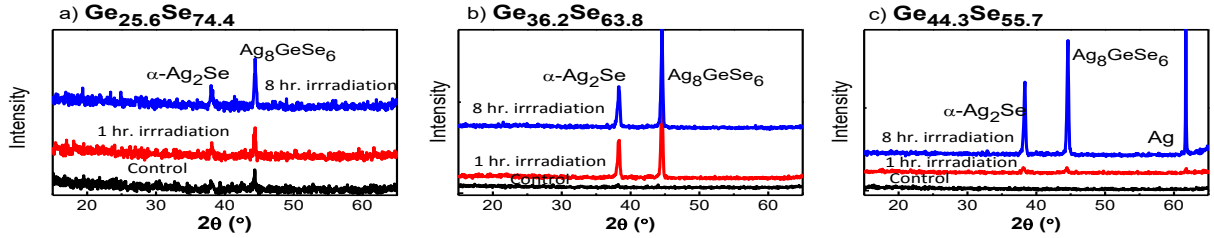

Fig. 5.

Fig.5 shows the result of X–ray diffraction. The composition forming after Ag diffusion into the chalcogenide films, caused by irradiation shows formation of the three component composition Ag_8GeSe_6 whose clusters grow with the increasing the irradiation intensity and Ge content in the samples. One interesting results is formation of αAg_2Se which is stable at temperature above 133°C. We suggest that the reason for this is the fact that this composition forms within the existing structure of the chalcogenide film, which requires the closest packed structure to form.