第68回応用物理学会春季学術講演会 講演予稿集 (2021 オンライン開催)

Synthesis of ternary type II clathrate films using AlGe alloy

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Type II clathrates (M_xIV_{136}, $0 \leq x \leq 24$) are one of the candidates of the high efficiency photovoltaic materials which are consisted by group IV elements (IV) with formations of cage like structures, and metallic atoms (M), typically alkali or alkaline earth elements. The metal atoms are included in the structures and act as electron donor. By removing the guest atoms, the properties of M_xIV_{136} varies from metallic to semiconducting. Recently, our group has succeeded to synthesize type II Si and Ge clathrates in film forms [1-3]. However, carrier type control had not been conducted which is necessary to apply the materials to semiconductor devices such as solar cells. In this research, we report preparations of $Na_x(Al_yGe_{1-y})_{136}$ films using the AlGe alloys with respect to work as acceptor in framework.

The AlGe films were prepared on sapphire substrates with Al/(Al+Ge) compositions of 2.4 (Al-02), 10 (Al-10) and 15 % (Al-15) by using rf co-sputtering. Subsequently, Na evaporation was conducted in a high vacuum and then the Na deposited AlGe film was immediately annealed for 6 h without exposure to air.

X-ray diffraction (XRD) patterns of the samples show formations of type II clathrate structures as shown in figure 1. According to the refinements of XRD patterns, the Na concentration of the samples were estimated about x = 2.8, x = 2.1, x = 2.1 for samples Al-02, Al-10, Al-15. The volumes of the unit cell of the $Na_x(Al_yGe_{1-y})_{136}$ films were decreased with increase of Al concentrations. It is suggested that the Al atoms are included in $Na_x(Al_yGe_{1-y})_{136}$ films with partial substitution of Ge atoms.

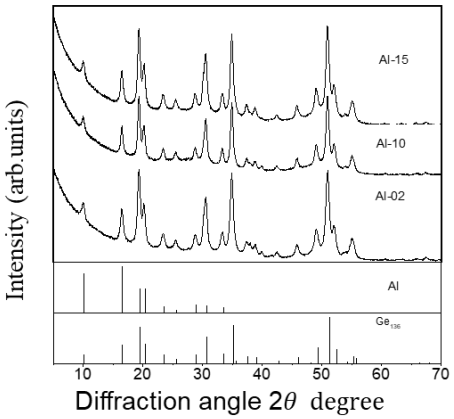

Figure 1. XRD patterns of type II clathrate films prepared with AlGe alloy on sapphire substrates.

Acknowledgment: The work reported here is supported by KAKENHI (No.16K21072, 17H03234, 20K0382).

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