29p-B6-4

Effect of solute transport on dissolution of Si into Ge melt and growth of SiGe

°M.Omprakash[1], M.Arivanandhan[1], R.Arun Kumar [1,2] H.Morii[1], T.Aoki[1], T.Koyama[1], Y.Momose[1], H.Ikeda[1], H.Tatsuoka[1], Y.Okano[3], T.Ozawa[4], Y.Inatomi[5], S.Moorthy Babu[6], Y.Hayakawa[1]
RIE Shizuoka Univ.[1],PSG college of Technology.[2] Osaka Univ.[3], Shizuoka Insti Sci. and Tech.[4], JAXA[5], Anna Univ.[6]
E-mail: omprakashmuthusamy@gmail.com

Introduction:

Thermoelectric energy conversion is one of the promising ways to convert the electric energy from waste heat [1]. Silicon-germanium ($Si_{1-x}Ge_x$) alloy semiconductor is a good material for thermo-electrical power generators at high temperature (from 700 to 1000ºC). It is highly challenging to optimize the figure of merit (ZT). Moreover, the thermoelectric properties of the $Si_{1-x}Ge_x$ alloy are strongly dependent on the composition of the material. Therefore, it is necessary to grow the $Si_{1-x}Ge_x$ crystals with homogeneous composition. After that, the optimum doping conditions is established by adjusting carrier concentration. In-situ observations of the growth process are important [2]. In the present study, the dissolution process of Si into Ge melt and SiGe growth was investigated by X-ray penetration method .

Experimental procedure:

Fig.1 indicates an experimental setup for X-ray penetration method. A rectangular shaped Si/Ge/Si sandwich sample was used. The dimension of Si (seed, feed) and Ge are 6 x 6 x 3 and 6 x 3 x 3 mm^3 (W x L x H), respectively. The crystals were polished with alumina abrasive and etched in the acid mixture of $HF:HNO_3$ (1:1) for Si and $HF:H_2O_2$ (1:1) for Ge. The sample was covered by BN tube inserted into the quartz ampoule, which was transparent to X-rays. The ampoule was evacuated about 10^{-4} Pa before sealing. The sealed ampoule was vertically fixed into the furnace. X-ray source was the tungsten target with acceleration voltage 150 kV and current 0.1 mA. X-rays were passed through the sample and the penetrated X-ray intensities were detected by the rectangular shaped CdTe detector. The penetrated X-ray intensities and images of the sample were measured as a function of time and temperature.

Results and discussion:

Fig. 2 shows X-ray intensity as a function of temperature. The sandwich sample loaded furnace was increased at a pre-determined heating rate to melt the Ge and the X-rays were allowed to penetrate through the sandwich sample from reference temperature (T_r) of 660 ºC. When T_r reached 930 ºC Ge was completely melted and the melting process was observed clearly from the intensity profile compared with RT intensity profile. Both changes of penetrated intensities and length of the Ge are detected due to density variation. The Si feed started to dissolve initially into Ge melt as observed by a shoulder region in the intensity profile at 976 ºC after 20 min and intensity of SiGe solution region increased as the temperature increased due to continuous dissolution of Si feed into Ge melt. Both seed and feed dissolution proceeded at 1140 ºC as observed the interface shift in the intensity profile. The intensity profile of SiGe solution continuously changed due to solute transport from seed interface towards to feed interface due to solutal convection induced by gravity and accumulates Si richer near the feed interface and suppress the further feed dissolution. Once the temperature reached 1200 ºC it was kept constant for about 4h. It was found that growth was started from feed interface after 343 min. The crystal must be Si richer and the solution near the interface should be Ge richer. Therefore near growth interface, a sudden change of intensity was observed at 1200 ºC after 343 min due to density variation between SiGe solid and liquid. After growth process, the grown sample was removed from the ampoule and polished to study the compositional distribution by EPMA. It was found that crystal was grown from feed interface to seed interface. Si composition of 0.7 was grown upto 1mm and it gradually decreased from 0.7 to 0.6 due to segregation phenomena shown in fig. 3.

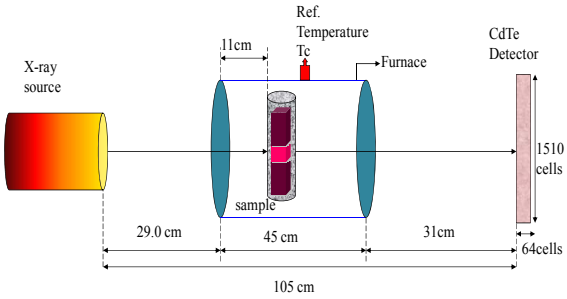

Fig. 1 Experimental set up for X-ray penetration method

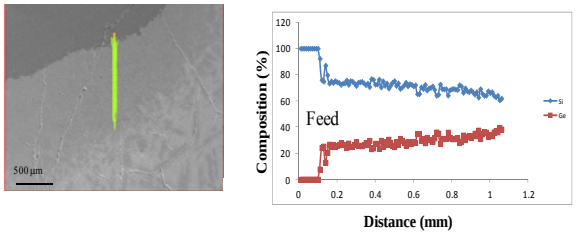

Fig.3 SiGe composition from feed interface upto 1mm

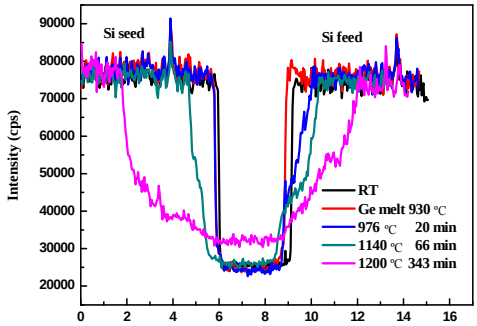

Fig. 2 X-ray intensity as a function of temperature

Reference

[1] M.Arivanandhan, Y.Hayakawa, et al., J.Cryst.Growth **519** (2011) 8532.
[2] G.Rajesh, M.Arivanandhan, Y.Hayakawa, et al., J.Cryst.Growth **312** (2010) 2677.