

## Effect of Pr filling on the unfilled skutterudite $\text{CoSb}_3$

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Effect of rare earth elements (*RE*) filling on unfilled skutterudite  $\text{CoSb}_3$  is a interesting subject from two view points; i.e., application as thermoelectric materials and basic scientific interest. There have been reported quite a few trials to attack this subject. To increase the filling factor, some trials on partial replacement of Co site by Fe,  $\text{Ce}_x(\text{Fe}_{1-y}\text{Co}_y)_4\text{Sb}_{12}$ , have been reported, however 100 % filling was unsuccessful [1]. Since  $\text{CoSb}_3$  is stable, it is difficult to increase the filling factor of *RE* in the antimony cage at ambient pressures. In contrast with the  $\text{REFe}_4\text{P}_{12}$  and  $\text{RERu}_4\text{Sb}_{12}$ , no successful reports on 100% filling has been made even in  $\text{REFe}_4\text{Sb}_{12}$  at ambient pressure [2]. Recently, we have succeeded in synthesizing high quality sample of  $\text{PrFe}_4\text{Sb}_{12}$  by using high pressure synthesis, and found the physical properties are quite sensitive to the Pr site filling [3]. The magnetic ground state changes from a ferrimagnet to a highly enhanced paramagnet, which is consistent with the large density of states near the Fermi energy associated with Fe-3*d* electrons in the band calculation on  $\text{LaFe}_4\text{Sb}_{12}$  [4]. This fact suggests an important role of 3*d* electrons in controlling the physical properties of these systems. To study the effect of 3*d* electrons on physical properties, we are trying to synthesize  $\text{PrCo}_4\text{Sb}_{12}$  under high pressures, in which Fe is substituted for Co having one more *d* electron.

After some trials under different conditions, we have obtained the filled skutterudite compound  $\text{Pr}_x\text{Co}_4\text{Sb}_{12}$ . Figure 1 shows powder x-ray diffraction pattern of this sample. We confirmed that the filled skutterudite was synthesized and a little unknown impurities included in the samples. The lattice constant was estimated to be 9.089 Å larger than that of unfilled skutterudite  $\text{CoSb}_3$ . In preliminary chemical composition analysis on field emission electron microscope, Pr site filling factor is estimated to be about 50 %. We will report the detail of synthesis techniques and the physical properties of  $\text{Pr}_x\text{Co}_4\text{Sb}_{12}$  in this study.

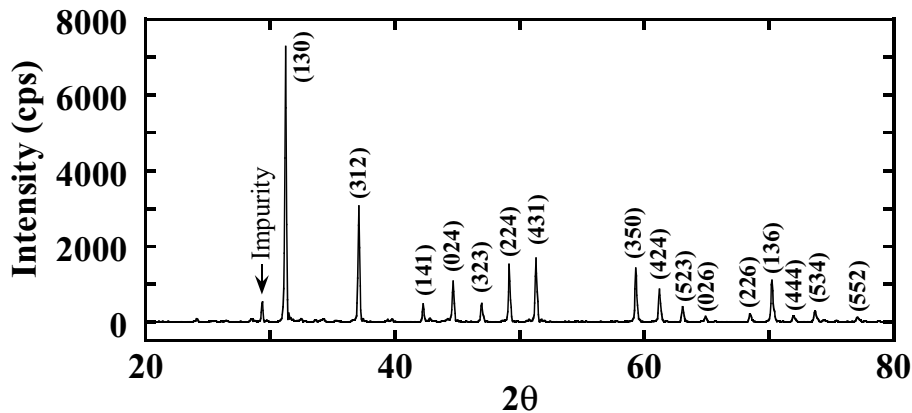


Figure 1: Powder x-ray diffraction pattern of  $\text{Pr}_x\text{Co}_4\text{Sb}_{12}$

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- [2] E. Bauer *et al.*, Phys. Rev. B **66** (2002) 214421.
- [3] K. Tanaka *et al.*, to be published in Physica B (2006).
- [4] K. Takegahara *et al.*, J. Phys. Soc. Jpn. **71** (2002) Suppl. pp. 240.