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学 位 論 文 題 目	

Development of Higher Borides and Heusler Alloys as High and Low Temperature Thermoelectric Materials

(高温及び低温熱電材料としての多ホウ化物及び Heusler 合金の開発)

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論　文　の　要　旨

The main focus of this work is the investigation and development of new materials forthe high and room temperature ranges. REB_{66} compounds and boron carbide were selected to investigate as well as the Full Heusler alloy Fe_2VAl for the high and low temperature regimes, respectively. Magnetic effects in thermoelectrics are also of interest and investigated. SmB_{66} was previously indicated to have enhanced properties possibly because of mixed valency effect of Sm. Single crystalline YbB_{66} as well as polycrystalline REB_{66} (RE=Y, Sm, Ho, Tm, Yb) fabricated for the first time by a synthesis route involving arc melting and spark plasma sintering were investigated in the applicant’s work. Strikingly, the thermoelectric properties of these compounds were discovered to be almost independent of the microstructure. The nature ofthe rare earth also found not to strongly affect the thermoelectric properties, differencesoriginating mainly from different compositions or the presence of secondary phases.Relatively large negative Curie-Weiss temperatures θ were observed with unusual rare earth dependence indicative of an unusualcoupling mechanism. The thermoelectric properties of phase-pure carbon-rich boron carbide $B_{4.05}C$ synthesized from a sucrose precursor by a solution-based method were reported and discussed using commercially available boron carbide powders as references. Electrical conduction mechanisms of boron carbide are also discussed, and the nearest-neighbor hopping conduction model was concluded to bemore valid than the often-used bipolaron model. The work on the Full Heusler alloy Fe_2VAl dealt with the connection of thermoelectric and magnetic properties via spin fluctuation. V was successfully substituted with Co resulting in ferromagnetic materials with Curie-temperatures shifted towards higher temperatures and maximum in the Seebeck enhancement could be observed at or above room temperature.

審　　査　　の　　要　　旨

〔批評〕

The applicant synthesized and investigated the properties of high temperature and room temperature thermoelectric materials which are maybe the most viable regions for future applications. In particular the thermoelectric and magnetic properties of REB_{66} were analyzed carefully. A previous study indicated that SmB_{66} has enhanced properties indicatedfrom mixed valency, however, the applicant obtained similar good properties in other REB_{66} which were indicated from magnetic properties to be trivalent, and the good properties origin were able to be attributed to metal rich compositions, with not much rare earth dependence apparent. Polycrystalline REB_{66} (RE=Y, Sm, Ho, Tm, Yb) were fabricated for the first time by a synthesis route involving arc melting and spark plasma sintering, which is much easier than the typical single crystal growth method, and strikingly, with thermoelectric properties similar to single crystals, indicating an independence of the microstructure.A different conduction mechanism from the usually assumed bipolaron conduction mechanism, was proposed for the prototypical boride thermoelectric material, boron carbide. Magnetic and thermoelectric properties of Co doped Fe_2VAl were found to be able to be controlled, with the transition temperature shifted to higher temperatures and the enhancement from spin fluctuation reaching a maximum at room temperature.The applicant has achieved interesting results into synthesis and investigation of thermoelectrical and magnetic properties. These achievements should accelerate the research,especially on higher boride thermoelectrics going forward. It is judged that the work by the applicant merits awarding of PhD of Engineering.

〔最終試験結果〕

The final examination committee conducted a meeting as a final examination on February17, 2020. The applicant provided an overview of dissertation, addressed questions and comments raised during Q&A session. All of the committee members reached a final decision that the applicant has passed the final examination.

〔結論〕

Based on the results of the above-mentioned dissertation defense and final examination, the final examination committee approved that the applicant is qualified to be awarded Doctor of Philosophy in Engineering.