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学位の種類	博士（工学）
学位記番号	博甲第 9638 号
学位授与年月日	令和 2 年 5 月 31 日
学位授与の要件	学位規則第4条第1項該当
審査研究科	数理物質科学研究科
学位論文題目	Development of New Synthesis Routes for High Temperature Thermoelectric Materials via Spark Plasma Sintering Utilizing Solid-State Reaction, Liquid Phase Sintering, and Gas/Solid Reaction (固相反応、液相焼結、気固反応を活用した放電プラズマ焼結による高温熱電材料の新規合成法の開発)
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論　文　の　要　旨

In this thesis, is reported on the development of new methods for synthesis of high temperature ceramic materials and development of their thermoelectric properties. The thesis reports on the first attempt of an n-type thermoelectric $MgTi_2O_5$/TiN conductive composite, which overcame the traditional trade-off relationship between σ and κ, fully densified p-n controllable polycrystalline $Y_xAl_yB_{14}$, and directly synthesized $YB_{22}C_2N$ without additives for high temperature thermoelectric applications. Rapid and effective synthesis routes for these ceramic materials via reactive SPS were also newly developed. The new processes for preparation of $MgTi_2O_5$/TiN composites, $Y_xAl_yB_{14}$ and $YB_{22}C_2N$ utilize solid-state reaction, liquid phase-assisted sintering, and gas/solid reaction technologies, respectively. These breakthrough techniques are expected to be applicable to the synthesis of other oxide-based composites, higher borides and rare earth borocarbonitrides. Furthermore, it is also expected that the newly developed direct synthesis technique by in-situ nitridation via reactive SPS utilizing N_2 gas/solid reaction could open the door for more accessible synthesis of not only borocarbonitrides but also other nitrides.

審　査　の　要　旨

〔批評〕

There is a need to develop n-type thermoelectric oxides, and the applicant focused on $MgTi_2O_5$ which is an insulator with negligible thermoelectric properties, but showed through composite formation with TiN that a huge improvement of properties could be attained. Interesting circumventing of conventional tradeoff through utilizing cation disorder and other effects were also demonstrated. Thermoelectric borides have been

predominantly p-type, and the applicant focused on two promising systems, $YAlB_{14}$ which has p, n control and $YB_{22}C_2N$, the n-type counterpart to boron carbide, which both have always had problems of densification and cumbersome synthesis. Fully densified p-n controllable $YAlB_{14}$ and direct synthesis of $YB_{22}C_2N$ was achieved for the first time. Rapid and effective synthesis routes for all materials via reactive SPS were also newly developed. The applicant has achieved interesting results into new synthesis approaches of oxide and boride ceramics and development of their thermoelectrical properties. These achievements should accelerate the research on ceramic material 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 April 8, 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.