High Temperature Thermoelectric Properties of W18O49-based Materials

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（$W_{18}O_{49}$ - 系材料の高温熱電特性）

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論　文　内　容　の　要　旨

Thesis Summary

This thesis is devoted to the investigations for high-temperature thermoelectric properties of $W_{18}O_{49}$-based materials. The study began with the synthesis strategies for acquiring monophasic ceramic $W_{18}O_{49}$, following by attempts on improving thermoelectric properties via oxygen vacancies manipulating. Next, the anisotropic nature of the structures and microstructure morphologies were investigated for the effects on thermoelectric performance. Finally, attempts on carrier concentration control by substitution W^{6+} with lower valance cations were reported with significant improvement in thermoelectric efficiencies.

This thesis consists of six chapters. Summaries of each chapter are presented in the following:

Chapter 1 presents the introduction, the background and motivation for this work. The current status of high temperature thermoelectric materials was briefly reviewed with the emphasis of the newly focused Tungsten suboxides materials.

The chapter 2 explains the main experimental processes involved in this study and all the corresponding apparatus were employed. Furthermore, the theories behind the characterization methodologies of the thermoelectric properties were discussed.

The chapter 3 elaborates the work in direct synthesis of $W_{18}O_{49}$. By reactive SPS, anisotropic thermoelectric properties of $W_{18}O_{49}$ could be partially influenced by the increment of applied pressure. The ordered orientation of crystallite under high pressure offers an intrinsically low thermal conductivity materials, despite its metallic nature, via more efficient intrinsic phonon scattering from the alignment of the tunnel structures. Theoretically, anisotropy could even be significantly strengthened by applying higher uniaxial pressure, thus, *ZT* was enhanced by employing this technique to 0.08 at 1073 K under sintered pressure of 50 MPa in comparison with the *ZT* of 0.06 of the less oriented samples.

The chapter 4 elucidates the processes in enhancing the thermoelectric performance of the $W_{18}O_{49}$-based materials by carrier tunning. Since the Tungsten oxides WO_x $(2 \leq x \leq 3)$ are a series of homologous structures, isolating monophasic polycrystalline ceramic materials was difficult. Many attempts were made from solid-state reaction in vacuum, to hot-pressing sintering and reactive spark plasma sintering. The $W_{18}O_{49}$ ceramic could be directly synthesize as a single phase through reactive spark plasma sintering, or pre-reacted in vacuum following by densification via SPS. The former method leaves the sample with anisotropic structures and thermoelectric properties, while the later is suitable for doping strategy through thermal diffusion mechanism.

Chapter 5 presents the work on the doping of Ti in $W_{18}O_{49}$-based materials for improving its thermoelectric performance. The $(W_{1-x}Ti_x)_{18}O_{49}$ samples $(0 \leq x \leq 0.25)$ were prepared by solid state reaction in vacuum followed by densification via spark plasma sintering method. The Ti substitution increased the Seebeck coefficient, the power factor, and decreased both the electronic and lattice thermal conductivity. The synergistic substitution effect on the electrical and thermal properties and inherently low lattice thermal conductivity of < 1 W K^{-1} m^{-1} originating from the tunnel structure led to the *ZT* of 0.2 at 1073 K ($x = 0.1$). The Jonker-type approximation between the Seebeck coefficient and electrical conductivity indicated that the power factor (and the *ZT*) of $W_{18}O_{49}$ can be further enhanced by decreasing the electron carrier concentration. For sample $x \geq 0.1$, secondary unknown phase was observed, indicating the solubility limit of Ti as a dopant. However, the thermoelectric performance of the material was continuously enhanced up to $x = 0.2$ reaching highest ZT value of 0.5 at 1073K, then decreasing drastically due to the domination of secondary phases.

Chapter 6 summarizes the findings and conclusions from the preceding chapters, with the discussion about the outlook for future developments of $W_{18}O_{49}$-based materials for thermoelectric applications.