MS13-2-11 Magnesioreduction synthesis of silicides: the structure-properties relationship #MS13-2-11

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Abstract

Thermoelectric (TE) generators are all-solid-state devices that enable the conversion of heat, including heat losses, into electricity thanks to the Seebeck effect. These generators most commonly consist of an assembly of n- and p-type TE materials connected electrically in series and thermally in parallel between two ceramic plates. Important research efforts have been undertaken during the past two decades to increase TE efficiency of numerous materials such as Bi_2Te_3 , $GeTe_2$ or PdTe₃ to enable a more extensive development of TE technology. However, many other factors must be taken into account when selecting materials for TE generator, such as the raw material costs and toxicity, mechanical properties, chemical and thermal stability, and thermal expansion coefficients. For all these reasons, Silicides (HMS and β -FeSi2) are considered as promising industrial thermoelectric materials for the mid-temperature applications (600 - 800 K), despite a moderate efficiency, however one of the main obstacle of a more diffuse application of such materials remain their synthesis. For such reason we develop the magnesiothermic reduction (MR) synthesis for such compaunds. The MR rapidity and lower operating temperature synthesis is coupled with a reduction of average powder grain size, with a high cristallinity.

Such cristallinity has allowed us fine characterize by X-ray and electron diffraction the materials obtained and study the relationship between their complex crystallographic structure (incommensurability for MnSi1.74 and presence of stacking fault for β -FeSi₂), and microstructure with the synthesis conditions and how such parameter influence the thermoelectric properties of such materials. In particular to (i) to precisely determine the γ ratio of the composite structure of V_xMn_{1-x}Si γ , which controls the VEC and therefore the transport properties [3,4], or (ii) to quantify the probability of stacking faults in β -Co_yFe_{1-y}Si₂ which can influence the lattice thermal conductivity[5].

References

(1) Le Tonquesse, S.; Alleno, É.; Demange, V.; Prestipino, C.; Rouleau, O.; Pasturel, M. Reaction Mechanism and Thermoelectric Properties of In0·22Co4Sb12 Prepared by Magnesiothermy. Materials Today Chemistry 2020, 16, 100223. https://doi.org/10.1016/j.mtchem.2019.100223.

(2) Le Tonquesse, S.; Alleno, E.; Demange, V.; Dorcet, V.; Joanny, L.; Prestipino, C.; Rouleau, O.; Pasturel, M. Innovative Synthesis of Mesostructured CoSb3-Based Skutterudites by Magnesioreduction. Journal of Alloys and Compounds 2019, 796, 176–184. https://doi.org/10.1016/j.jallcom.2019.04.324.

(3) Le Tonquesse, S.; Verastegui, Z.; Huynh, H.; Dorcet, V.; Guo, Q.; Demange, V.; Prestipino, C.; Berthebaud, D.; Mori, T.; Pasturel, M. Magnesioreduction Synthesis of Co-Doped β-FeSi2: Mechanism, Microstructure, and Improved Thermoelectric Properties. ACS Appl. Energy Mater. 2019, 2 (12), 8525–8534. https://doi.org/10.1021/acsaem.9b01426.

(4) Le Tonquesse, S.; Joanny, L.; Guo, Q.; Elkaim, E.; Demange, V.; Berthebaud, D.; Mori, T.; Pasturel, M.; Prestipino, C. Influence of Stoichiometry and Aging at Operating Temperature on Thermoelectric Higher Manganese Silicides. Chemistry of Materials 2020, 32 (24), 10601–10609. https://doi.org/10.1021/acs.chemmater.0c03714.

(5) Le Tonquesse, S.; Dorcet, V.; Joanny, L.; Demange, V.; Prestipino, C.; Guo, Q.; Berthebaud, D.; Mori, T.; Pasturel, M. Mesostructure - Thermoelectric Properties Relationships in V Mn1–Si1.74 (x = 0, 0.04) Higher Manganese Silicides Prepared by Magnesiothermy. J. Alloys Compd. 2020, 816, 152577. https://doi.org/10.1016/j.jallcom.2019.152577.

Rietveld refinement of washed MnSi1.74

