Optimization of Ni doping to improve thermoelectric properties of β-$Fe_{1-x}Ni_xSi_2$

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Iron silicide compound is an abundant and non-toxic material which has 3 different kinds of phases namely cubic ε-phase, tetragonal α-phase, and orthorhombic β-phase with space group of $P2_13$, *P4/mmm*, and *Cmce*, respectively. β-$FeSi_2$ is a semiconductor having a narrow band gap of about 0.73 eV which is relevant in high temperature thermoelectric (TE) application due to strong oxidation resistance and good thermal stability; however, the bipolar effect, which deteriorates the Seebeck coefficient (*S*) as temperature increases, usually occurs in the pristine β-$FeSi_2$ due to such a small band gap and low carrier concentration (n_H) [1-2]. Komabyashi *et al*., reported that the TE's parameters of thin film's $Fe_{0.94}Ni_{0.06}Si_{2.05}$ measured at room temperature such as: *S*, electrical resistivity (ρ), and power factor ($PF = S^2\rho^{-1}$) was -113 μV/K, 0.076 Ω.cm, and 17 $\mu W.m^{-1}. K^{-2}$, respectively [3]. In addition, Tani and Kido reported that the ρ of bulk's β-$Fe_{1-x}Ni_xSi_2$ increases with the addition of Ni amount owing to the increase in n_H [4], which is also significant to the reduction of bipolar effect. However, the optimum doping level of Ni to enhance the *PF* and the effect of Ni to thermal conductivity (κ) as well as TE's performance ($ZT = S^2\rho^{-1}\kappa^{-1}T$) of β-$FeSi_2$ has not yet been investigated. In the current work, we have studied in detail about the effect and the optimization of Ni dopant on the TE's properties of bulk's β-$Fe_{1-x}Ni_xSi_2$ prepared by arc-melting techniques. The powder XRD data were measured by CuKα diffractometer (SmartLab, Rigaku). The *S* and ρ were measured by ResiTest8300 and homemade apparatus, and the κ was measured by power conversion efficiency measurement system (PEM-2, ULVAC-RIKO). As a result, the addition of Ni significantly reduces the bipolar due to the increase in n_H and the *S* of bulk's Ni-doped samples are remarkably more stable than that of non-doped sample above 400 K. We observed that both $|S|$ and ρ remarkably decreases with increasing x, while κ is not significantly varied with x. The highest PF = 130 $\mu W.m^{-1}. K^{-2}$ is obtained in β-$Fe_{0.99}Ni_{0.01}Si_2$ at 760K, which is 7 times larger than the thin film sample reported by Komabyashi *et al*., at room temperature measurement. Therefore, the highest ZT = 0.01 is also obtained in β-$Fe_{0.99}Ni_{0.01}Si_2$ at 760 K with $n_H = 2.3(9) \times 10^{17}$ cm^{-3} due to the stability in $|S|$, the significant reduction in ρ, and no remarkable effect in κ.

References

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