Optimizing the Condition for High Thermoelectric Performance of Flexible $Ag_2(S,Se)$ Material through Synergic Approach

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The development of thermoelectric generators (TEGs) for alternate source of energy is currently one of major topics in renewable power generations as the TEGs are capable of supplying an input power (μW to mW) to many sensors, electronic gadgets, and wearable devices in low temperature range around 300 – 400 K [1]. Improvement of *dimensionless figure of merit*, *ZT*, of the thermoelectric materials used in such device are very important. Besides, if these materials are flexible [2], the number of potential applications is significantly enlarged.

Recently, Ag_2S is found to be an interesting material, as near room temperature it possesses very low thermal conductivity (~ 0.5 W/m K) and large Seebeck coefficients (~ –900 μV/K) together with good flexibility [3]. However, the value of *ZT* was found to be very low due to large electrical resistivity (~ 10^6 mΩ cm). To improve the *ZT*, the partial substitutions of Se, Te at sulfur site are investigated [4,5]. Interestingly, the ductile nature of Ag_2S material found to be excellent with Se substitutions at sulfur site for $Ag_2S_{1-x}Se_x$ ($0 < x < 0.55$) compositions, and the maximum *ZT* of ~0.6 at 300 K is achieved near room temperature [4]. The value of *ZT* is significantly improved, however, to make an efficient TE device which can sufficiently produce the required electric power using human body heat, the *ZT* of the material about unity or larger near room temperature is highly desirable.

ZT of Ag_2S based material get significantly improved via tuning the charge carrier concentration to increases electrical conductivity (σ). However it simultaneously leads to decreases in *S*, and an overall enhancement in power factor ($PF = S^2\sigma$) is limited. The modification of electronic structure near chemical potential is an effective way to significantly improve *S* of a semiconductor with capable of keeping large *S* with a small change in κ.

In this study, therefore, we modify the electronic structure of $Ag_2S_{1-x}Se_x$ by non-toxic 3*d* transition metal elements substitution on Ag site. By this approach we successfully improve the S and achieved an effective increase of *ZT* near 300 K.

Polycrystalline samples were prepared by using standard melting method. The measurements of all the samples were made for several cycles to check the reproducibility of TE properties.

Structural characterization of as-prepared powder sample was carried out by using Bruker D8 Advance with Cu K_α source, and found to be in single phase. Chemical composition and grain structure analyses were performed using Scanning Electron Microscope-Energy Dispersive X-ray spectroscopy (SEM-EDX), HITACHI SU 6600. Heat and electron transport properties were measured on hot-pressed bulk samples. Thermal conductivity measurement was done by using Laser flash analysis (NETZSCH LFA 457); whereas for Seebeck coefficient and electrical resistivity measurement, experimental systems developed in our laboratory were employed [4]. Longitudinal and transverse velocity measurements at room temperature were carried by using the commercial Olympus 5072PR pulser/receiver and ultrasonic transducers connected with Texio made digital storage oscilloscope system (DCS-1102B).

Transition metal elements (Cu to V) were partially substituted for Ag-site with several different concentrations below 1 at.%. We confirmed that *n*-type behavior was kept unchanged regardless of the substitutions. Notably, we found an appropriate condition where *S* was increased while the small magnitudes of ρ and κ were maintained. As a result, large magnitudes both in power factor and *ZT*, PF ~1390 $mWm^{-1}K^{-2}$ and ZT_{max} = ~ 0.8 at 310 K, were obtained. These numbers let us believe that Ag_2S -based thermoelectric materials would be usable in energy harvesting devices.

In this JSAP meeting, we will discuss in more details about the optimizing condition for high thermoelectric performance of flexible $Ag_2(S,Se)$ material through synergic approach.

References

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