Enhancing transport and thermoelectric properties of Heusler based alloys

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Introduction

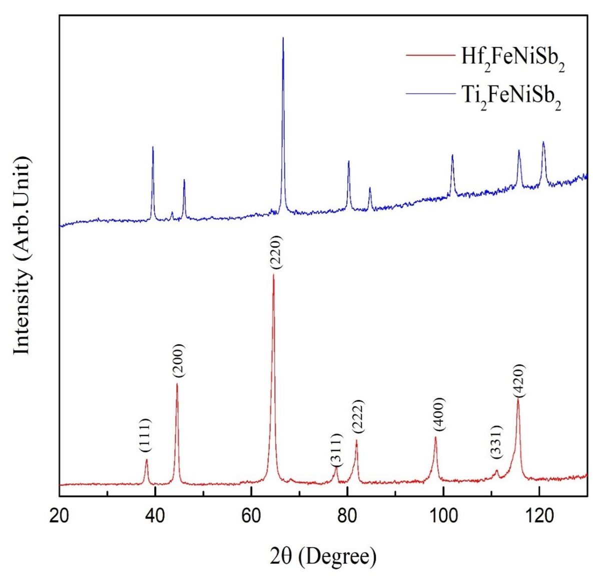
- **Thermoelectric materials** allow one to convert thermal energy into electricity and vice versa, and play therefore a vital role in solving the crisis of energy and environmental pollution.
- The **thermoelectric conversion efficiency** is determined by the dimensionless thermoelectric figure of merit $ZT = S^2\sigma T/K_{tot}$.
- **Half Heusler alloys** have been considered as a suitable option for waste heat recovery applications owing to **their high-temperature stability and a large ZT ~ 1** for both, *n*- and *p*-type compounds.
- Alongside with the half Heusler alloys, growing attention has been paid to so-called **double half Heusler (DHH)** alloys who exhibit significantly **lower thermal conductivity** as compared to the traditional half Heusler alloys.
- Several methods for tuning the lattice thermal conductivity by increasing phonon scattering via mass fluctuation or grain boundary scattering have been proposed and investigated
- **The melt spinning** method was found to be an efficient way to decrease the grain size, as the lattice thermal conductivity is sensitive to the grain size. As the average grain size reduced from 10 μm to 0.05 μm, the lattice thermal conductivity decreased from 10 to 4 $Wm^{-1}K^{-1}$. [1]
- In the current research, we adopted **the arc melting and melt-spinning methods** followed by Spark Plasma Sintering (SPS) to obtain double half Heuslers alloys $Ti_2FeNiSb_2$ and $Hf_2FeNiSb_2$.

Experimental work

- For preparing the samples, the precursors in a pure elemental form were weighed according to the nominal formula of both compounds $Ti_2FeNiSb_2$ and $Hf_2FeNiSb_2$
- The ingots were melted by arc melting under protective inert atmosphere
- Flakes were produced by putting the produced ingots inside quartz ampules with ~ 0.3 mm nozzle to be meltdown via **melt spinning** using Ar pressure to push them on the flying wheel at varying speed ~ 20-40 m/s.
- Using **Spark Plasma Sintering (SPS)**, tablets were formed by placing the powder in a graphite mold, then compacted for 15 minutes at a pressure of ~ 500 Kg/cm² and temperature of 1073 K.
- **X-ray Diffraction (XRD)** data were collected for powdered samples at room temperature.
- **Scanning electron microscopy (SEM)** was used to obtain the morphology.
- **Thermal diffusivity** was measured by a flash diffusivity method from 300 to 873 K under an Argon flow.
- **The electrical conductivity** σ, and **the Seebeck coefficient** *S*, were measured in a temperature range from 300 to 900 K via the standard four-probe and differential methods

XRD analysis

- All of the diffraction peaks are **well indexed** to the dominant compounds of $\mathbf{Hf_2FeNiSb_2}$, and $\mathbf{Ti_2FeNiSb_2}$ exhibiting a cubic crystal structure within the space group (F-43m) with a lattice parameter of 6.055 Å.
- The good indexicality and sharpness of the peaks indicate **the homogeneity of the obtained samples** and their consistency of fine grains, which affect the samples' thermal conductivity.



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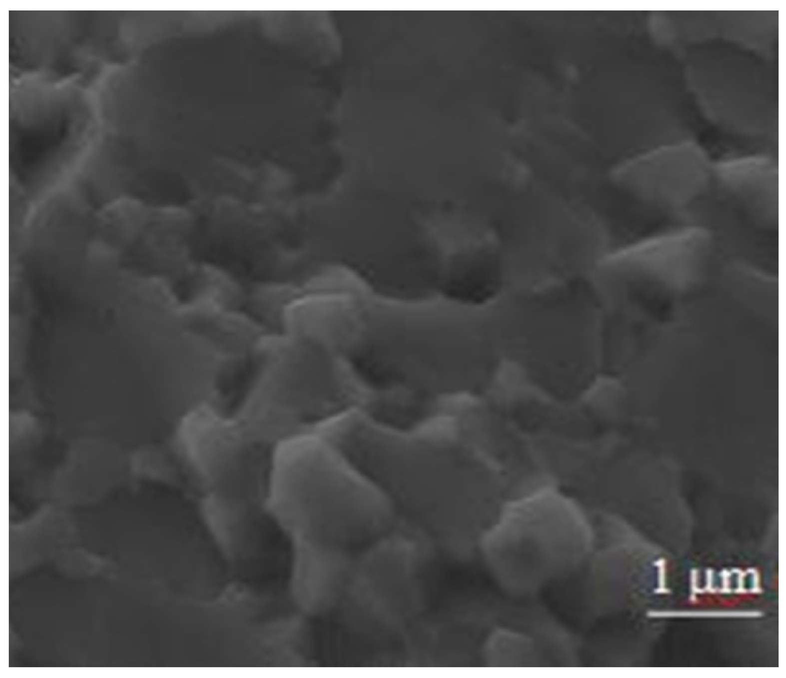
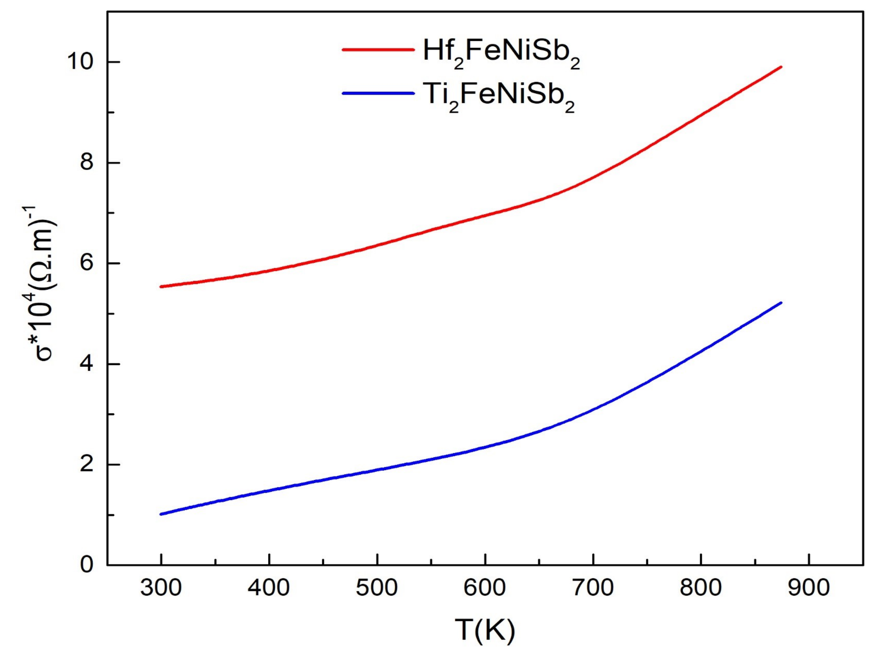
SEM Analysis

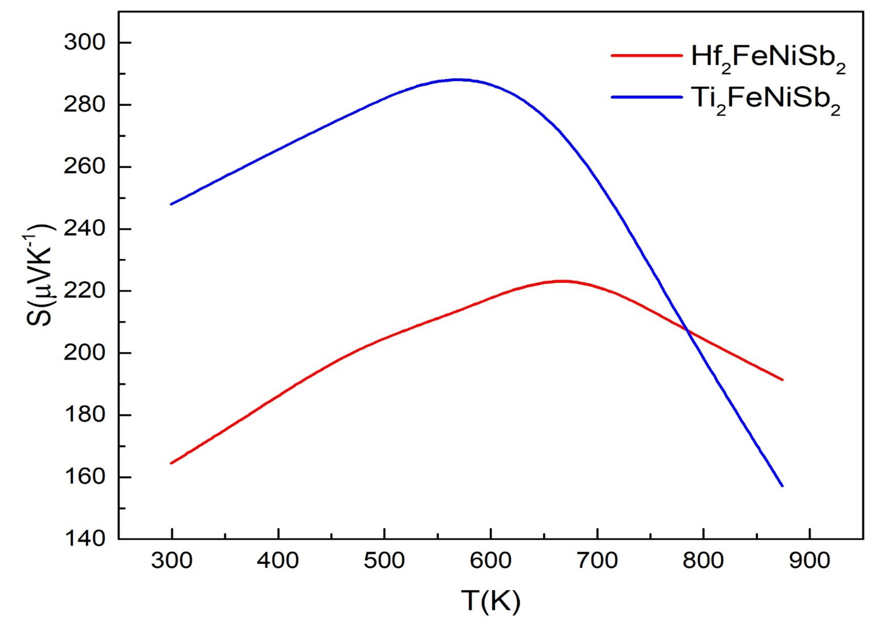
- Microstructure and surface morphology after treating the $Hf_2FeNiSb_2$ sample by SPS and annealing processes was investigated.
- **Fine grains of high-density** can be observed. Also, It is clearly seen that the sample are polycrystalline of high crystallinity, homogeneity and density.

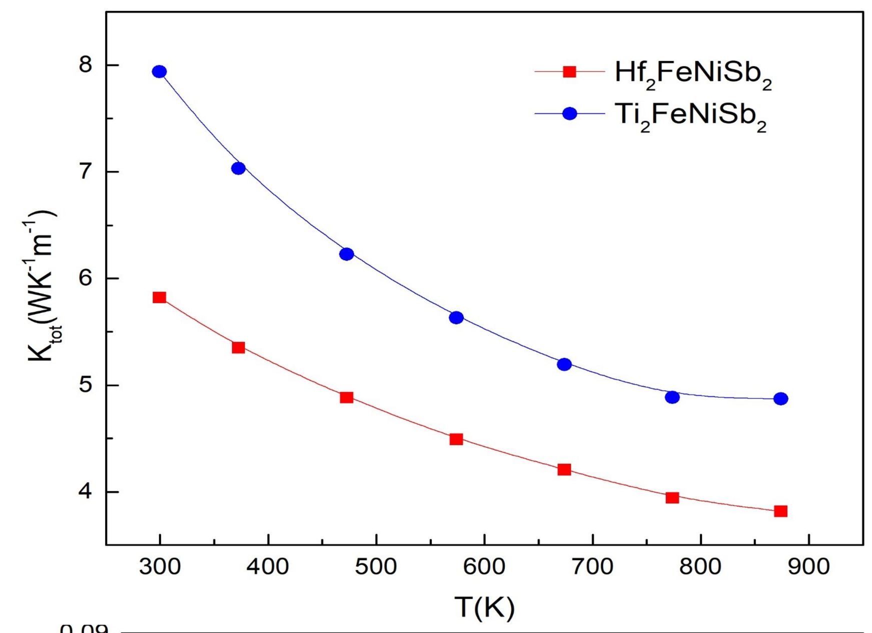


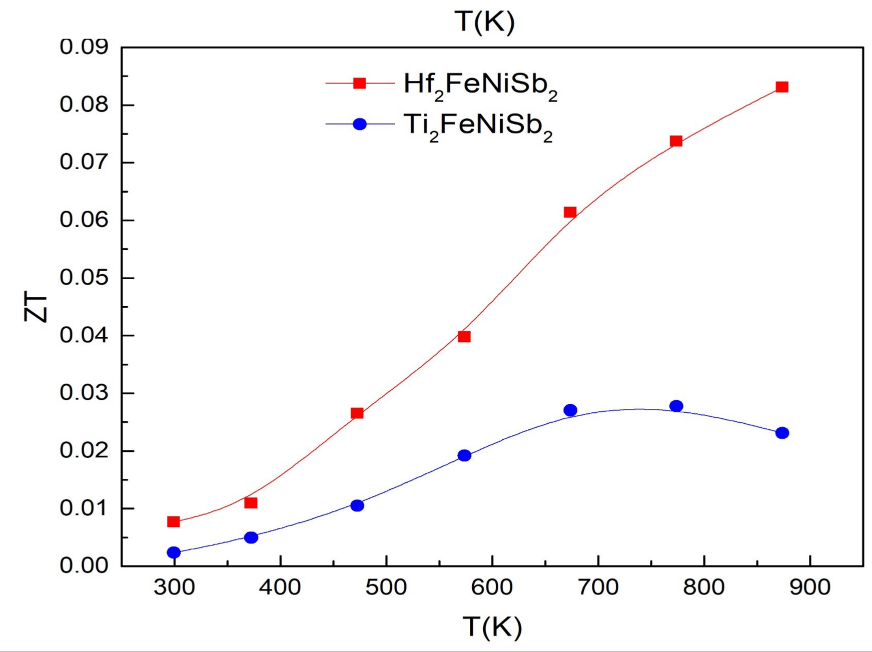
Transport and thermoelectric properties

- **The electrical conductivity** σ gradually increases with increasing temperature, till 650 K, for both samples due to their semiconducting nature.
- **The electrical conductivity** increases more rapidly and reaches maximum values of 9097 and 5240 $\Omega^{-1}m^{-1}$ for $Hf_2FeNiSb_2$ and $Ti_2FeNiSb_2$, respectively, at 873 K, which falls in line with the experimental results for $Ti_2FeNiSb_2$ which was reported previously. [2]
- **The Seebeck coefficient S** increases with increasing temperature, reaches a maximum, and then steeply decreases.
- This particular behaviour arises from the majority carrier (holes) which are responsible for the increase of the Seebeck coefficient up to specific temperature.
- **The Seebeck coefficient** maximum values were obtained for $Hf_2FeNiSb_2$ and $Ti_2FeNiSb_2$ as 228 and 293 μV/K at 675 and 575 K, respectively.
- The room temperature **thermal conductivity** $\mathbf{K_{tot}}$ is as high as 5.82 and 7.95 W m^{-1} K^{-1} for $Hf_2FeNiSb_2$ and $Ti_2FeNiSb_2$, respectively.
- These obtained values indicate that the compounds have lower intrinsic **thermal conductivity** than TiCoSb half Heusler alloy because of the smaller group velocity phonons and the disordered scattering by Fe/Ni. [3]
- **The lattice thermal conductivity** $\mathbf{K_L}$ of $Hf_2FeNiSb_2$ and $Ti_2FeNiSb_2$ are 5.78 and 7.93 $Wm^{-1}K^{-1}$, respectively, i.e., is of about one-third of that of TiCoSb (18.7 $Wm^{-1}K^{-1}$), indicating the decrease in the total thermal conductivity mainly comes from the lattice thermal conductivity contribution. [4]
- **The thermoelectric figure of merit ZT** of $Hf_2FeNiSb_2$ and $Ti_2FeNiSb_2$ reaches only 0.082 and 0.027, respectively, at 874K.









Conclusion

- An investigation of the thermoelectric properties of double half Heusler $Hf_2FeNiSb_2$ and $Ti_2FeNiSb_2$, prepared by melt spinning method was carried out.
- The obtained samples exhibit **the p-type semiconductor** behaviour as indicated from the positive sign of **the Seebeck coefficient.**
- The prepared compounds owe lower intrinsic **thermal conductivity** compared to TiCoSb half-Heusler alloy, attributed to the smaller group velocity phonons and the disordered scattering by Fe/Ni.
- **The thermal conductivity** values have to be further reduced, e.g., by doping of the parent alloys with substitutional elements like Ti.

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