Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Panoscopically optimized thermoelectric performance of half-Heusler/full-Heusler based *in-situ* bulk composite $Zr_{0.7}Hf_{0.3}Ni_{1+x}Sn$: Energy and Time efficient way

A. Bhardwaj[1,2], N. S. Chauhan[1,2] Bhagyashree Sancheti[1], T. D. Senguttuvan[1,2] and D. K. Misra*[1,2],

[1]Physics of Energy Harvesting Division, National Physical Laboratory, Council of Scientific and Industrial Research, New Delhi 110012, India.

[2]Academy of Scientific & Innovative Research (AcSIR), CSIR-National Physical Laboratory (CSIR-NPL) campus, New Delhi-110012, India.

Supplementary Information:

The SEM morphology taken corresponding to HH $Zr_{0.7}Hf_{0.3}NiSn$ clearly reveals a single phase contrasts as shown in Fig. S1(a). Fig S1(b) shows energy dispersive X-ray analysis (EDAX) spectrum recorded from the sample of HH $Zr_{0.7}Hf_{0.3}NiSn$ indicating almost nominal composition of HH $Zr_{0.7}Hf_{0.3}NiSn$. However, the SEM morphology obtained from composite of HH(1-x)/FH(x) with compositions $Zr_{0.7}Hf_{0.3}Ni_{1+x}Sn$ (0.03, 0.05 & 0.10) with increasing FH fractions display two phase contrasts which are presented in supporting information S1(c, e & f). One can clearly see that with increasing FH fractions, the size of FH grains also increases and these grains were noted to be of varying length scales S1(c, e & f). The compositional analysis of these phases was performed employing EDAX to verify the homogeneities of the samples. The compositions of HH and FH in all the compositions $Zr_{0.7}Hf_{0.3}Ni_{1+x}Sn$ have been quantified and averaged from values taken at 8 positions and no obvious impurities phase were observed other than HH and FH in any of the composite samples. A representative EDS spectrum and quantification of elements for the best performing sample $Zr_{0.7}Hf_{0.3}Ni_{1+x}Sn$ with x=0.03, have been presented in fig. S1(d). The

EDAX analysis confirms the minor phase to be full-Heusler, while matrixes phase to be HH, which are consistent with the XRD results.

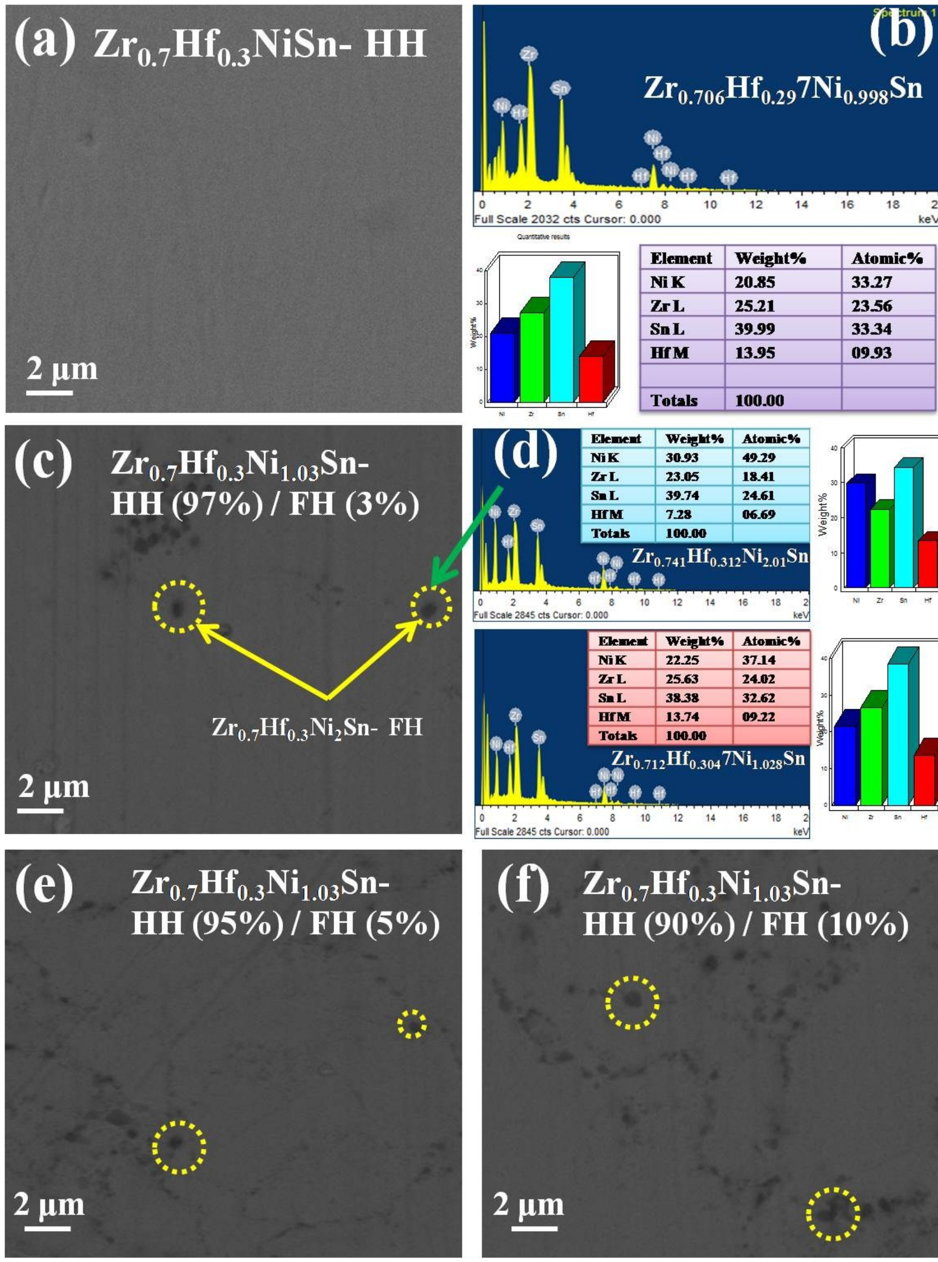

Figure S1: (a) The back scattered SEM micrograph of $Zr_{0.7}Hf_{0.3}NiSn$ representing a single phase contrast which corresponds to half heusler phase confirmed by the EDAX pattern shown in Fig (b); (c) SEM of $Zr_{0.7}Hf_{0.3}Ni_{1.03}Sn$ representing dominantly two phase contrasts; (d) the EDAX pattern of dark contrast and overall phase; (e & f) SEM micrograph of HH (1-x)/FH(x) $Zr_{0.7}Hf_{0.3}Ni_{1+x}Sn$ (0.05 & 0.10) composites showing dominantly two phase contrasts with varying FH grain sizes.

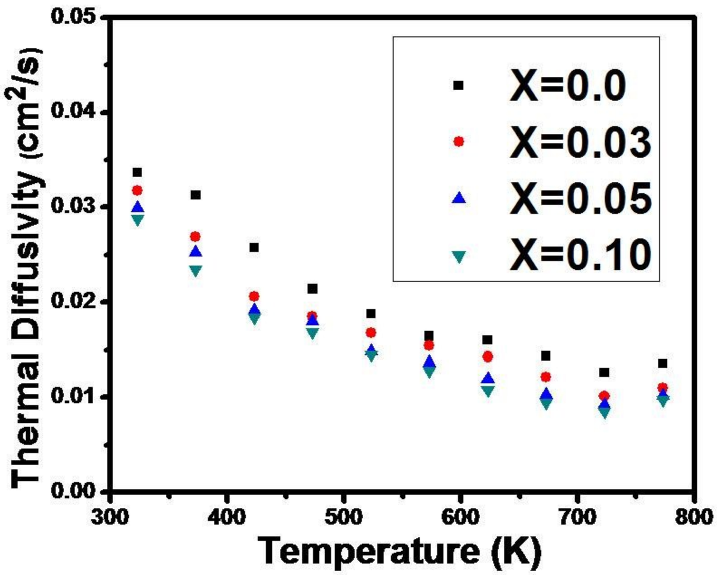

Figure S2: Temperature dependence of thermal diffusivity of composite of HH(1-x)/FH(x) with compositions $Zr_{0.7}Hf_{0.3}Ni_{1+x}Sn$ (0, 0.03, 0.05 & 0.10).

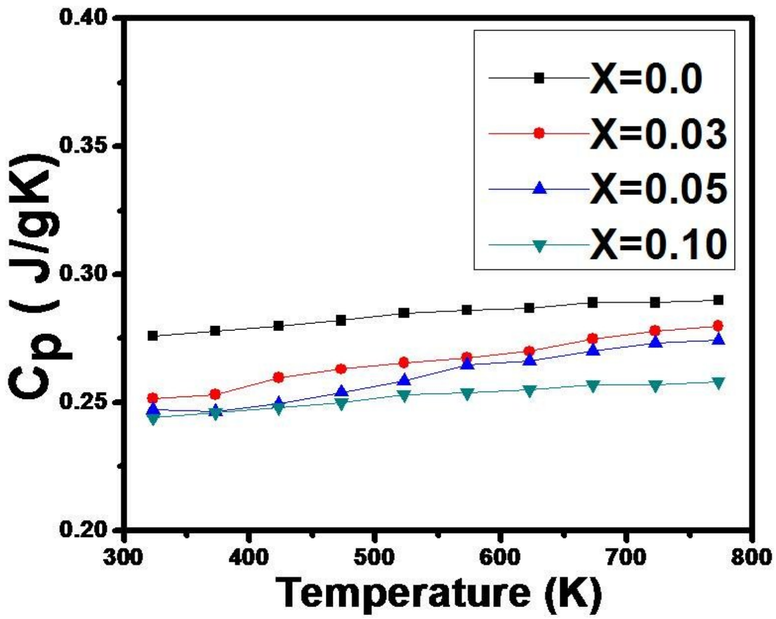

Figure S3: Temperature dependence value of specific heat (c_p) of composite of HH(1-x)/FH(x) with compositions $Zr_{0.7}Hf_{0.3}Ni_{1+x}Sn$ (0, 0.03, 0.05 & 0.10).

Table ST1: Crystal density data along with geometrical pellet density of HH (1-x)/FH(x) composites derived from the compositions $Zr_{0.7}Hf_{0.3}Ni_{1+x}Sn$ ($0.0 \leq x \leq 0.10$).

Composition:	$Zr_{0.7}Hf_{0.3}NiSn$	$Zr_{0.7}Hf_{0.3}Ni_{1.03}Sn$	$Zr_{0.7}Hf_{0.3}Ni_{1.05}Sn$	$Zr_{0.7}Hf_{0.3}Ni_{1.1}Sn$
Crystal Density (kg/m^3)	8724.27	8766.52	8805.21	8873.65
Geometrical pellet density (kg/m^3)	8584.68	8677.10	8678.41	8717.47
Relative %	98.4%	98.98%	98.56%	98.24%