Supplementary Information for

Enhanced thermoelectric performance in TiNiSn-based half-Heuslers

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Table S1. Refined lattice parameters (*a*), phase fraction (f), microstrain (S) and goodness-of-fit (χ^2) for the arc-melted $Ti_{1-x}Zr_xNiSn_{0.95}$ phases.

x	*a* (Å)	f	S (%)[a]	χ^2
0	5.9293(1)	0.34(1)	0	2.9
	5.9354(1)	0.66(1)	0.02(1)	
0.05	5.9459(2)	0.16(1)	0.01(1)	2.6
	5.9501(1)	0.84(1)	0	
0.5	5.9909(1)	0.23(1)	0.70(1)	2.4
	6.0301(1)	0.10(1)	0.70(1)	
	6.0612(1)	0.67(1)	0.30(2)	
1	6.1085(1)	0.08(1)	0	2.5
	6.1148(1)	0.92(1)	0	

[a] microstrain: S (%) = π/0.18 (LY-LY_i); LY_i = 0.08°; GW fixed at 0.12°, LY_i = instrumental contribution taken from x = 0 sample.

Fig. S1. Laboratory X-ray powder diffraction patterns for the arc-melted $Ti_{1-x}Zr_xNiSn_{0.95}$ samples after two weeks of annealing at 900 °C. Data were collected over a period of 8 hours on a Bruker D8 Advance diffractometer with monochromated Cu $K_{\alpha 1}$ radiation. Small amounts of full-Heusler (*) and Ti_5Sn_3 (#) remain after prolonged annealing.

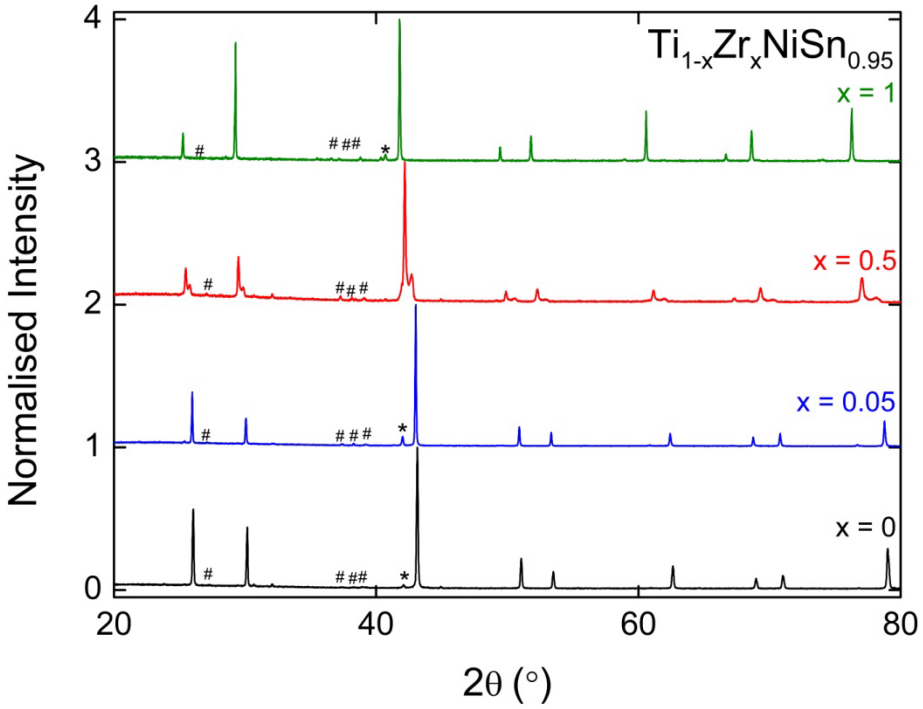

Fig. S2. Thermoelectric power factor (S^2/ρ) for the $Ti_{1-x}Zr_xNiSn_{0.95}$ samples.

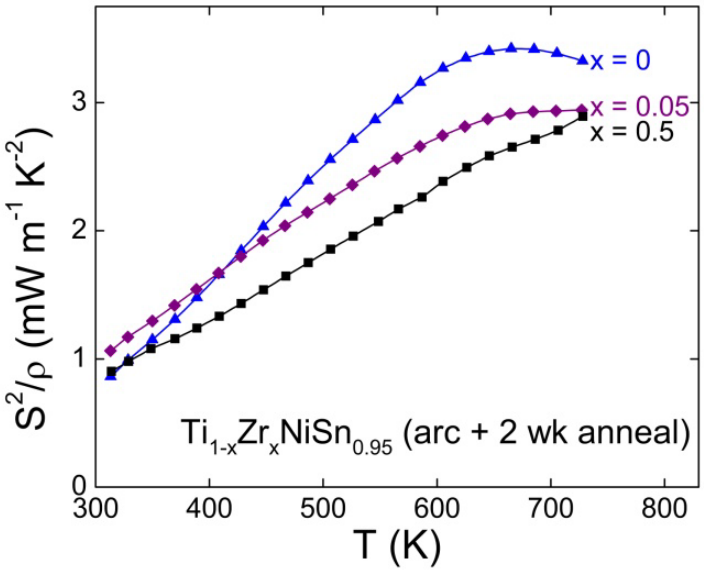

Fig. S3. Rietveld fit to Polaris neutron powder diffraction data for the nominally stoichiometric TiNiSn sample. Prepared by arc-melting and annealing for two weeks at 900 °C. Top markers (blue) are for Ti_5Sn_3 (2.3(1) wt%), middle markers (red) are for $TiNi_{1.76(2)}Sn$ (4.7(2) wt%), and bottom markers are for $TiNi_{1.036(1)}Sn$.

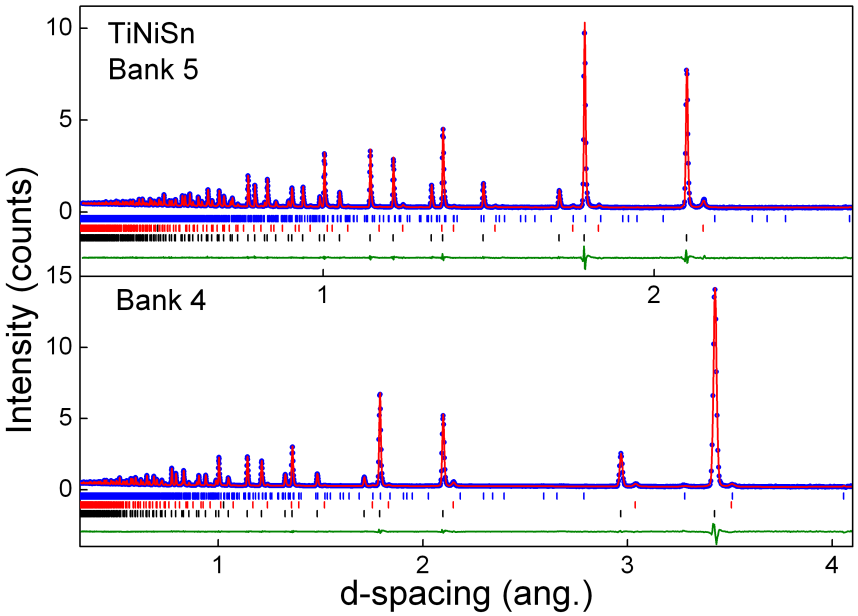

Table S2. Refined structural parameters for nominally stoichiometric TiNiSn from the Rietveld fit against Polaris neutron powder diffraction data.

	Wyckoff	*x*	*y*	*z*	occupancy	U_{iso} (Å^2)
Ti	4a	0	0	0	1.002(2)	0.00478(7)
Ni1	4c	0.25	0.25	0.25	1.000(3)	0.00525(5)
Ni2	4d	0.75	0.75	0.75	0.036(3)	0.00426(5)
Sn	4b	0.5	0.5	0.5	1.000(2)	0.00525(5)

Space group F-43m, a = 5.9327(1) Å; Final fit statistics: $\chi^2 = 6.5$; Bank 5: $wR_p = 2.5\%$, $R_p = 3.8\%$; Bank 4: $wR_p = 1.9\%$, $R_p = 3.6\%$; Bank 3: $wR_p = 2.5\%$, $R_p = 3.1\%$. The sample contains $TiNi_{1.76(2)}Sn$ impurity (4.7(2) wt%, a = 6.0753(3) Å) and Ti_5Sn_3 (2.3(1) wt%) impurities.

Fig. S4. Temperature dependence of the electrical resistivity (ρ), Seebeck coefficient (S), power factor (S^2/ρ), the total (κ) and lattice thermal conductivities ($\kappa_{lat} = \kappa - LT/\rho$; $L = 2.4 \times 10^{-8}$ W Ω K^{-2}), and the thermoelectric figure of merit (ZT) for the arc-melted nominally stoichiometric TiNiSn sample.