Supplementary Material

Physical Properties of {Ti,Zr,Hf}$_2$Ni$_2$Sn Compounds

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Abstract

Physical properties i.e. electrical resistivity (4.2-800K), Seebeck coefficient (300-800K), specific heat (2-110K), Vickers Hardness and elastic moduli (RT) have been defined for single-phase compounds at slightly nonstoichiometric compositions: $Ti_{2.13}Ni_2Sn_{0.87}$, $Zr_{2.025}Ni_2Sn_{0.975}$, $Hf_{2.055}Ni_2Sn_{0.945}$. From X-ray single crystal and TEM analyses $Ti_{2+x}Ni_2Sn_{1-x}$, x~0.13(1), is isotypic with the U_2Pt_2Sn-type (space group $P4_2/mnm$, ternary ordered version of the Zr_3Al_2-type), also adopted by the homologous compounds with Zr, Hf.

For all three polycrystalline compounds (relative densities >95%) the electrical resistivity of the samples is metallic-like with dominant scattering from static defects mainly conditioned by off-stoichiometry. Analyses of the specific heat curves C_p vs T and C_p/T vs T^2 reveal Sommerfeld coefficients of $\gamma_{Ti2Ni2Sn}$ = 14.3(3) mJ/molK^2, $\gamma_{Zr2Ni2Sn}$ = 10(1) mJ/molK^2, $\gamma_{Hf2Ni2Sn}$ = 9.1(5) mJ/molK^2 and low-temperature Debye-temperatures: θ_D^{LT} = 373(7) K, 357(14) K and 318(10) K. Einstein temperatures were in the range 130-155 K. Rather low Seebeck coefficients (<15 μV/K), power factors (pf < 0.07 mW/mK^2) and an estimated thermal conductivity of λ < 148 mWcm^{-1}K^{-1}, yield thermoelectric figures of merit ZT < 0.007 at ~800 K.

Whereas for polycrystalline Zr_2Ni_2Sn elastic properties were determined by resonant ultrasonic spectroscopy (RUS): E = 171 GPa, ν = 0.31, G = 65.5 GPa, B = 147 GPa, the accelerated mechanical property mapping (XPM) mode was used to map hardness and elastic moduli of T_2Ni_2Sn. Above 180 K, Zr_2Ni_2Sn reveals a quasi-linear expansion with CTE = 15.4×10^{-6}K^{-1}.

The calculated density of states is similar for all three compounds and confirms a metallic type of conductivity. The isosurface of *elf* shows a spherical shape for Ti/Zr/Hf atoms and indicates their ionic character, while the $[Ni_2Sn]^{n-}$ sublattice reflects localizations around the Ni and Sn atoms with a large somewhat diffuse charge density between the closest Ni atoms.

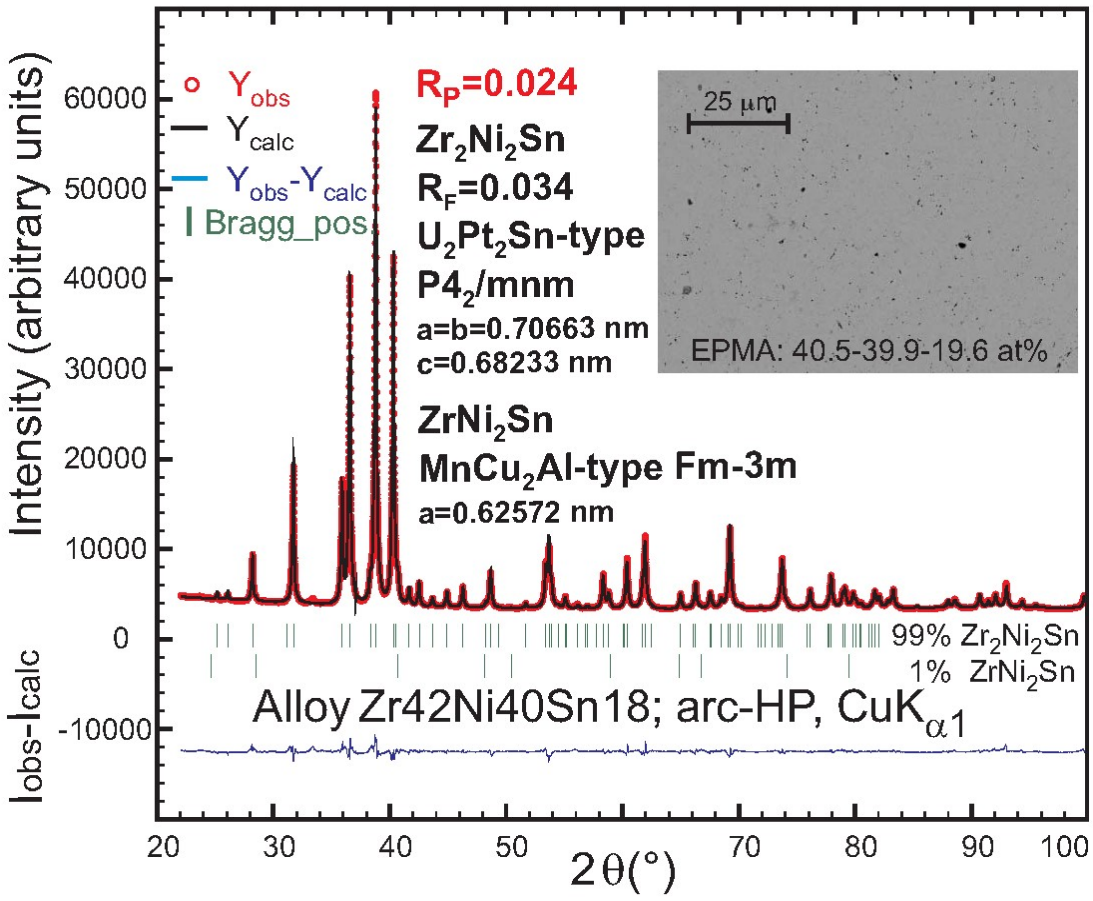

Fig. S1: Rietveld refinement of hot-pressed sample prepared from the nominal composition Zr42Ni40Sn18 (in at.%). The insert shows the microstructure and the composition $Zr_{2+x}Ni_2Sn_{1-x}$, x=0.025 from the corresponding backscatter photo.

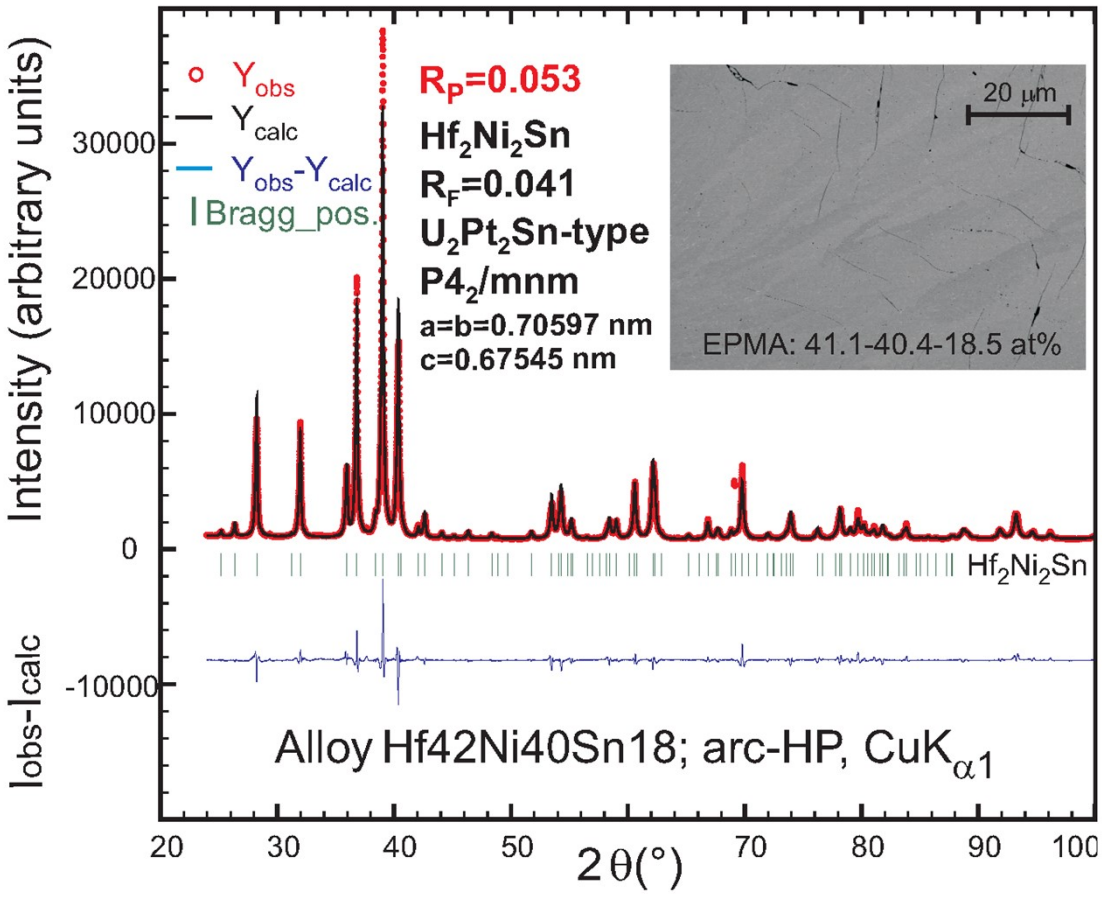

Fig. S2: Rietveld refinement of hot-pressed sample prepared from the nominal composition Hf42Ni40Sn18 (in at.%). The insert shows the microstructure and the composition $Hf_{2+x}Ni_2Sn_{1-x}$, x=0.055 from the corresponding backscatter photo.

Table S1: Interatomic distances < 0.4 nm for $Ti_{2+x}Ni_2Sn_{1-x}$, x ~ 0.13(1). Standard deviations <0.00008 nm.

Atom1	Atom 2	
Ti1	4 Ni1	0.25466
	2 Ni1	0.25926
	4 Sn1	0.29578
	2 Ti2	0.32215
	2 Ti2	0.33477
	1 Ti1	0.35707
	2 Ti2	0.38359
Ti2	2 Ni1	0.25412
	4 Ni1	0.26502
	1 Ti2	0.28426
	4 Sn1	0.30553
	2 Ti1	0.32215
	2 Ti1	0.33477

Atom1	Atom 2	
Ni	1 Ni1	0.24643
	1 Ti2	0.25412
	2 Ti1	0.25466
	1 Ti1	0.25926
	2 Ti2	0.26502
	2 Sn1	0.26932
Sn	4 Ni1	0.26932
	4 Ti1	0.29578
	4 Ti2	0.30553
	2 Sn1	0.32009