Supporting Information

Improving the thermoelectric performance of double half-Heusler compounds ZrNi(In,Sb)

Shiyang He [a,b,ǂ], Amin Bahrami [a*,ǂ], Pingjun Ying [a], Lars Giebeler [a], Xiang Zhang [c], Kornelius Nielsch [a,b], Ran He[a*]

[a] *Leibniz Institute of Solid State and Materials Science, 01069 Dresden, Germany*

[b] *Institute of Materials Science, Technische Universität Dresden, 01062 Dresden, Germany*

[c] *National Center for International Joint Research of Micro-Nano Molding Technology, School of Mechanics and Safety Engineering, Zhengzhou University, 450001 Zhengzhou, China*

* Correspondence to: e-mails: a.bahrami@ifw-dresden.de (A. Bahrami); r.he@ifw-dresden.de (R. He)

ǂ These authors contributed equally to this work.

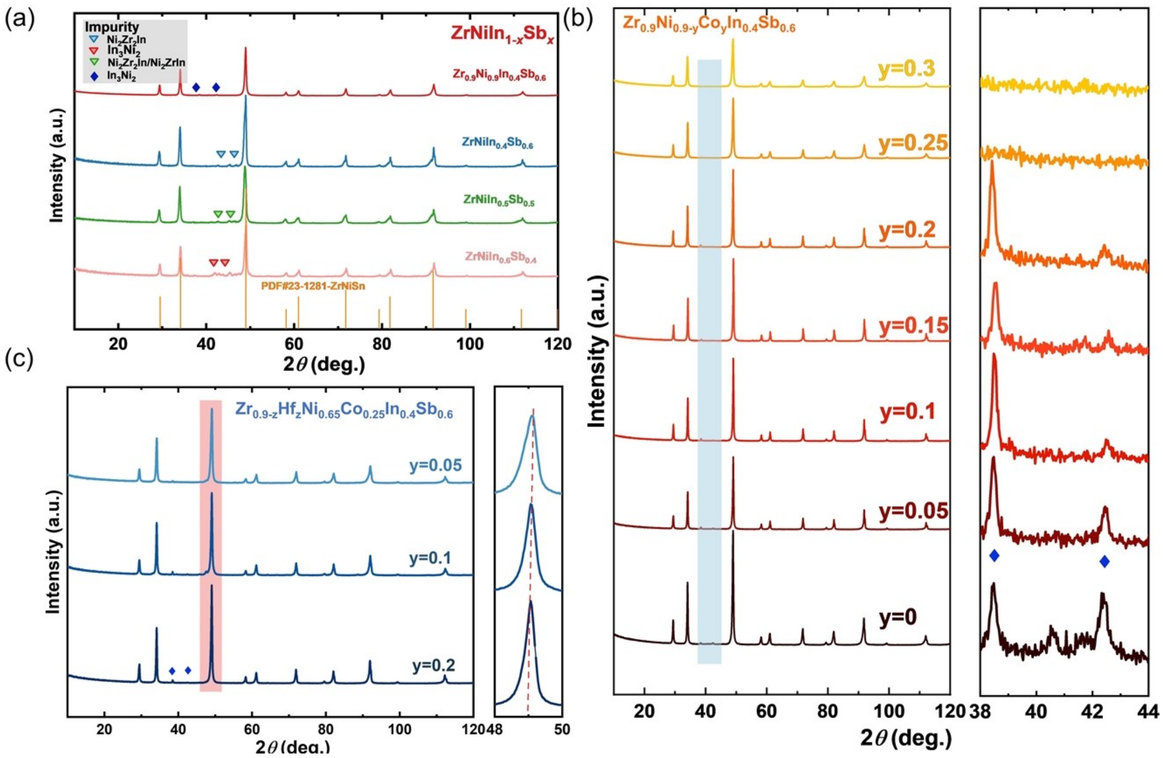

Fig. S1 XRD patterns of $ZrNiIn_xSb_{1-x}$-based materials.

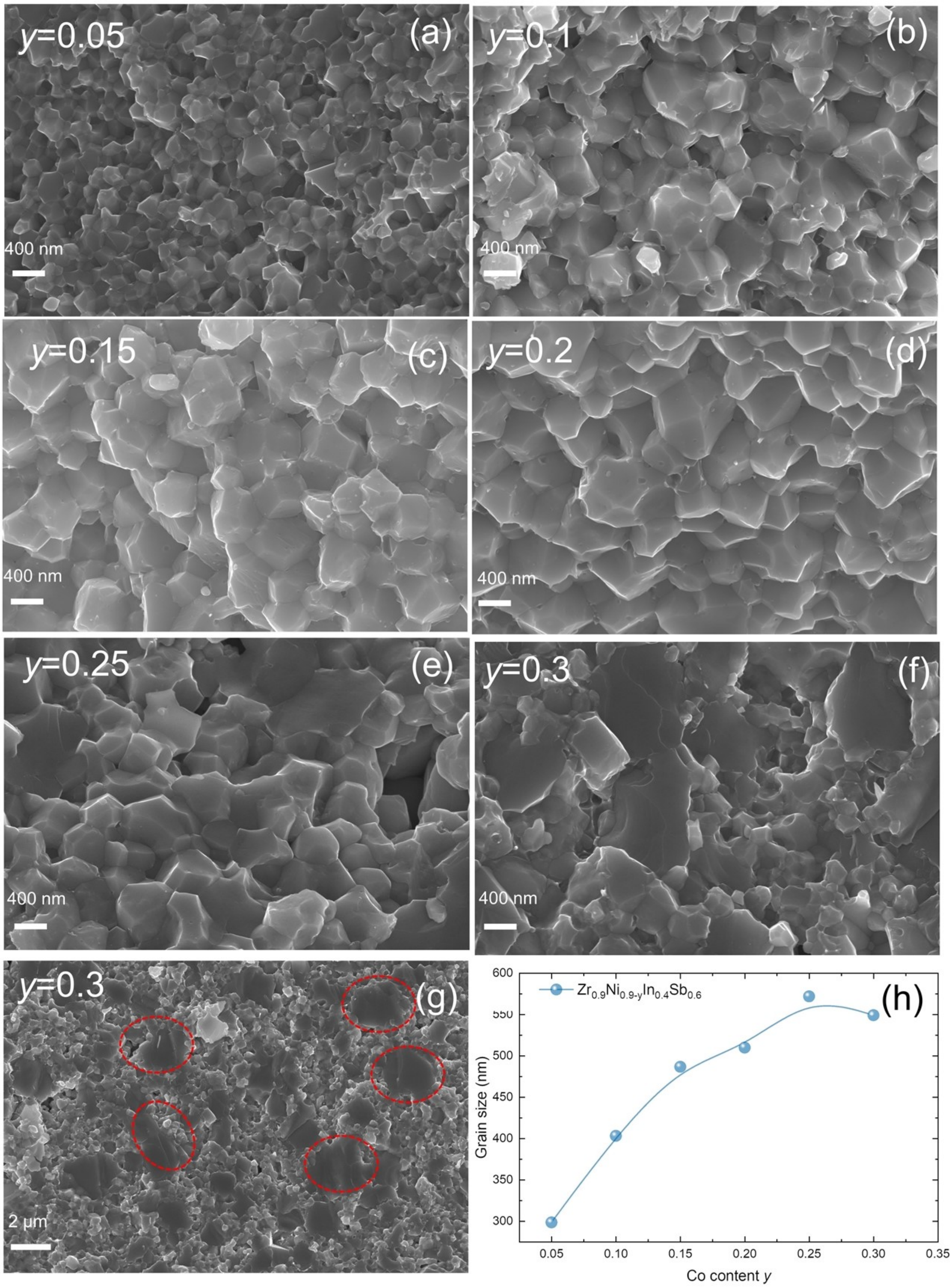

Fig. S2 The SEM images for (a) y=0.05, (b) y=0.1, (c) y=0.15, (d) y=0.2, (e) y=0.25, and (f) - (g) y=0.3 in $Zr_{0.9}Ni_{0.9-y}Co_yIn_{0.4}Sb_{0.6}$ and (h) Co-content dependence of grain size.

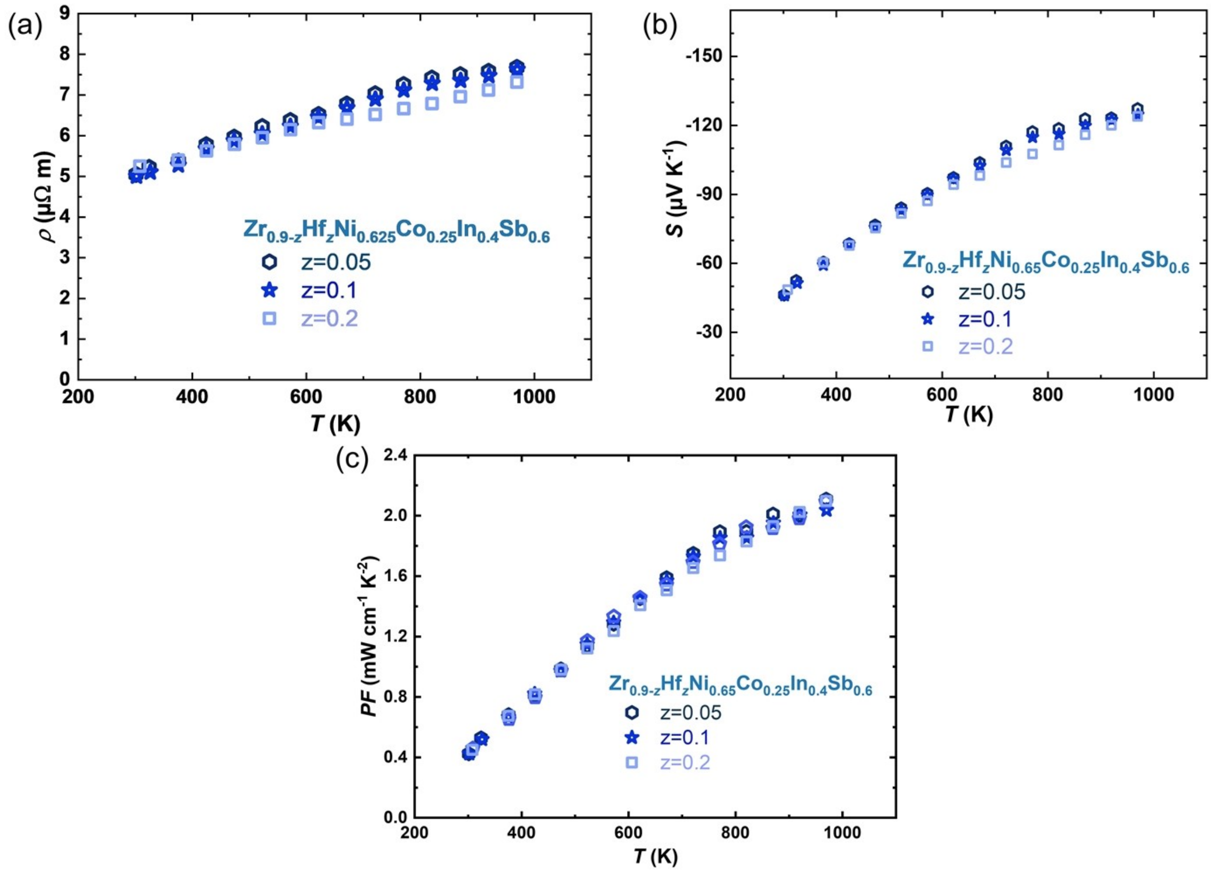

Fig. S3 Temperature dependence of electrical properties for $Zr_{0.9-z}Hf_zNi_{0.625}Co_{0.25}In_{0.4}Sb_{0.6}$ (z = 0.05 - 0.2)

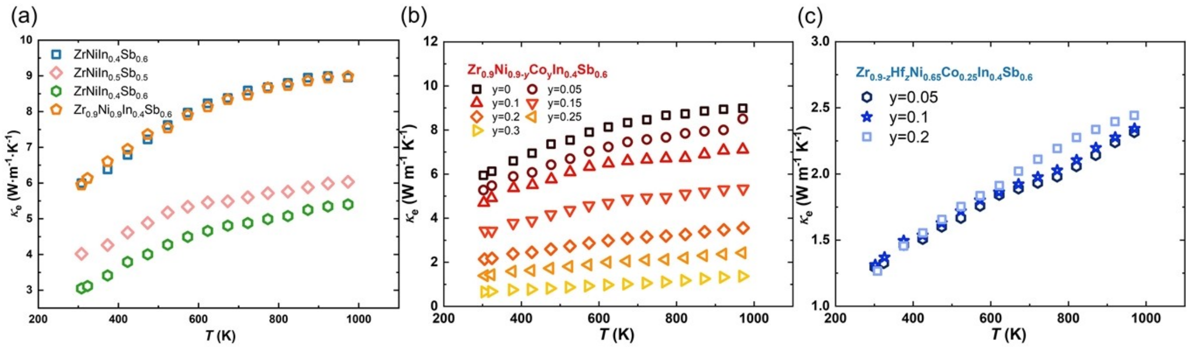

Fig. S4 Temperature dependence of κ_e for $ZrNiIn_xSb_{1-x}$, $Zr_{0.9}Ni_{0.9-y}Co_yIn_{0.4}Sb_{0.6}$ and $Zr_{0.9-z}Hf_zNi_{0.625}Co_{0.25}In_{0.4}Sb_{0.6}$ samples.

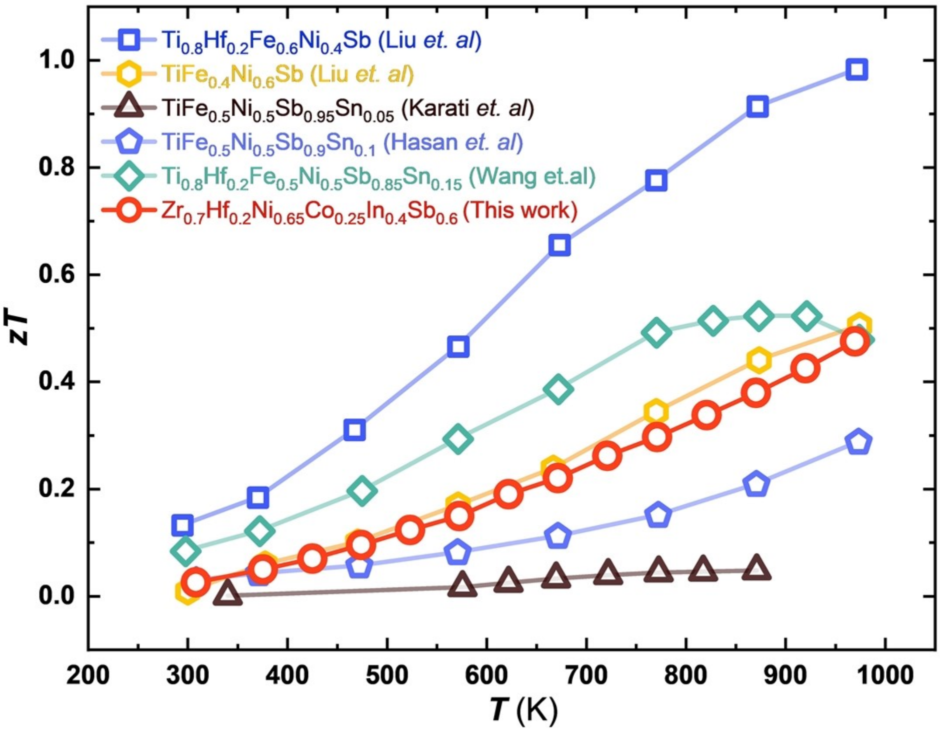

Fig. S5 Comparison of temperature dependence of *zT* for reported double HH compounds[1–4].

Table S1 Results of the Rietveld analyses for compounds with the nominal compositions $ZrNiIn_{0.5}Sb_{0.5}$, $ZrNiIn_{0.4}Sb_{0.6}$, $Zr_{0.9}Ni_{0.9}In_{0.4}Sb_{0.6}$, $Zr_{0.9}Ni_{0.65}Co_{0.25}In_{0.4}Sb_{0.6}$, and $Zr_{0.7}Hf_{0.2}Ni_{0.65}Co_{0.25}In_{0.4}Sb_{0.6}$

	$ZrNiIn_{0.4}Sb_{0.6}$		**$ZrNiSb_{0.5}In_{0.5}$**			**$Zr_{0.9}Ni_{0.9}Sb_{0.4}In_{0.6}$**	**$Zr_{0.9}Ni_{0.65}Co_{0.25}Sb_{0.4}In_{0.6}$**		**$Zr_{0.7}Hf_{0.2}Ni_{0.65}Co_{0.25}Sb_{0.4}In_{0.6}$**		
Phase	ZrNiInSb	Zr_2Ni2In	ZrNiInSb	Zr_2Ni_2In	Ni_2ZrIn	ZrNiInSb	ZrNiInSb	In_3Ni_2*	ZrNiInSb	Ni_2ZrIn	HfO_2
Space group	*F*-43*m*	*P*4$_2$/*mnm*	*F*-43*m*	*P*4/*mbm*	*Fm*-3*m*	*F*-43*m*	*F*-43*m*	*P*-3m*1*	*F*-43*m*	*Fm*-3*m*	*P*1 2$_1$/*c* 1
a / Å		7.1095(4)		7.1579(4)	6.2864(8)			4.3973(4)			5.100(4)
b / Å	6.10130(3)		6.10055(4)			6.09816(1)	6.09368(4)		6.08992(5)	6.2752(9)	5.197(4)
c / Å		6.7742(5)		3.3485(2)				5.2982(10)			5.299(4)
β / °											99.02(6)
γ / °								120			
V / Å^3	227.126(3)	342.40(7)	227.042(5)	171.56(3)	248.43(10)	226.755(1)	226.276(4)	88.72(5)	225.858(6)	247.1(1)	138.7(3)
phase content / wt%	98	2	93	6	1	~98#	99	1	96	3	1
measured reflections	31	296	31	159	34	31	31	120	31	34	408
R_f	1.2	13.3	1.5	13.6	56.7	1.4	1.3	33	1.3	42.7	32
R_I	1.49	11.7	1.6	12.2	46.4	1.6	1.7	31.9	1.6	20.9	41.4

R_{wp}	11.8		13.9			13.3	12.6		12.1	
R_p	9.4		9.6			9.8	8.7		11.2	
R_e	12.2		14.2			12.6	12.6		11.8	
χ^2	0.94		0.95			1.2	1.0		1.1	
atomic parameters										
Zr1 / Hf1	4*a* (0. 0. 0)									
Occ.	1.00 / -		0.998(15) / -			0.933(6) / -	0.932(6) / -		0.471(11) / 0.361	
Biso / Å^2	0.47(4)		0.78(6)			0.48(5)	0.46(4)		0.68(13)	
Ni1 / Co1 / Zr2	4*c* (1/4. 1/4. 1/4)									
Occ.	1.00 / - / -		1.004(13) / - / -			0.946(6) / - / -	0.669(4) / 0.255(4) / 0.016(4)		0.77(13) / 0.24(13) / -	
Biso / Å^2	0.54(3)		0.80(6)			0.54(4)	0.60(5)		0.62(5)	
In1/Sb1	4*b* (1/2. 1/2. 1/2)									
Occ.	0.61 / 0.36		0.51 / 0.46			0.63 / 0.32	0.63 / 0.31		0.67 / 0.27	
Biso / Å^2	0.38(3)		0.48(4)			0.54(4)	0.55(3)		0.48(12)	
Reference	5	6–8	5	9	10	5	5	11,12	5	10

values for minor phase estimated, at least one more phase

* angle γ = 120° for the hexagonal setting of $P\bar{3}m1$, not given in the table for clarity

Table S2 The results of ICP of $ZrNiIn_xSb_{1-x}$ -based double HH.

Nominal Composition	Element content					
$ZrNiIn_{0.4}Sb_{0.6}$		Zr	Ni	In	Sb	
	Measured *wt%*	33.834± 0.35	22.363± 0.25	15.360± 0.16	27.524± 0.15	
	Measured *at%*	33.361	34.272	12.033	20.333	
	Nominal *at%*	33.333	33.333	13.333	20.000	
$ZrNiIn_{0.5}Sb_{0.5}$		Zr	Ni	In	Sb	
	Measured *wt%*	34.215± 0.79	22.316± 0.38	19.554± 0.34	23.054± 0.49	
	Measured *at%*	33.641	34.102	15.275	16.982	
	Nominal *at%*	33.333	33.333	16.67	16.67	
$Zr_{0.9}Ni_{0.9}In_{0.4}Sb_{0.6}$		Zr	Ni	In	Sb	
	Measured *wt%*	33.290± 0.21	21.833± 0.20	14.422± 0.23	30.219± 0.18	
	Measured *at%*	32.855	33.490	11.309	22.345	
	Nominal *at%*	32.143	32.143	14.286	21.429	
$Zr_{0.9}Ni_{0.65}Co_{0.25}Sb_{0.6}In_{0.4}$		Zr	Ni	In	Sb	Co
	Measured *wt%*	33.102± 0.21	15.825± 0.27	14.231± 0.14	30.439± 0.10	5.842± 0.04
	Measured *at%*	32.822	24.388	11.211	22.612	8.967
	Nominal *at%*	32.143	23.214	14.286	21.429	8.929

Reference:

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