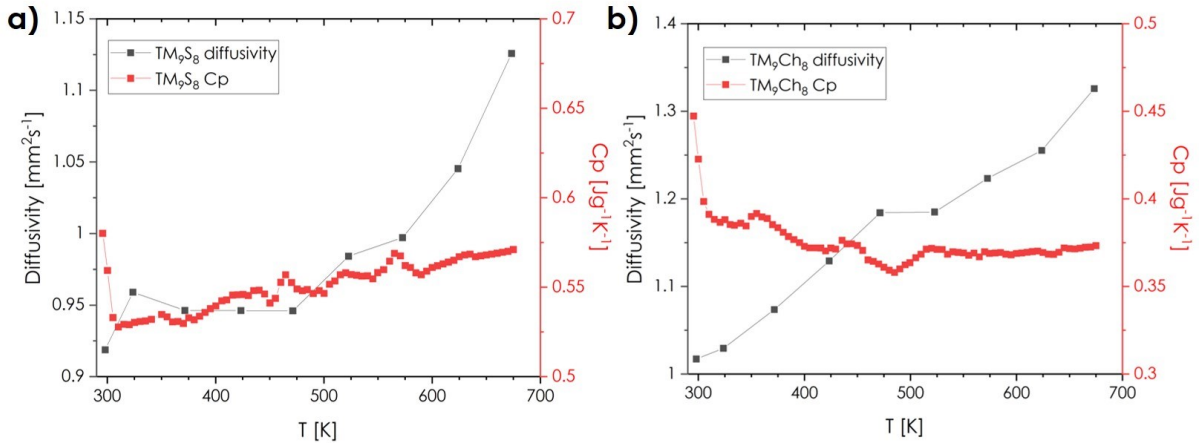
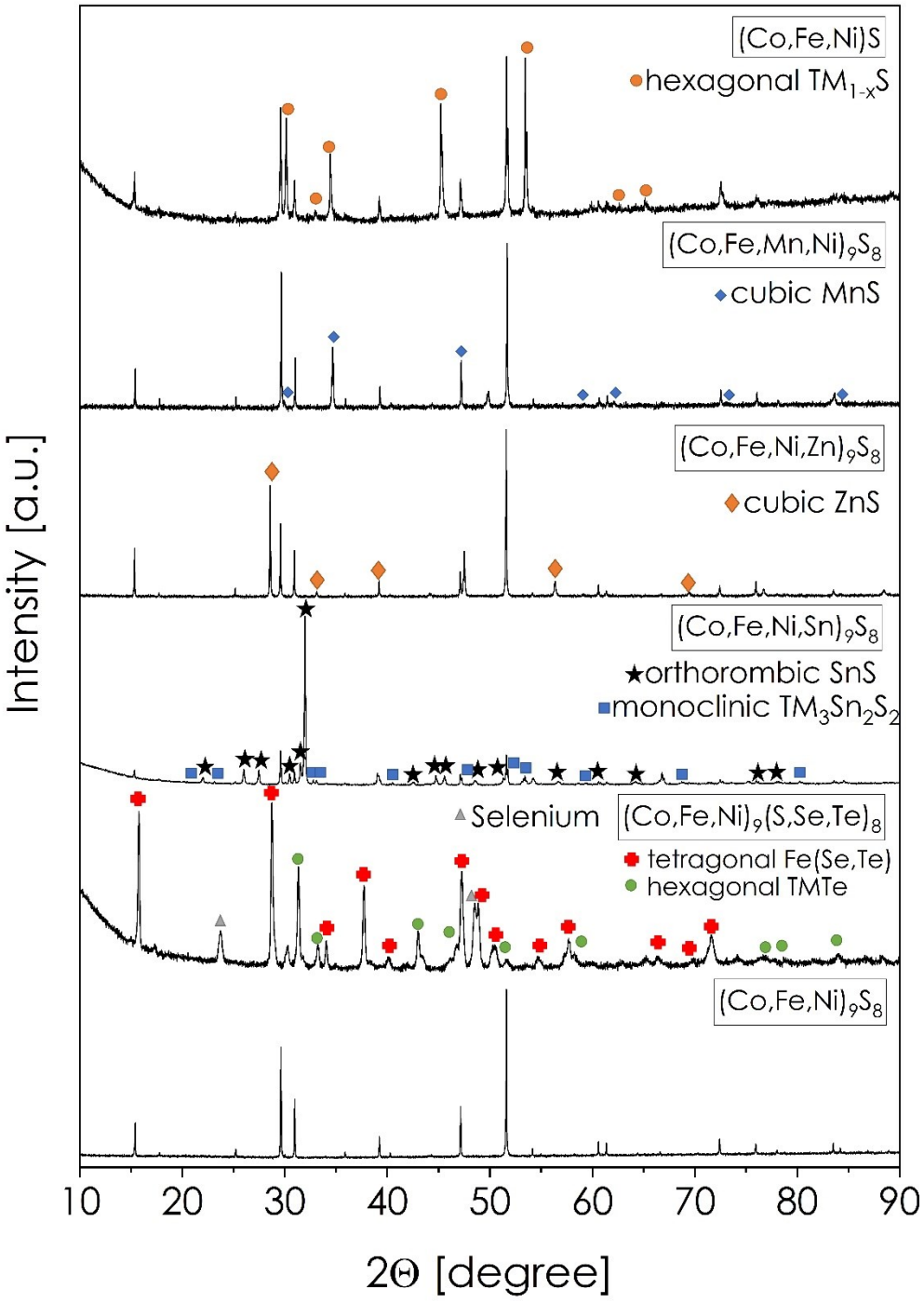
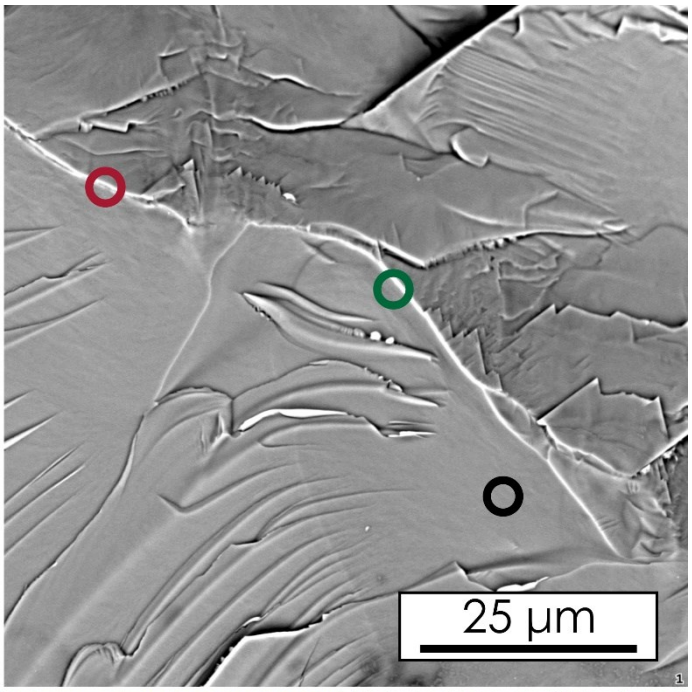
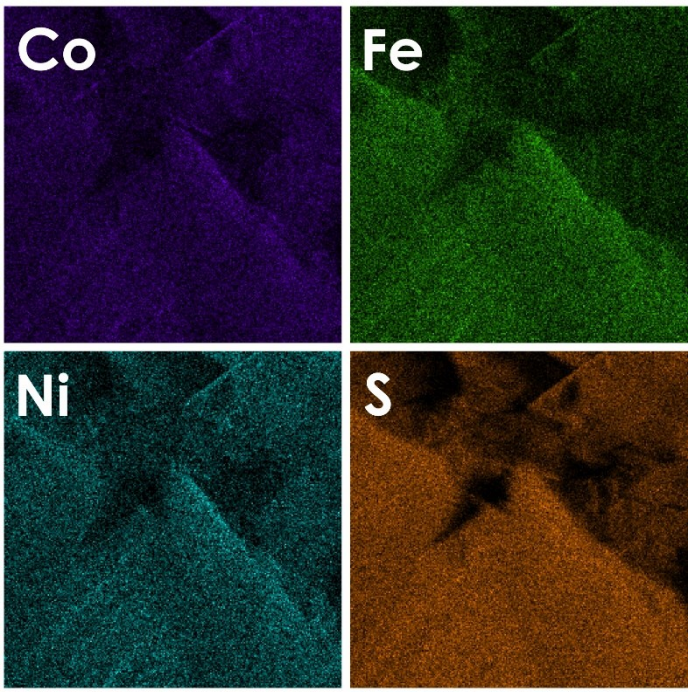


SI.Fig. 1. Thermal diffusivity and specific heat (Cp) measurements for TM_9S_8 (a) and TM_9Ch_8 IHP-sintered pellets.



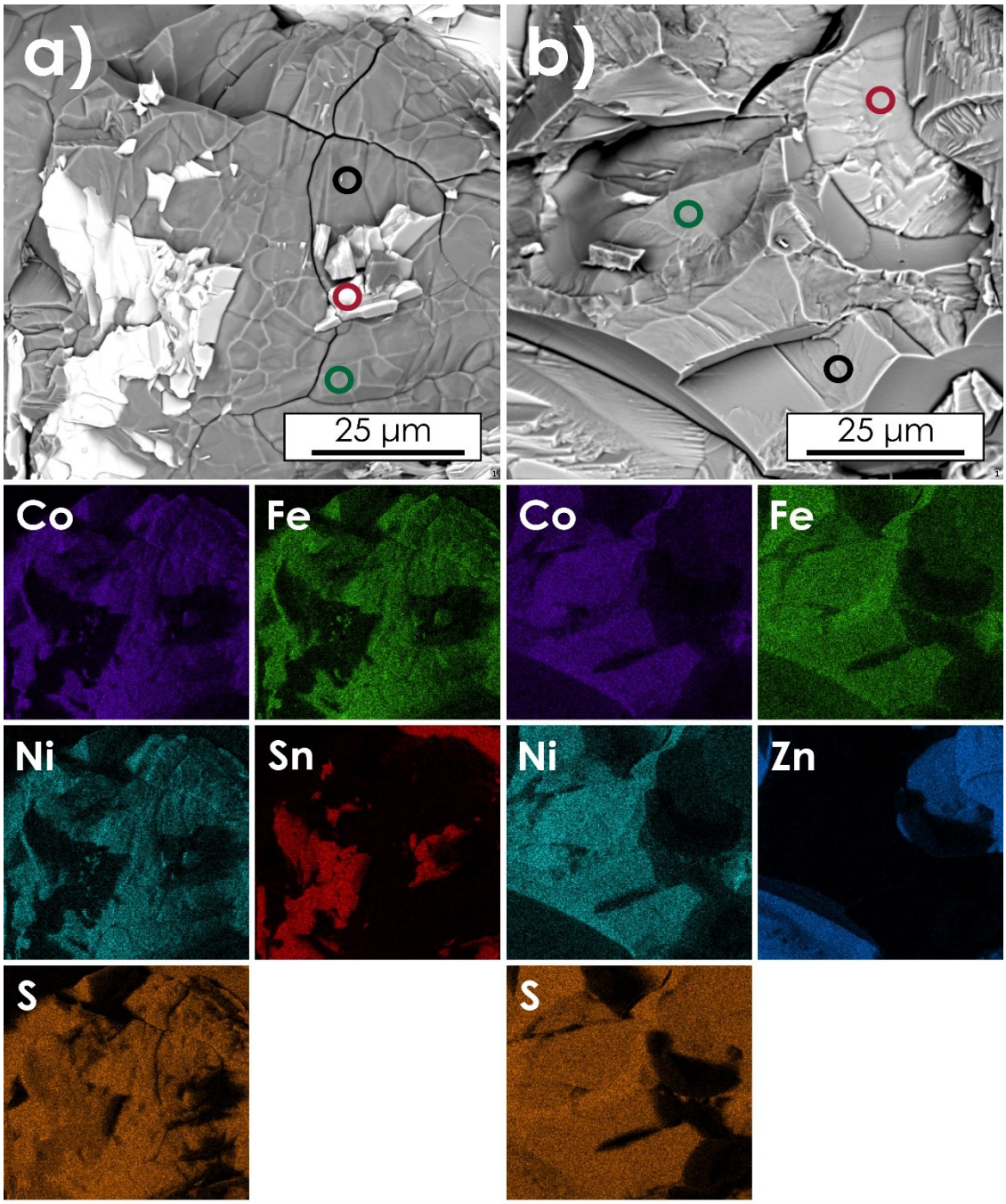
SI.Fig. 2. *XRD diffraction patterns of the multi-phase (Co,Fe,Ni)S, $(Co,Fe,Mn,Ni)_9S_8$, $(Co,Fe,Ni,Sn)_9S_8$, $(Co,Fe,Ni,Zn)_9S_8$, $(Co,Fe,Ni)_9(S,Se,Te)_8$, and single-phase $(Co,Fe,Ni)_9S_8$ pentlandite powder samples.*





	Co	Fe	Ni	S
avg	22.6	19,4	17.3	40.8
○	14.3	22.2	15.8	47.7
○	14.0	22.2	12.8	51.1
○	17.9	19.9	14.6	47.6

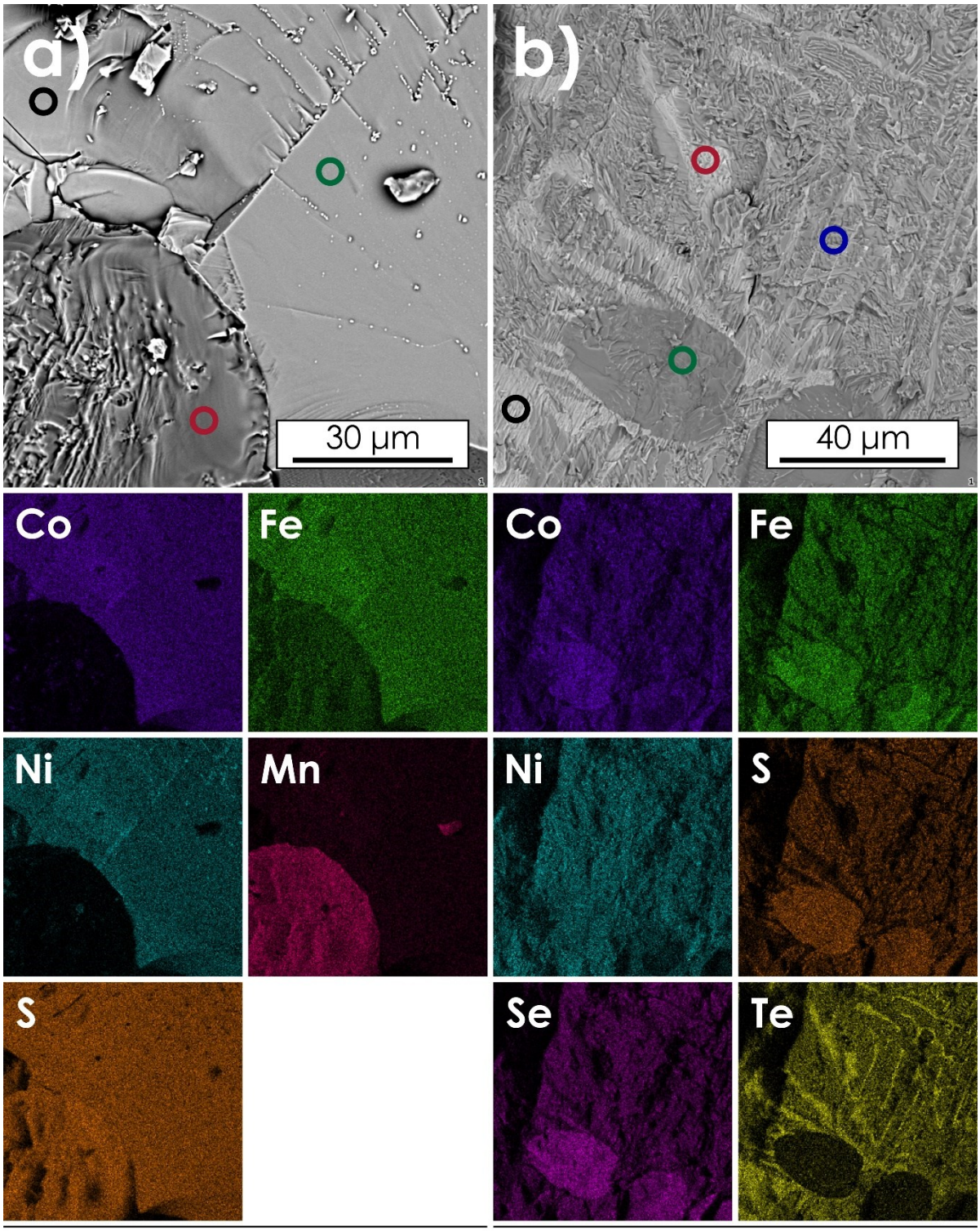
SI.Fig. 3. SEM micrographs together with the corresponding EDS mappings and point/area analysis. All data was collected from the fractured, as-synthesized **(**Co,Fe,Ni)S ingot.



	Co	Fe	Ni	Sn	S
avg	21.3	13.4	16.2	11.4	37.8
O	2.6	1.6	1.8	54.1	39.9
O	23.1	14.1	18.3	0.1	44.4
O	24.6	14.3	19.3	0.9	41.0

	Co	Fe	Ni	Zn	S
avg	14.5	13.8	16.0	12.7	43.0
O	6.3	7.6	1.3	36.1	48.7
O	17.9	12.3	23.4	0.0	46.4
O	19.2	11.0	22.9	0.0	46.9

SI.Fig. 4. SEM micrographs together with the corresponding EDS mappings and point/area analysis. All data was collected from the fractured, as-synthesized **(**Co,Fe,Ni,Sn)$_9$S$_8$ (a), and (Co,Fe,Ni,Zn)$_9$S$_8$ ingots.



	Co	Fe	Ni	Mn	S
avg	16.8	17.9	17.1	24.0	23.5
○	1.6	19.1	0.1	44.7	34.6
○	21.9	14.9	18.7	0.1	44.4
○	21.0	14.9	18.3	0.5	45.3

	Co	Fe	Ni	S	Se	Te
avg	18.0	16.3	16.4	12.4	19.4	17.6
○	18.7	3.3	26.8	1.1	2.5	47.6
○	21.9	15.0	11.1	21.3	27.9	2.9
○	15.3	7.1	22.5	4.0	10.4	40.7
○	16.9	19.2	23.5	8.3	12.0	20.1

SI.Fig. 5. SEM micrographs together with the corresponding EDS mappings and point/area analysis. All data was collected from the fractured, as-synthesized **(**$Co,Fe,Mn,Ni)_9S_8$ (a), and $(Co,Fe,Ni)_9(S,Se,Te)_8$ ingots.

SI. Table 1 Structural data of the TM_9S_8 pentlandites,

Wyckoff position	TM_9S_8 powder		TM_9S_8 pellet	
	Occupancy	B iso	Occupancy	B iso
4a	Co – 0.42, Fe – 0.28, Ni – 0.36	Co – 0.50, Fe – 0.50, Ni – 0.50	Co – 0.42, Fe – 0.26, Ni – 0.32	Co – 0.50, Fe – 0.50, Ni – 0.50
32f	Co – 0.42, Fe – 0.28, Ni – 0.36	Co – 0.50, Fe – 0.5, Ni – 0.5	Co – 0.42, Fe – 0.26, Ni – 0.32	Co – 0.50, Fe – 0.5, Ni – 0.50
8c	S – 1.0	S – 1.15	S – 1.0	S – 1.15
24e	S – 1.0	S – 0.50	S – 1.0	S – 0.50

SI. Table 2 Structural data of the TM_9Ch_8 pentlandites.

Wyckoff position	TM_9Ch_8 powder		TM_9Ch_8 pellet	
	Occupancy	B iso	Occupancy	B iso
4a	Co – 0.30, Fe – 0.34, Ni – 0.36	Co – 1.21, Fe – 1.27, Ni – 1.27	Co – 0.30, Fe – 0.34, Ni – 0.36	Co – 1.21, Fe – 1.27, Ni – 1.27
32f	Co – 0.30, Fe – 0.34, Ni – 0.36	Co – 1.21, Fe – 1.27, Ni – 1.27	Co – 0.30, Fe – 0.34, Ni – 0.36	Co – 1.21, Fe – 1.27, Ni – 1.27
8c	S – 0.50, Se – 0.50	S – 0.78, Se – 0.40	S – 0.44, Se – 0.56	S – 0.78, Se – 0.50
24e	S – 0.50, Se – 0.50	S – 0.78, Se – 0.40	S – 0.60, Se – 0.40	S – 0.78, Se – 0.45