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# **Supporting Information**

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Mechanically Robust BiSbTe Alloys with Superior Thermoelectric Performance: A Case Study of Stable Hierarchical Nanostructured Thermoelectric Materials

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#### Supporting Information

# Mechanical robust BiSbTe alloys with superior thermoelectric performance: A case study of stable hierarchical nanostructured thermoelectric materials

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Figure S1. XRD patterns of MS powders before sintering.



Figure S2. Free fracture surfaces of MS-PAS samples.



Figure S3. Backscattered electron (BSE) images of polished surfaces of MS-PAS samples.



**Figure S4.** Backscattering image (BSI) and elemental mapping of Bi, Sb and Te of carefully polished surface for MS10 sample.



**Figure S5.** (a) Temperature dependence of carrier concentration of ZM and MS-PAS bulk specimens. (b) Electron and phonon mean free path donoted as EMFP and PMFP, respectively, as a function of MS linear speed. The linear speed of 0 m/s corresponds to the ZM ingot.



**Figure S6.** High speed video observations of crack propagation in compressive test for ZM and MS10 samples.



**Figure S7.** Optical images of micro bricks (dimension  $1.6 \times 1.4 \times 1.4 \text{ mm}^3$ ) cut from (a) (c) ZM ingots and (b) (d) MS10 specimens. The arrows indicate the machining defects in ZM ingots such as surface cracks, edge chipping, pitting and delamination.



**Figure S8** FESEM images of free fracture surface of (a)-(b) unannealed and (c)-(d) annealed MS10 specimens.



**Figure S9.** Plasma activated sintering profile of Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub> powders.



Figure S10. The flow chart of MS-PAS process. The size of PAS bulk material is  $\Phi 20x15$  mm.

**Table S1** The table lists the number of samples measured at room temperature, °C and 200 °C, and temperature dependent flexural strength of ZM and MS-PAS samples fitted by Weibull distribution.

Specimen	Temperature [°C]	Number of samples	Characteristic strength [Mpa]	Weibull modulus
ZM	25	12	9.6 (6.1, 12.2)	3.2 (1.2, 5.3)
MS10	25	16	65.2 (51.8, 70.7)	11.4 (10.0, 12.7)
	100	14	65.6 (53.4, 76.5)	9.6 (7.9, 11.4)
	200	11	56.3 (50.2, 61.5)	13.2 (9.8, 16.6)
MS20	25	15	66.2 (55.8, 72.7)	12.2 (10.5, 13.9)
	100	10	70.1 (54.8, 76.5)	10.0 (8.4, 11.7)
	200	10	61.1 (47.6, 65.1)	10.2 (7.8, 12.6)
MS30	25	12	64.0 (57.2, 73.0)	12.8 (7.7, 18.0)
	100	10	65.2 (59.9, 72.6)	13.0 (8.2, 17.8)
	200	11	61.7 (46.3, 68.7)	8.4 (6.8, 10.0)
MS40	25	14	69.6 (54.0, 80.8)	6.8 (5.4, 8.2)
	100	12	72.8 (66.2, 77.9)	18.5 (15.2, 21.8)
	200	11	65.1 (54.9, 70.7)	11.5 (9.1, 13.9)

**Table S2.** The table lists the number of samples measured at room temperature,  $100 \,^{\circ}$ C and  $200 \,^{\circ}$ C, and temperature dependent compressive strength of ZM and MS-PAS samples fitted by Weibull distribution.

Specimen	Temperature [°C]	Number of samples	Characteristic strength [Mpa]	Weibull modulus
ZM	25	14	12.7 (6.3, 18.0)	3.6 (2.9, 4.2)
	100	15	10.2 (6.7, 12.7)	4.3 (3.1, 5.6)
	200	15	10.7 (7.0, 13.4)	5.7 (5.0, 6.5)
MS10	25	13	113.7 (59.8, 140.1)	4.0 (3.5, 4.5)
	100	11	96.3 (64.7, 140.8)	3.3 (2.2, 4.5)
	200	10	84.6 (71.2, 88.3)	18.4 (14.1, 22.7)
MS20	25	15	133.7 (106.0, 144.4)	15.2 (11.5, 18.9)
	100	14	108.2 (88.0, 111.0)	14.3 (9.2, 19.4)
	200	15	94.1 (78.1, 96.6)	12.4 (8.8, 16.0)
MS30	25	11	116.3 (100.7, 122.6)	14.9 (11.6, 18.3)
	100	10	101.6 (90.9, 104.8)	17.6 (10.8, 24.4)
	200	10	95.4 (75.3, 99.3)	21.9 (12.0, 31.9)
MS40	25	14	135.3 (110.5, 155.5)	17.8 (13.6, 22.0)
	100	12	119.4 (92.8, 131.1)	14.3 (10.2, 18.4)
	200	11	111.8 (98.6, 115.6)	19.5 (16.3, 22.7)