Supplementary Information

Realizing high thermoelectric performance in MXene incorporated $Yb_{0.4}Co_{3.96}Ti_{0.04}Sb_{12}$ via carrier and phonon engineering

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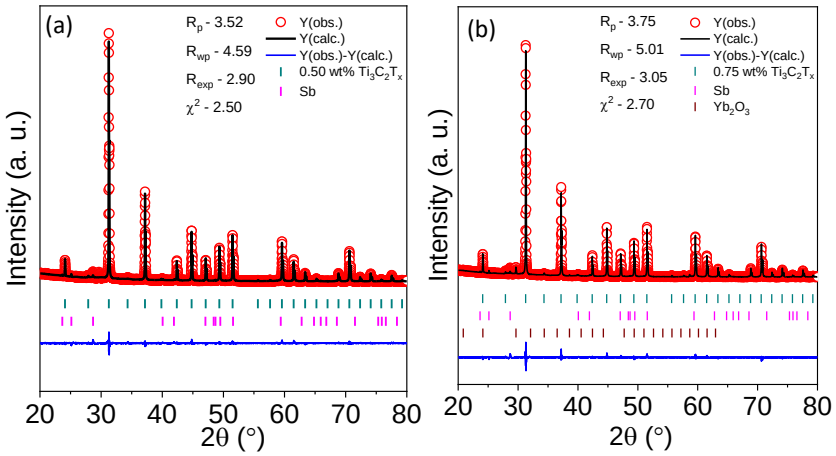

Fig. S1 The Rietveld refinement of (a) 0.50 wt% $Ti_3C_2T_x$/ $Yb_{0.4}Co_{3.96}Ti_{0.04}Sb_{12}$ and (b) 0.75 wt% $Ti_3C_2T_x$/ $Yb_{0.4}Co_{3.96}Ti_{0.04}Sb_{12}$

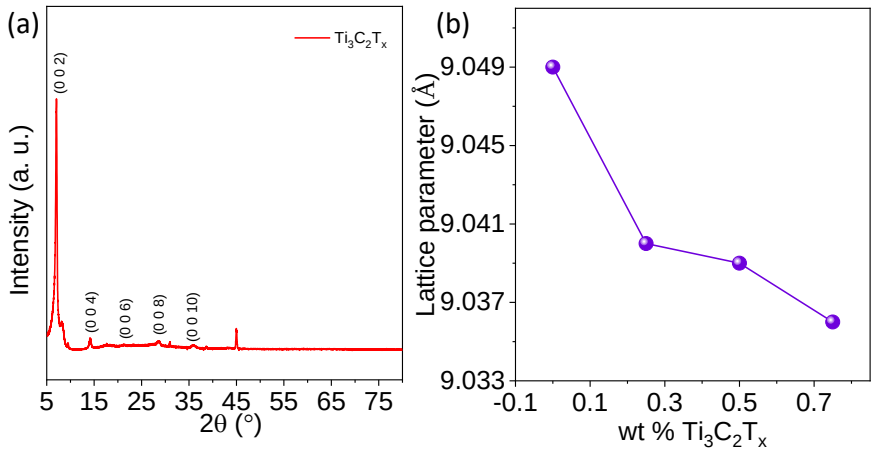

Fig. S2 (a) Powder XRD pattern of $Ti_3C_2T_x$ phase (b) Variation of lattice parameters with $Ti_3C_2T_x$ fraction

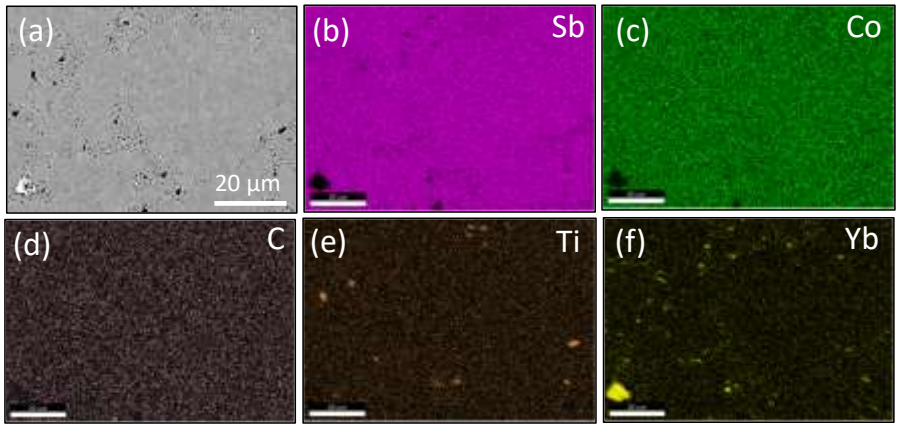

Fig. S3 BSE micrograph (a) and elemental mapping of (b) Sb, (c) Co, (d) C, (e) Ti, and (f) Yb elements in 0.25 wt % $Ti_3C_2T_x$

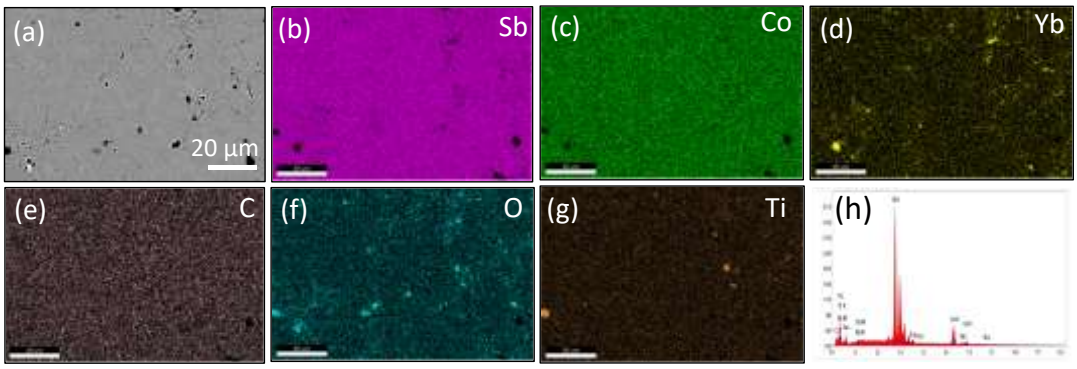

Fig. S4 BSE micrograph (a) and elemental mapping of (b) Sb, (c) Co, (d) Yb, (e) C, (f) O, and (g) Ti elements in 0.75 wt % $Ti_3C_2T_x$. (h) EDS spectra

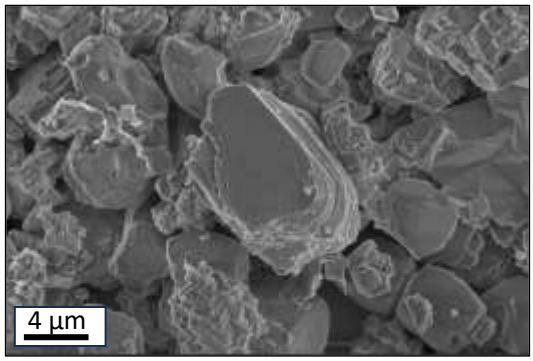

Fig. S5 Secondary electron micrograph of $Ti_3C_2T_x$ layers

Table S1

Composition	***d*** ***(gcm^{-3})***	**Lattice constant** ***a (Å)***	**Volume** ***(Å^3)***
$Yb_{0.4}Co_{3.96}Ti_{0.04}Sb_{12}$	7.74 (> 98 %)	9.049(0)	740.97
0.25 wt % $Ti_3C_2T_x$	7.59 (> 98 %)	9.040(8)	738.76
0.50 wt % $Ti_3C_2T_x$	7. 55 (> 98 %)	9.039(7)	738.68
0.75 wt % $Ti_3C_2T_x$	7.52 (> 98 %)	9.036(1)	737.81

Table S2: Quantitative analysis of Sb3d, Sb3p and Ti3p energy levels

Sample	Sb3d - energy level (eV)				Sb3p - energy level (eV)		Ti3p - energy level
	$Sb3d_{5/2}$		$Sb3d_{3/2}$		$Sb3p_{3/2}$	$Sb3p_{1/2}$	
$Yb_{0.4}Co_{3.96}Ti_{0.04}Sb_{12}$	527.66	529.83	537.13	539.02	811.95	765.95	-
0.25 wt % $Ti_3C_2T_x$	527.84	530.13	537.25	539.29	813.14	766.75	33.99
0.50 wt % $Ti_3C_2T_x$	528.47	530.77	538.54	540.38	813.47	767.41	34.52
0.75 wt % $Ti_3C_2T_x$	526.42	528.50	537.54	539.76	812.99	766.83	34.18

Table S3: Quantitative analysis of Co2p energy level

Sample	Co2p - energy level						O1s Energy level
	$Co2p_{3/2}$		$Co2p_{1/2}$		Satellite peak		
	Co^{3+}	Co^{2+}	Co^{3+}	Co^{2+}	Co^{3+}	Co^{2+}	
$Yb_{0.4}Co_{3.96}Ti_{0.04}Sb_{12}$	777.66	782.88	792.31	797.40	786.61	802.52	-
0.25 wt % $Ti_3C_2T_x$	778.33	783.92	795.22	801.59	789.58	807.76	-
0.50 wt % $Ti_3C_2T_x$	779.26	784.05	794.19	800.28	788.33	805.86	-
0.75 wt % $Ti_3C_2T_x$	778.28	782.59	793.15	799.67	787.08	806.17	530.54

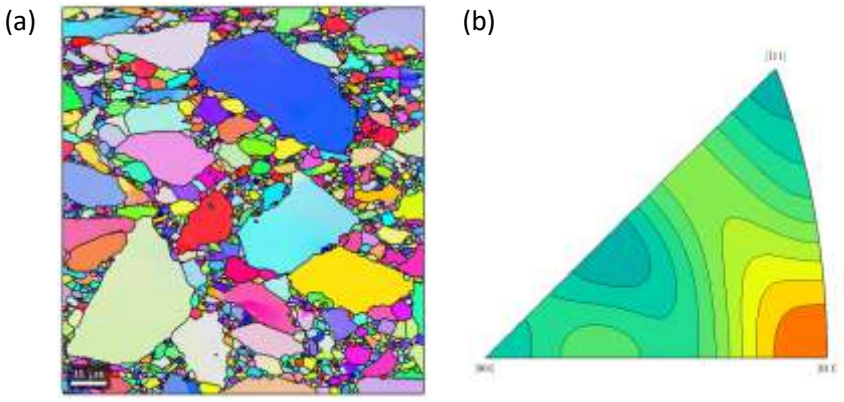

Fig. S6 (a) Inverse pole figure (IPF) map showing the microstructure and (b) Inverse pole figure (IPF) showing the texture along the normal direction (ND) corresponding to $Yb_{0.4}Co_{3.96}Ti_{0.04}Sb_{12}$

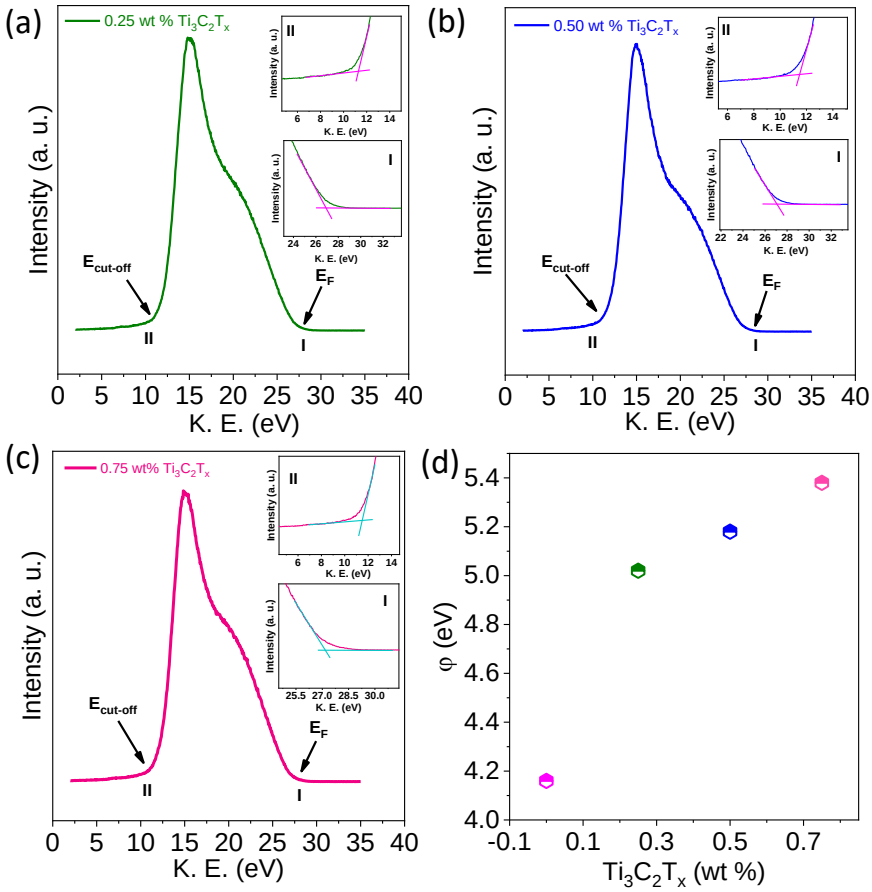

Fig. S7 UPS spectrum of (a) 0.25 wt % $Ti_3C_2T_x$ (b) 0.50 wt % Ti_3C_2Tx (c) 0.75 wt % $Ti_3C_2T_x$ composites, the insets show the close-ups view of the inelastic cut-off $E_{cut\text{-}off}$ (II) and Fermi edge E_F (I) of the UPS spectrum (d) Variation of work function with Mxene weight percentage

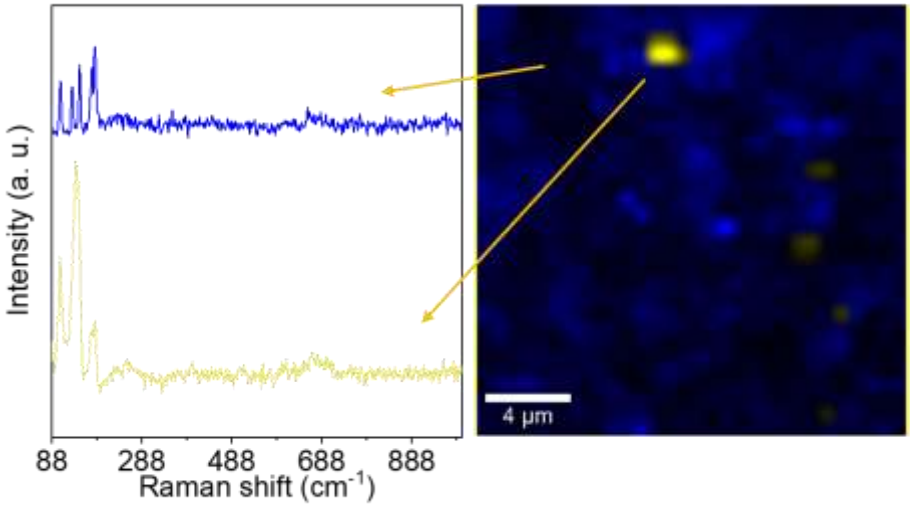

Fig. S8 Raman mapping performed on the selected area where blue region denotes the matrix $Yb_{0.4}Co_{3.96}Ti_{0.04}Sb_{12}$ and yellow region corresponds to $Ti_3C_2T_x$ inclusions rich area.

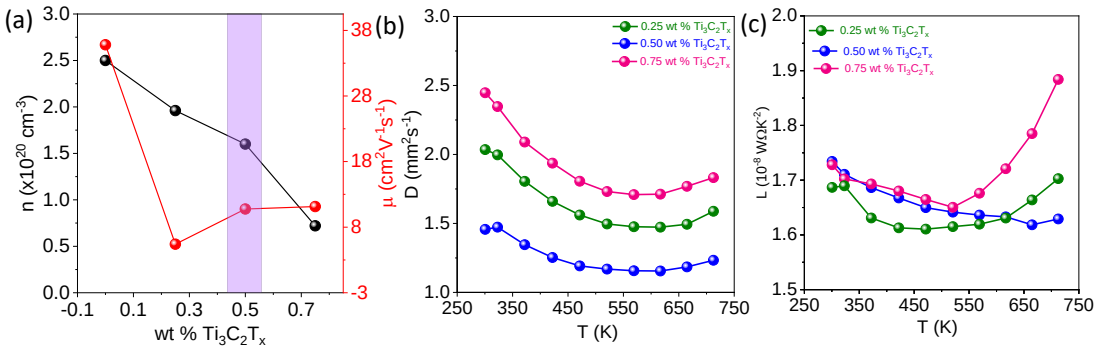

Fig. S9 (a) Variation of carrier concentration and mobility with weight percentage of $Ti_3C_2T_x$ (b) Variation of thermal diffusivity with temperature (c) Variation of Lorenz number with temperature

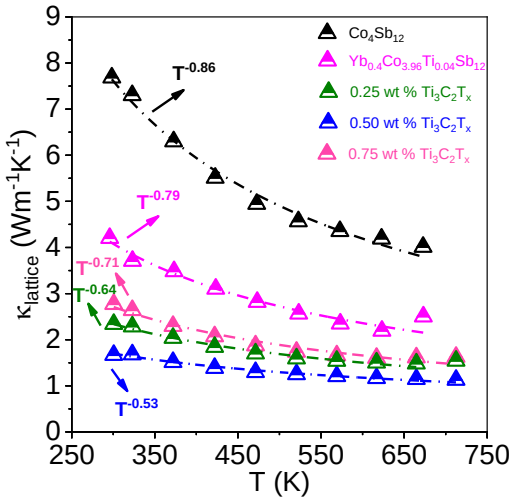

Figure S10 The temperature dependent lattice thermal conductivity data fitted as a function of T^{-x}

Calculations of PF, P_d, zT_{engg}, and η_{max} parameters

The typical thermoelectric efficiency (η_{max}) of a thermoelectric material is evaluated by the dimensionless parameter zT where, the S, σ, and κ values are assumed as temperature independent factors during the calculations. The term ZT_{engg}, a quantitative metric was introduced by H. S. Kim,[1] that evaluates the efficiency of thermoelectric (TE) conversion with the consideration of temperature dependent featured TE properties. This metric is particularly useful for accurately assessing the thermoelectric efficiency of a material with a significant temperature difference between the cold and hot sides of the thermoelectric legs. The calculations are made using the relations mentioned below;

a. Average figure of merit,

$$ZT_{avg} = \frac{\frac{S^2(T)}{\rho(T)} \times T}{\kappa(T)}$$

b. Engineering power factor,

$$PF_{eng} = \frac{\left(\int_{T_c}^{T_H} S(T)\, dT\right)^2 \Delta T}{\int_{T_c}^{T_H} \rho(T)\, dT}$$

c. Engineering figure of merit,

$$(zT)_{eng} = \frac{PF_{eng}}{\int_{T_c}^{T_H} \kappa(T)\, dT}$$

d. Power density,

$$P_d = \frac{PF_{eng}\Delta T}{L} \frac{m_{opt}}{(1+m_{opt})^2}; \qquad m_{opt} = \sqrt{1 + (ZT)_{eng}\left(\alpha/\eta_c - 1/2\right)}$$

e. Maximum TE efficiency,

$$\eta_{max} = \frac{\eta_c \sqrt{1 + (ZT)_{eng}\left(\frac{\alpha}{\eta_c} - \frac{1}{2}\right)} - 1}{\sqrt[\alpha]{1 + (ZT)_{eng}\left(\frac{\alpha}{\eta_c} - \frac{1}{2}\right)} - \eta_c}$$

$$\eta_c = \frac{\Delta T}{T_H}; \qquad \alpha = \frac{S(T_H)\Delta T}{\int_{T_c}^{T_H} S(T) dT}$$

where, ρ (T), S(T), η_C, α and κ (T) represents the resistivity, Seebeck coefficient, Carnot efficiency, Thomson coefficient and total thermal conductivity respectively and m_{opt} is the ratio of external load resistance (R_L) to internal resistance (R_{int}).

References:

(1) Kim, H. S.; Liu, W.; Chen, G.; Chu, C. W.; Ren, Z. Relationship between Thermoelectric Figure of Merit and Energy Conversion Efficiency. *Proc Natl Acad Sci U S A* **2015**, *112* (27), 8205–8210.