Supporting Information

Interfacial modulation to low lattice thermal conductivity and enhanced thermoelectric performance in *n*-type Mg$_3$(Sb, Bi)$_2$-based materials *via* Graphene and MXene

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1. Lorenz number and density of state mass calculations

The Lorenz number (L) and density of state mass (m^*) is calculated using the single parabolic band (SPB) model with measured Seebeck coefficient (S) and carrier concentration (n) according to the following equations [1]:

$$S=\frac{k_{\mathrm{B}}}{e}\left[\left(\frac{(2+\lambda)F_{1+\lambda}(\mu)}{(1+\lambda)F_{\lambda}(\mu)}\right)-\mu\right]$$

$$F_i(\mu)=\int_0^{\infty}\frac{\varepsilon^i}{1+\exp(\varepsilon-\mu)}d\varepsilon$$

$$L=\left(\frac{k_{\mathrm{B}}}{e}\right)^2\left[\frac{\lambda+3}{\lambda+1}\frac{F_{\lambda+2}(\mu)}{F_{\lambda}(\mu)}-\left(\frac{\lambda+2}{\lambda+1}\right)^2\left(\frac{F_{\lambda+1}(\mu)}{F_{\lambda}(\mu)}\right)^2\right]$$

$$m^*=\frac{h^2}{2k_{\mathrm{B}}T}\left(\frac{n}{4\pi F_{1/2}(\mu)}\right)^{\frac{2}{3}}$$

Where μ is reduced Fermi level, $F_i(\mu)$ is Fermi integral, k_{B} is the Boltzmann constant, h is the Planck constant and λ is scattering parameter.

2. Weighted mobility calculation

Weighted mobility (μ_{W}) (electron mobility weighted by the density of electronic states) is obtained using the following equation [2]:

$$\mu_{\mathrm{W}}=\frac{331}{\rho}\left(\frac{T}{300}\right)^{-\frac{3}{2}}\left[\frac{\exp\left(\frac{|S|}{k_B/e}-2\right)}{1+\exp\left[-5\left(\frac{|S|}{k_B/e}-1\right)\right]}+\frac{\frac{3|S|}{\pi^2 k_B/e}}{1+\exp\left[5\left(\frac{|S|}{k_B/e}-1\right)\right]}\right]$$

Where ρ is electrical resistivity. Weighted mobility analysis can elucidate the electronic structure and scattering mechanisms in materials. At the optimal carrier concentration, the maximum of zT depends on the ratio of $\mu_{\mathrm{W}}/\kappa_{\mathrm{L}}$.
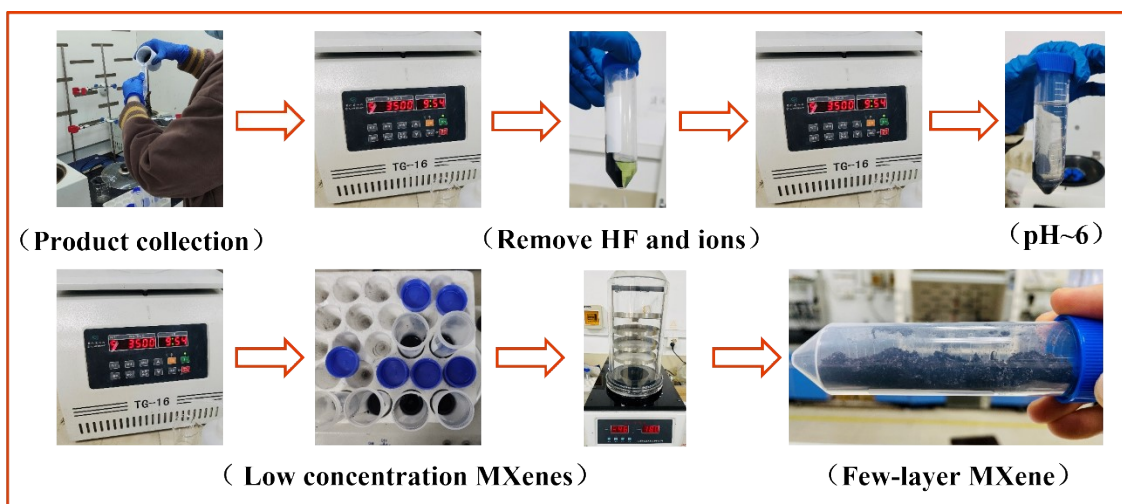
（Product collection）　　　　（Remove HF and ions）　　　　（pH~6）

（Low concentration MXenes）　　　　（Few-layer MXene）

Fig. S1 Few-layer MXene preparation process.

Table S1 The density of $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% Graphene or MXene samples.

sample name	density (g cm^{-3})	relative density (%)
$x = 0$	5.12	98.5
$x = 0.2$ G	5.03	96.7
$x = 0.4$ G	4.96	95.3
$x = 0.6$ G	4.9	94.2
$x = 0.3$ M	5.06	97.3
$x = 0.6$ M	5.02	96.5
$x = 0.9$ M	4.95	95.2
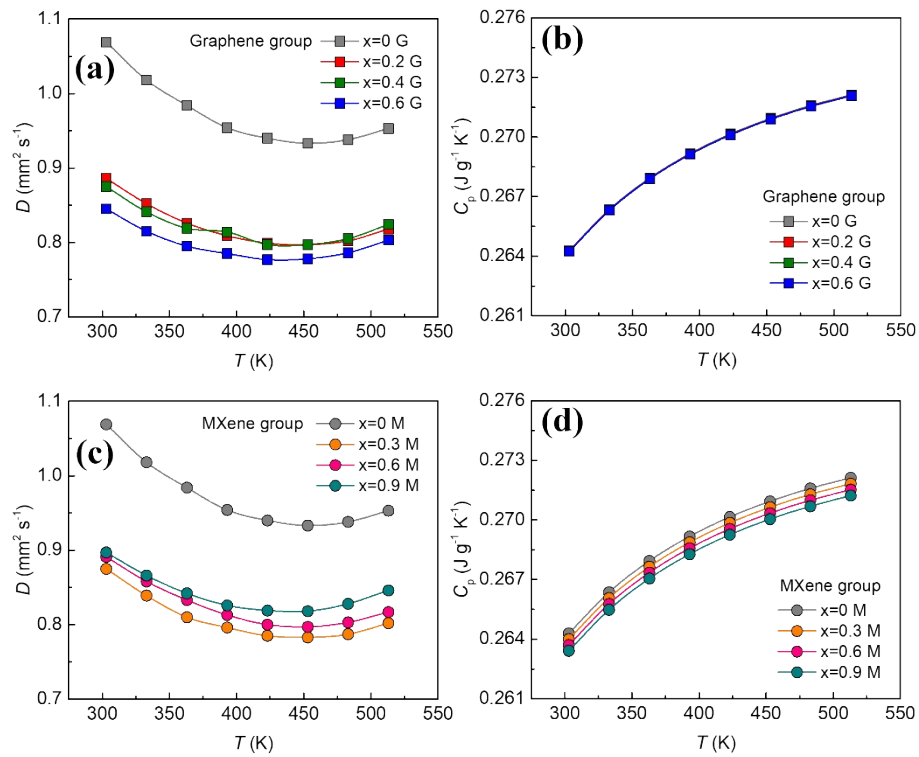
Fig. S2 (a) Thermal diffusion coefficient and (b) specific heat of $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% Graphene. (c) Thermal diffusion coefficient and (d) specific heat of $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% MXene.
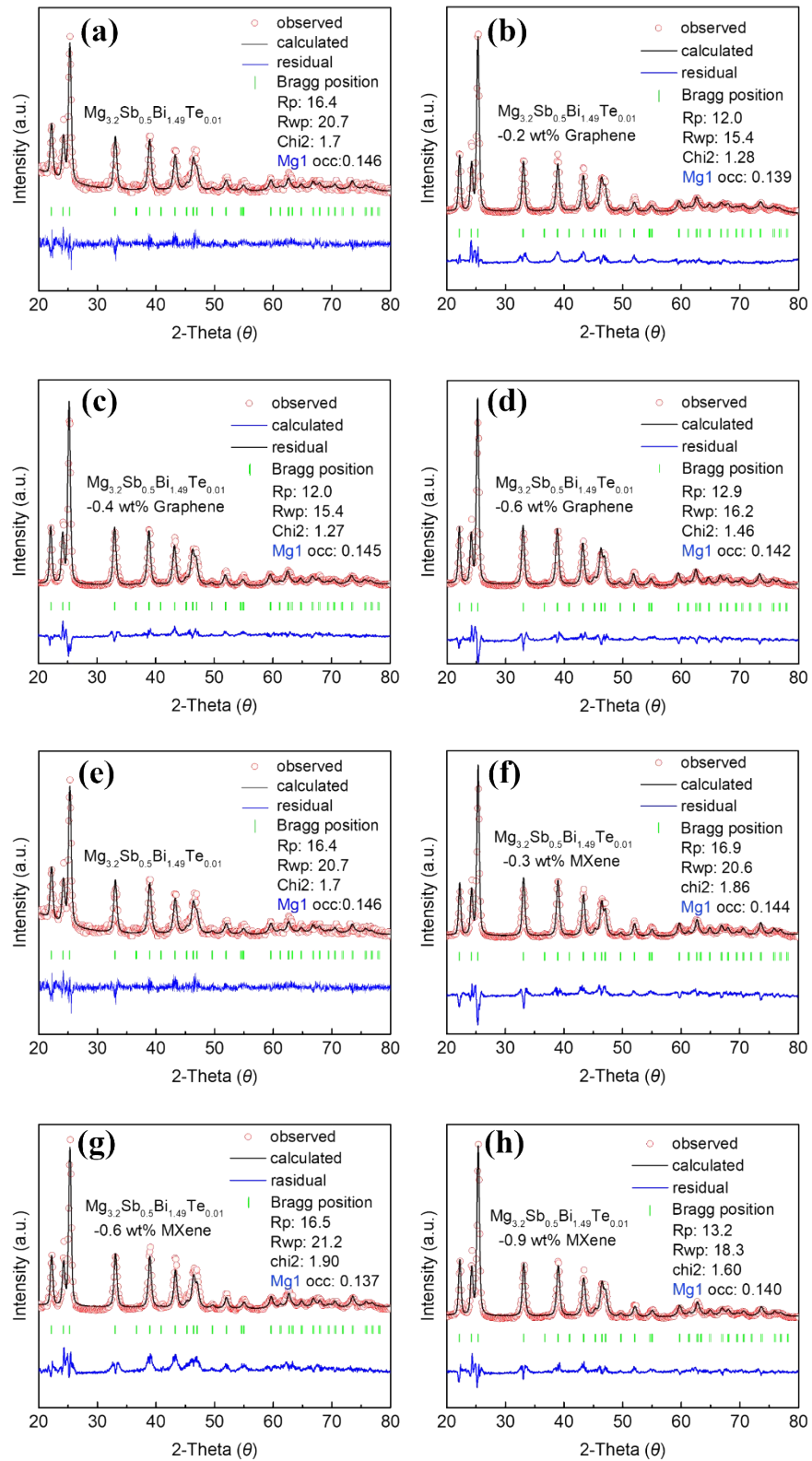
Fig. S3 (a)-(d) The XRD refinement of $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% Graphene (x = 0, 0.2, 0.4 and 0.6. respectively). (e)-(h) The XRD refinement of $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% MXene (x = 0, 0.3, 0.6 and 0.9. respectively).
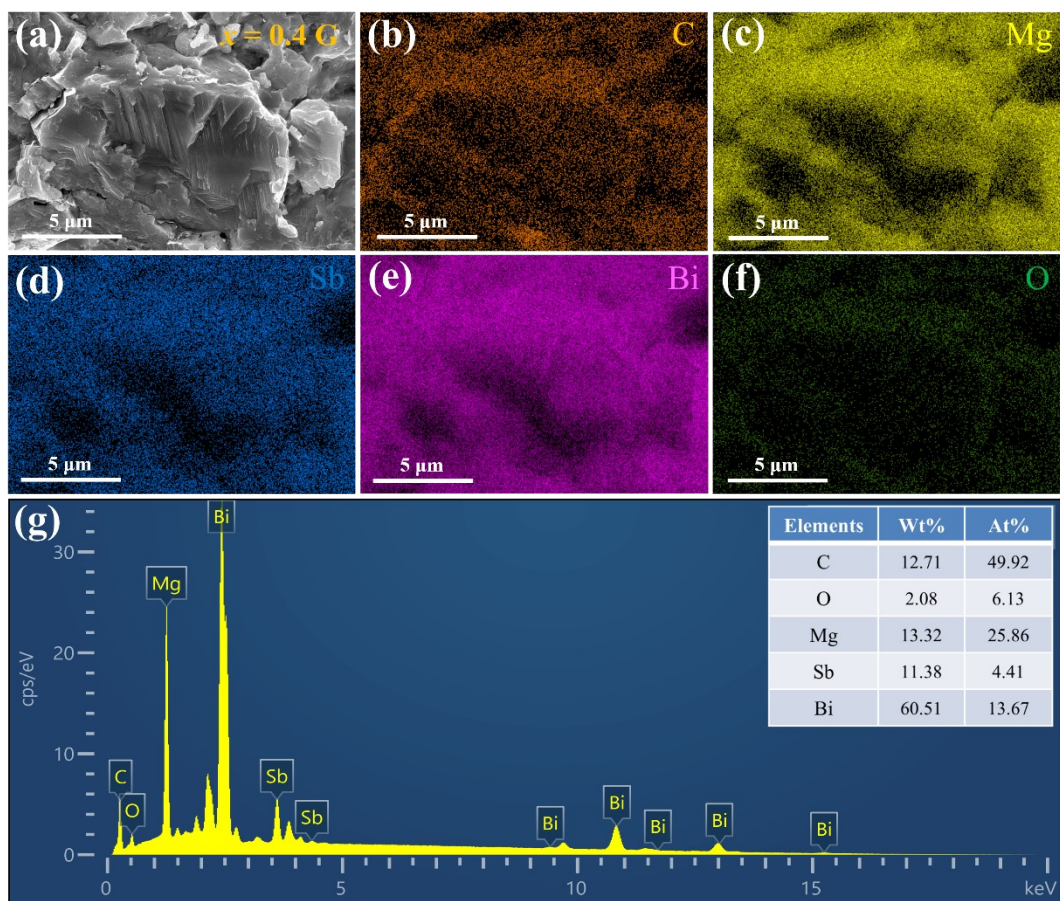
Fig. S4 (a) The cross-section SEM image of *x* = 0.4 G with (b-f) elemental maps of C, Mg, Sb, Bi and O, and (g) spectrogram of the total number of distribution maps.
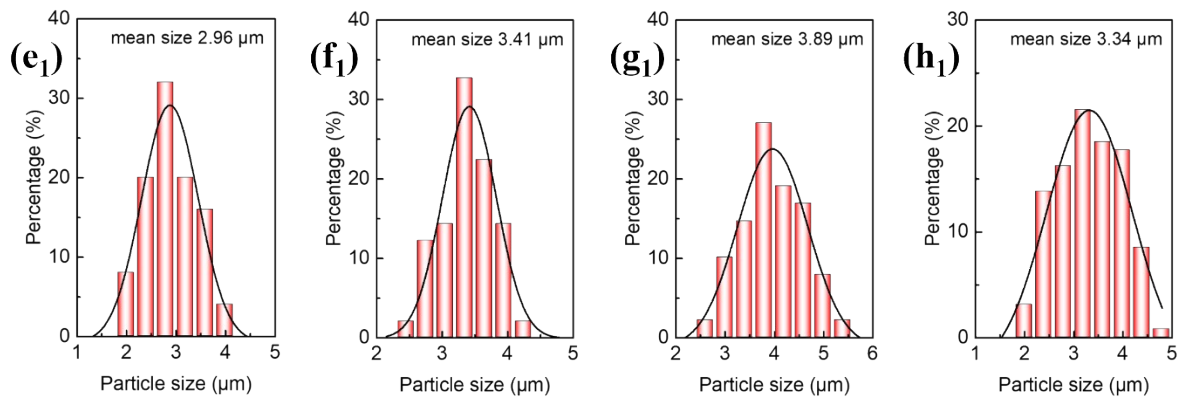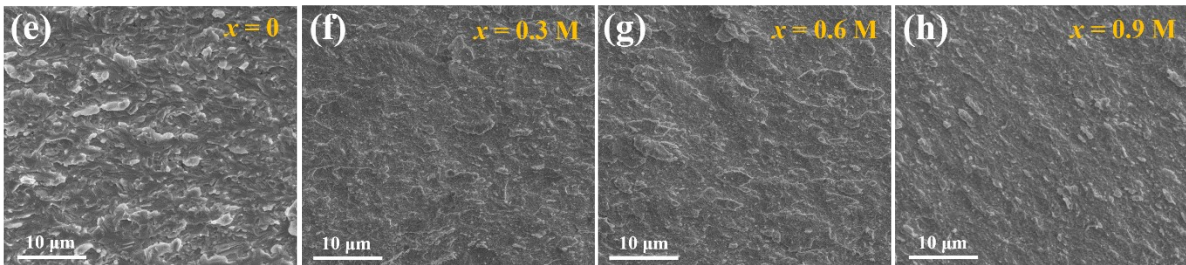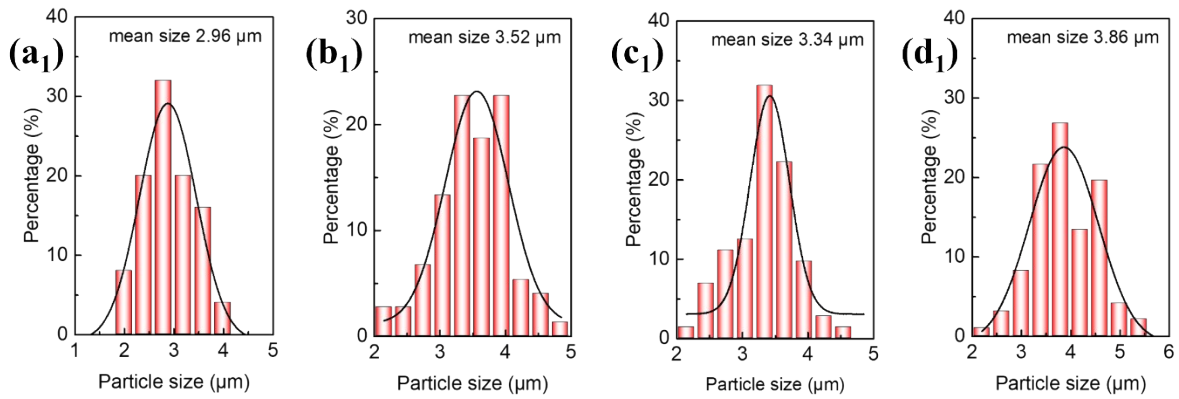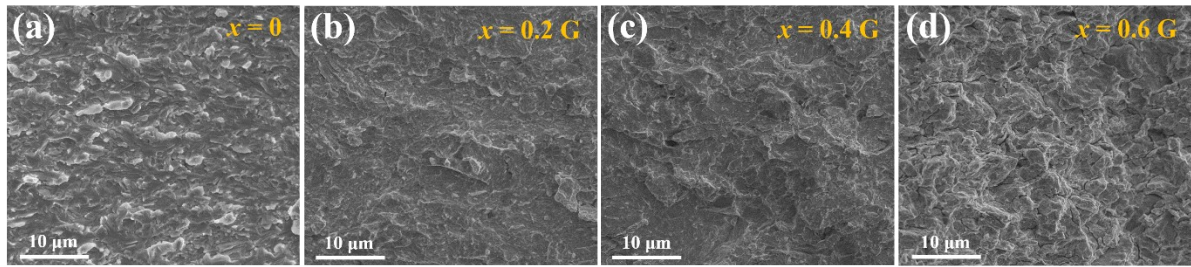
Fig. S5 (a)-(d) Cross-section SEM images of the $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% Graphene and (a₁)-(d₁) are the corresponding grain size statistics respectively. (e)-(h) Cross-section SEM images of the $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% MXene and (e₁)-(h₁) are the corresponding grain size statistics respectively.
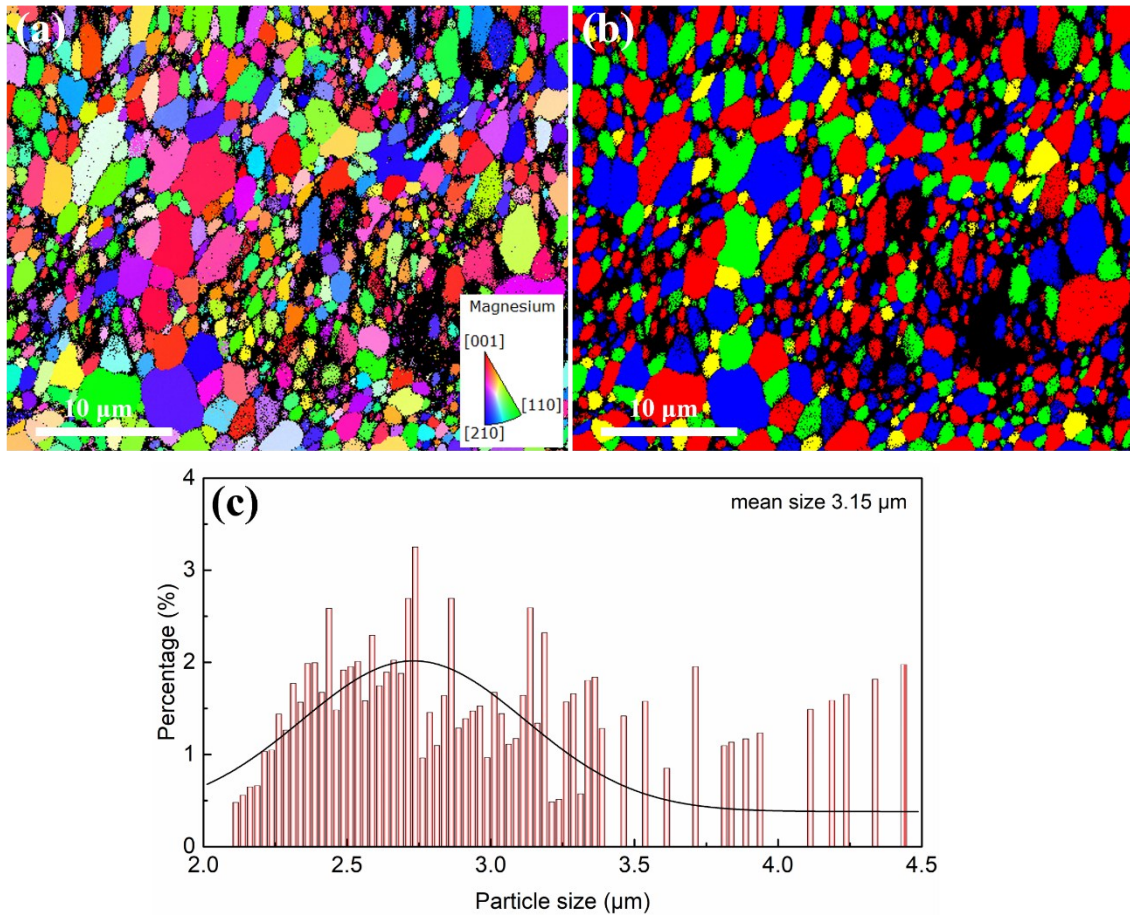
Fig. S6 (a) and (b) The electron back-scattering diffraction (EBSD) image of $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-0.6 wt% MXene sample, (c) the statistics of grain size obtained from the EBSD analyses.

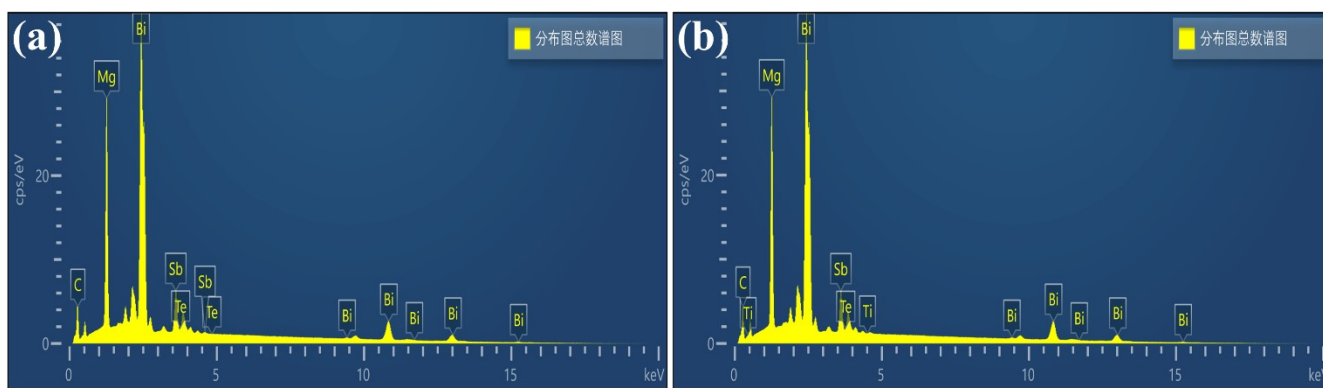**Fig. S7** Spectrogram of the total number of distribution maps in (a) $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-0.4 wt% Graphene and (b) $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-0.6 wt% MXene samples.
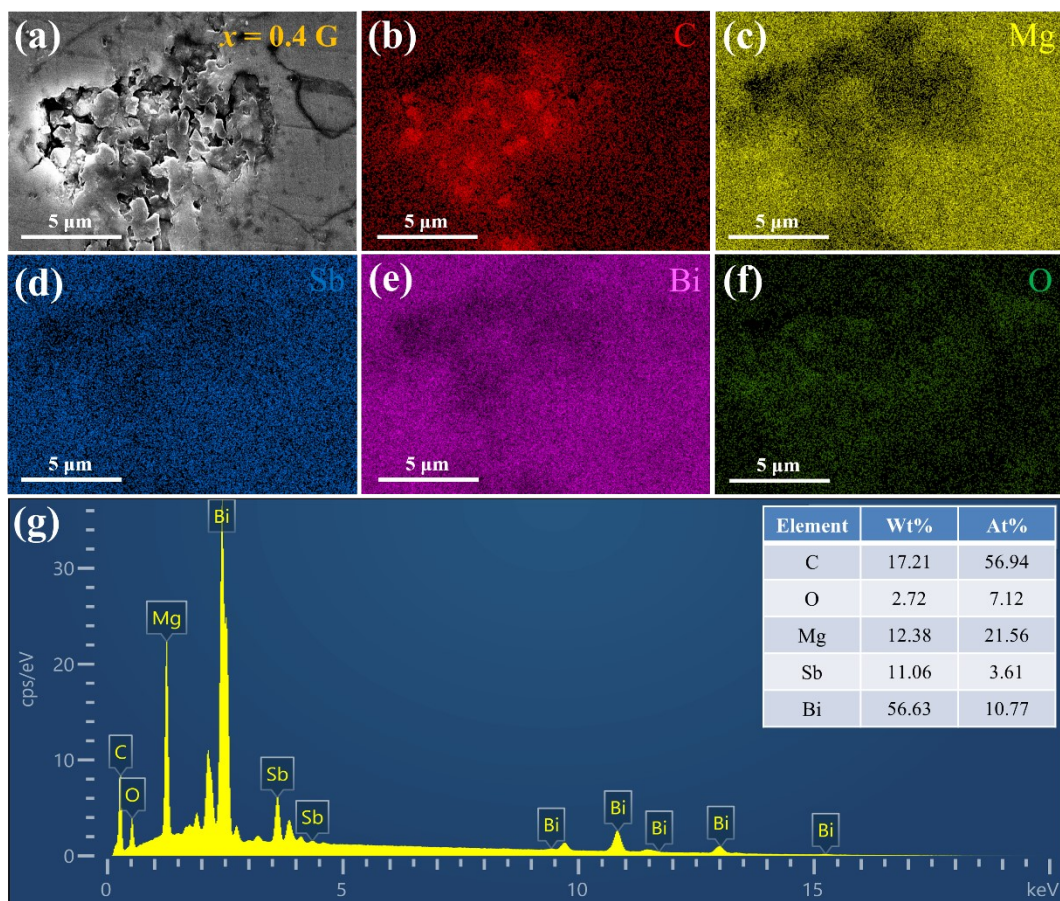
Fig. S8 (a) The polished surface SEM image of x = 0.4 G with (b-f) elemental maps of C, Mg, Sb, Bi and O, and (g) spectrogram of the total number of distribution maps.
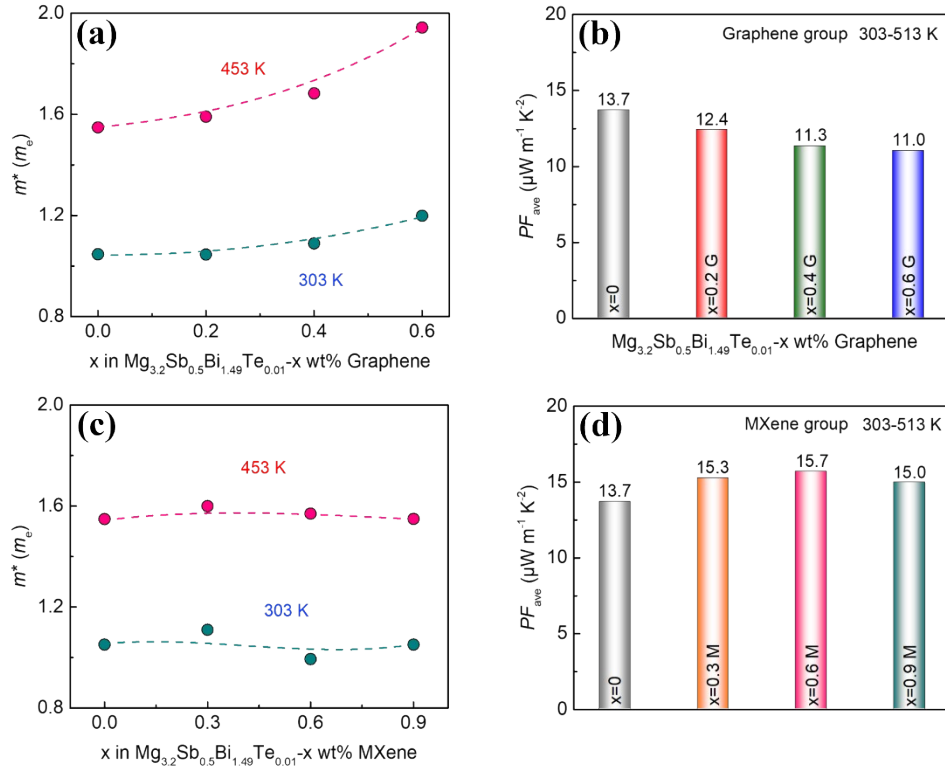
Fig. S9 (a) Density of states effective mass m^* and (b) average power factor between 303 and 513 K of $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% Graphene. (c) Density of states effective mass m^* and (d) average power factor between 303 and 513 K of $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% MXene.
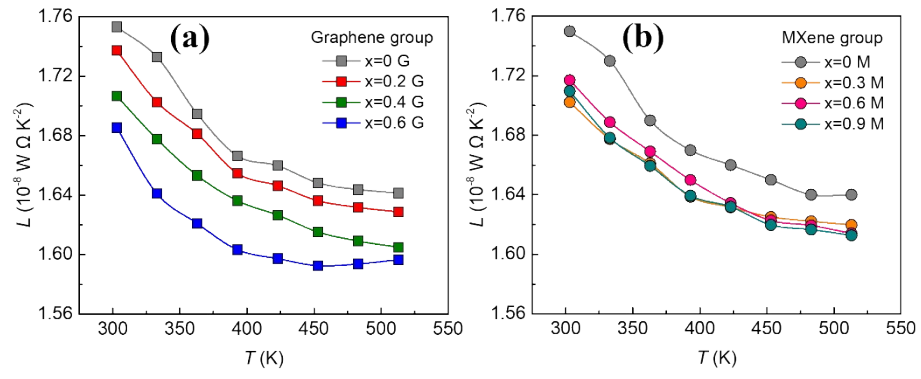
Fig. S10 Lorenz parameter of (a) $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% Graphene and (b) $Mg_{3.2}Sb_{0.5}Bi_{1.49}Te_{0.01}$-$x$ wt% MXene.

References

1. J. J. Kuo, S. D. Kang, K. Imasato, H. Tamaki, S. Ohno, T. Kanno and G. J. Snyder, *Energy Environ. Sci.*, 2018, **11**, 429-434.

2. G. J. Snyder, A. H. Snyder, M. Wood, R. Gurunathan, B. H. Snyder and C. Niu, *Adv. Mater.*, 2020, **32**, e2001537.