Electronic Supplementary Information



Figure S1: (a) surface profile of a 6 μ m Sb₂Te₃ film on Al₂O₃ s substrate, (b) surface profile of 6 μ m Bi₂Te₃ film on Al₂O₃ substrate.

S2. Measurement set up



Figure S2: Image of the system set-up used for measuring the voltage from the thermopower experiments. Ignition takes place at one end of the fuel/thermoelectric film/Al₂O₃ sample and the reaction wave self-propagates to the other end resulting in a voltage signal.

S3. Oscillation dependance on β value



Figure S3: Region in the parameter $(\beta, \ell/\beta)$ space where combustion waves and extinction occurs. Figure reproduced from reference (1S).

S4. Thermal conductivity comparison of the substrates



Figure S4: Difference in thermal conductivities of Al_2O_3 and terracotta substrates. Figure reproduced from reference (2S).

In order to show the difference in thermal conductivities of the Al₂O₃ and terracotta substrates, both were heated at one 25 end for the same duration using a custom-made blowtorch and the temperature profile was obtained at the other end to see how fast thermal conduction occurs through the substrate. In Fig. S1, we can observe that temperature for Al₂O₃ rises rapidly to about 130 °C in about 40 sec. The terracotta 30 substrate however reaches a maximum temperature of around 40 °C and takes about twice the time taken by Al₂O₃ to reach peak temperature. We note that the temperature continues to rise even after the heat source is withdrawn. This is because heat takes time to travel from one end to the other and finally 35 dissipates, depending on the thermal conductivity of the material. Therefore, the peak temperature is obtained a few seconds after the heat source is turned off. However we can see that terracotta takes approximately 60 seconds longer than Al₂O₃ to reach its peak temperature. A similar observation is 40 made in the cooling durations of the two substrates. While Al₂O₃ cools down to room temperature in about 150 seconds from the point of its peak temperature, terracotta takes approximately 240 seconds to cool down to room temperature. These observations clearly demonstrate that the thermal 45 conduction through terracotta is significantly slower than Al₂O₃. Also it further supports our experimental results, which show that the reaction velocities for Al₂O₃ substrates are considerably higher than terracotta. This endorses our approximation that the thermal conduction takes place through 50 the substrate.

S4.1 Calculation of specific power

In order to obtain the specific power, the masses of the fuel (nitrocellulose & sodium azide) and thermoelectric films were ⁵⁵ considered. The relationship between mass, density and volume was used for calculating the mass of the Sb₂Te₃ and Bi₂Te₃ films as: $m = \rho \times V$, where, *m* is the mass in grams (g), ρ is the density in g.cm⁻³ and *V* is the volume in cm³.

 ρ =6.5 g.cm⁻³ and 7.7 g.cm⁻³ for Sb₂Te₃ and Bi₂Te₃ ⁶⁰ respectively.(3S, 4S) and V depends on the dimensions of the sample and is calculated using the formula, $V = l \times w \times h$ where l is the length, w is the width and h is the thickness of the thermoelecteric film in cm, respectively.

For fuels, nitrocellulose and NaN₃ in our case, we knew the concentration of the two in their respective solutions. As

- ⁵ mentioned in *Section 2.2*, 15g.L⁻¹ nitrocellulose and 20g.L⁻¹ NaN₃ were used by drop casting. Hence, we know the amount of solution for each fuel and therefore the corresponding mass for each sample, depending on the volume of the liquid placed on the thermoelectric films. The sum of the masses of the
- ¹⁰ thermoelectric film and the fuels gives the total mass of the materials used. The corresponding power from the sample is then scaled to per kg of the materials to obtain the specific power of the sample in kW kg⁻¹.
- 15 Note:

The mass of Al₂O₃ has not been considered as its effect was negligible compared to the masses of Sb₂Te₃/Bi₂Te₃ and the fuels. The entire thickness of 100 μ m does not contribute to the thermal conduction. As can be seen from Fig. S4, it takes

- $_{20}$ 25 sec for the heat to be thermally conduced through 100 µm of Al₂O₃ from one side to another. Taking the short thermopower reaction time [Fig. 3(b)] of approximately 0.5 sec as an example (most of them are shorter than this), we can therefore estimate that only a 2 µm thick Al₂O₃ substrate will
- suffice for the purpose of the thermal conduction in these experiments. Even assuming an extreme effective thickness of 5 times larger than 2 μ m (10 μ m) for Al₂O₃ thermal conductivity, the effective volume is calculated to be 1 cm × 0.2 cm × 10⁻³ cm =2 × 10⁻⁴ cm³. Considering the density of
- $_{30}$ Al₂O₃ to be 3.6 g. cm⁻³,(5S) the total effective mass of Al₂O₃ substrate is only 0.7 mg, which is negligible (the total fuel/thermoelectric film mass ranged between 16 to 29 mg).

35 S5. Comparison of heat conduction rate of Sb₂Te₃ and Bi₂Te₃



Figure S5: a) Measurement set up, b) Difference in thermal conduction of Sb_2Te_3 and Bi_2Te_3 films

The thermal diffusivity (α) is defined as follows:

$$\alpha = \frac{\kappa}{\frac{\rho c}{p}}$$
(S1)

where, κ is the thermal conductivity, ρ is the density and c_P is the specific heat. The density of Sb₂Te₃ and Bi₂Te₃ is approximately 6.5 g/cm³ and 7.7 g/cm³, respectively.(*3S,4S*) The specific heats of Sb₂Te₃ and Bi₂Te₃ are 0.19 J/g.K and ⁴⁵ 0.16 J/g.K respectively.(*6S,7S*) Therefore, the product ρc_P in the denominator is 1.235 and 1.232 for Sb₂Te₃ and Bi₂Te₃ respectively, which are almost equal. As a result it can be said that the thermal diffusivity (α) is directly proportional to the thermal conductivity κ in the case of our experiments.

Figure S5 (a) shows the measurement set up. The Sb_2Te_3/Al_2O_3 and Bi_2Te_3/Al_2O_3 samples of ~ 12 mm length were used for the measurement and the thickness of both Sb₂Te₃ and Bi₂Te₃ films were equal to 6 µm. A heater with a 55 sharp tip was used as the heating source and was positioned at one end on the sample. The temperature profile was acquired at the opposite end using a TENMA 727730 thermocouple. The heat source was kept in place until the temperature at the other end reached a constant value (approximately 200 s) in 60 equilibrium with the ambient. Figure S5(b) shows the temperature profile for the two samples. It shows that the temperature rises to approximately 47°C for both samples. However, the rate of rise is approximately 2.2 times higher for the Al₂O₃/Sb₂Te₃ sample which is analogous to the ratio of the 65 thermal conductivites of Sb₂Te₃ and Bi₂Te₃. This result endorses our explanation that the rate of heat conduction in Sb₂Te₃ is \sim 2.5 times faster than that in Bi₂Te₃; based on the ratio of their thermal conductivity.

Heat conduction is defined as
$$H = \frac{\Delta Q}{\Delta t} = \kappa A \frac{\Delta T}{x}$$

⁷⁰ where $\frac{\Delta Q}{\Delta t}$ is the rate of heat flow, κ is the thermal

conductivity, A is the total cross sectional area of the conducting surface, ΔT is the temperature difference and x is the thickness of the conducting surface separating the two temperatures. As the dimensions, thickness, and temperature ⁷⁵ difference of our Sb₂Te₃ and Bi₂Te₃ samples are similar the thermal conductivity is the most dominant factor affecting the rate of heat conduction. Hence, we believe that our assumption in the paper is consistent with the theory. Also experimental results of Figure S5 shown in the supplementary ⁸⁰ prove the same."

S6. Comparison of electrical conductivities of Sb_2Te_3 and Bi_2Te_3

⁸⁵ In order to compare the electrical conductivities of the two films four point probe measurements were carried out on Sb₂Te₃ and Bi₂Te₃. The following equation was then used to calculate the resistivity:

$$\rho = R_{Sh} \times t \quad ; \tag{S2}$$

in which ρ is the resistivity of the film in Ω .m, R_{sh} is the sheet resistance of the film in Ω / and *t* is the thickness of the film in m.

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5 Sb₂Te₃ film

The four point probe showed that the sheet resistance of the Sb₂Te₃ film was 3.103 Ω / for the Sb₂Te₃ film of approximately 6µm. Therefore the resistivity of the film was calculated to be $1.86 \times 10^{-5} \Omega$.m. Inverse of the resistivity ¹⁰ gives the conductivity.

Thus, the electrical conductivity of the Sb_2Te_3 film was found to be 5.3×10^4 S/m.

Bi₂Te₃ film

 $_{15}$ A similar approach was used to calculate the electrical conductivity of the Bi_2Te_3 film. The sheet resistance of the Bi_2Te_3 film was found to be 1.75 $\Omega/$.

Therefore the electrical conductivity of the film was calculated to be 1.1×10^5 S/m.

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The above measurements show that the electrical conductivity of Bi_2Te_3 is an order of magnitude higher than Sb_2Te_3 . The values obtained are also comparable with the literature values of 10^4 and 10^5 S/m for Sb_2Te_3 and Bi_2Te_3 films

25 respectively.(8S-10S)

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