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Walia, S., Weber, R., Sriram, S., Bhaskaran, M., Latham, K., Zhuiykov, S., & Kalantar Zadeh, K. (2011). Sb₂Te₃ and Bi₂Te₃ based thermopower wave sources. *Energy & Environmental Science*, 4(9), 3558–3564. <https://doi.org/10.1039/c1ee01370j>

Document Version: Accepted Manuscript

Published Version: <https://doi.org/10.1039/c1ee01370j>

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Citation:

Walia, S, Weber, R, Sriram, S, Bhaskaran, M, Latham, K, Zhuiykov, S and Kalantar Zadeh, K 2011, 'Sb₂Te₃ and Bi₂Te₃ based thermopower wave sources', *Energy & Environmental Science*, vol. 4, no. 9, pp. 3558-3564.

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Version: Accepted Manuscript

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<http://dx.doi.org/10.1039/c1ee01370j>

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Sb₂Te₃ and Bi₂Te₃ based thermopower wave sources†

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Received (in XXX, XXX) Xth XXXXXXXXXX 200X, Accepted Xth XXXXXXXXXX 200X

First published on the web Xth XXXXXXXXXX 200X

DOI: 10.1039/b000000x

Exothermic chemical reactions from nitrocellulose are coupled onto Sb₂Te₃ (antimony telluride) and Bi₂Te₃ (bismuth telluride) layers to generate self propagating oscillating thermopower waves. P-type Sb₂Te₃ and N-type Bi₂Te₃ are employed due to their large Seebeck coefficients, high electrical conductivities and their complementary semiconducting properties. Sources based on both materials exhibit high power to mass ratios: up to 0.6 kW kg⁻¹ for Sb₂Te₃ and 1.0 kW kg⁻¹ for Bi₂Te₃. Having both P- and N-type semiconductors in the system, the combination of the outputs can be used for generating sources with polarities alternating in time.

1 Introduction

The miniaturization of power sources, while maintaining a high specific power (also called power-to-weight ratio), is necessary for applications in advanced micro/nanoscale intelligent electronics, telecommunications systems, as well as implantable bio-devices.⁽¹⁻³⁾ Integrated miniaturised microsystems such as micro-electro-mechanical systems (MEMS) will struggle to attain their potential until power sources driving these systems match their dimensional scale and deliver the required power density.⁽⁴⁾ Miniaturization is also desirable for ignition systems of heat engines, where a large power surge is required solely at the start, and not over the course of operation of the engine; thereby, eliminating the need for accommodating bulky and heavy batteries for the entire journey.⁽⁵⁾ It is known that the deployment of hybrid cars with ultimate efficiency has been held back by the lack of suitable sized and high specific power batteries, with small volume and weight.⁽⁶⁾ Although there have been recent advances in fabricating 3-D Li-ion micro/nano batteries, their specific power and energy densities are still low (~0.3 kW kg⁻¹ and 0.4 kWh kg⁻¹, respectively).⁽⁷⁻⁸⁾ A reduction in size gives rise to problems such as ionic flow disruption around electrodes as well as electrical shorts that hamper their ability to perform effectively.^(4, 9-10) Additionally, microlithography processes have proved cumbersome and expensive to transfer to batteries.⁽¹¹⁾

Energy can also be released *via* systems such as fuel cells; however, they generally require large volumes and expensive electronics while exhibiting low specific powers.⁽¹²⁻¹⁴⁾ Additionally, conventional batteries and fuel cells are only capable of producing direct current (DC) power, while oscillatory, alternating current (AC) sources are desirable for most electrical applications.

Energy can also be stored and released at high rates using technologies such as supercapacitors. They exhibit a specific power of at least 10 kW kg⁻¹.⁽¹⁵⁻¹⁷⁾ The major disadvantage of supercapacitors is their high self-discharge rate. This renders them impractical for long-term energy storage as they have to be charged frequently in order to maintain the stored energy. Thus, none of the aforementioned technologies have

so far enabled reliable miniaturized power sources with high specific power.

Choi *et al.*⁽¹⁸⁻¹⁹⁾ have recently demonstrated that chemically driven ‘thermopower waves’ guided by multi-walled carbon nanotubes (MWNT) are capable of producing specific powers as large as 7 kW kg⁻¹. In such thermopower devices the highly conductive MWNT core allows the axial transportation of free electrons, that are generated *via* a large temperature gradient across the device. This temperature gradient is produced as a result of an exothermic reaction of a highly reactive solid fuel covering the MWNTs. In these systems, the high thermal conductivity of MWNTs guarantees the continuation of the reaction and the propagation of thermopower waves. The main limitation of the work was the low voltage generated, which averaged 30-50 mV with a maximum reported voltage of 210 mV for masses in the range of just a few milligrams. The amplitude of oscillations was also low (20-30 mV generally). The reason for this limitation is that MWNTs exhibit a relatively low Seebeck coefficient (80 μ V K⁻¹).⁽²⁰⁾ In order to increase the output voltage, thermoelectric materials such as antimony telluride (Sb₂Te₃) and bismuth telluride (Bi₂Te₃) with large respective Seebeck coefficients of 287 and 243 μ V K⁻¹ can potentially replace MWNTs.⁽²¹⁻²⁴⁾ One of the main requirements of a thermopower source core material is its high electrical conductivity. A higher electrical conductivity can increase the power output as it results in larger currents.⁽²⁵⁾ This means that materials such as silicon (Si), even in its highly-doped state, with a large Seebeck coefficient (~1500 μ V K⁻¹) are rendered inefficient due to their low electrical conductivity.⁽²⁶⁾ Sb₂Te₃ and Bi₂Te₃, on the other hand, both have high electrical conductivities. The electrical conductivity of Bi₂Te₃ is of the order of 10⁵ S m⁻¹, while Sb₂Te₃ exhibits an electrical conductivity in the range of 0.5-1.0×10⁴ S m⁻¹ [See ESI† for details].⁽²⁷⁻²⁹⁾

High thermal conductivity is also required for sustaining the propagation of the thermopower waves. The thermal conductivity of Sb₂Te₃ and Bi₂Te₃ is low (2.5 and 1.0 Wm⁻¹ K⁻¹, respectively).⁽³⁰⁻³²⁾ To compensate for this and to achieve the desired effective thermal conductivity, we can place the materials on highly thermally conductive

substrates (for instance Al_2O_3 with thermal conductivity of $20 \text{ W m}^{-1} \text{ K}^{-1}$).⁽³³⁾

We have recently demonstrated thermopower wave generation by coupling exothermic chemical reaction of nitrocellulose to Bi_2Te_3 films supported by either a highly thermally conductive alumina (Al_2O_3) substrate in comparison to a much less thermally conductive terracotta (baked natural polysilicate) substrate.⁽³⁴⁾ These devices produced a high specific power of the order of 1 kW kg^{-1} and generated voltages of the order of 100-150 mV with large oscillation amplitudes in the range of 50-140 mV. The generated voltage and the amplitude of oscillations for Bi_2Te_3 -based devices were significantly larger than MWNT-based thermopower devices.

In this work, we use Sb_2Te_3 as the complementary thermoelectric material and compare its performance to previously demonstrated Bi_2Te_3 -based thermopower devices.⁽³⁴⁾ Sb_2Te_3 is a P-type semiconductor and Bi_2Te_3 is N-type.⁽³⁵⁻³⁶⁾ Such a contrast ensures that the voltage output from these thermopower wave sources, with identical characteristics (one based on Bi_2Te_3 and the other based on Sb_2Te_3), produce both positive and negative polarities. The combination of these two can result in an alternating signal, which is required for practical applications. Similar to our previous work ⁽³⁴⁾, we use Al_2O_3 and terracotta as the substrates in our investigations. They are chosen due to their contrasting thermal conductivities, which will help observing the effect of thermal properties of the system on the thermopower wave propagation. A detailed comparison of Sb_2Te_3 - and Bi_2Te_3 -based thermopower wave devices is presented in this work.

2 Experimental

2.1 Deposition of Sb_2Te_3 and Bi_2Te_3 films

The Sb_2Te_3 and Bi_2Te_3 films were deposited on Al_2O_3 and terracotta substrates using RF magnetron sputtering, under identical deposition conditions. High purity (99%) Sb_2Te_3 and Bi_2Te_3 targets (Vin Karola Instruments) were used for sputtering. The chamber was pumped down to a pressure of 2×10^{-5} Torr before starting the sputtering process. Al_2O_3 and terracotta substrates with linear dimensions of $12\text{-}15 \text{ mm (L)} \times 4\text{-}6 \text{ mm (W)} \times 100 \mu\text{m (H)}$ were utilised after being cleaned using acetone, isopropyl alcohol, and distilled water. The substrates were held at a constant temperature of 100°C throughout the sputtering process, in which a power of 100 W and an argon atmosphere with 10 mTorr process pressure were used. Sputtering time of 90 mins resulted in a $6 \mu\text{m}$ thick Sb_2Te_3 and $5 \mu\text{m}$ Bi_2Te_3 films, respectively. Electrical contacts between the copper tape electrodes and the thermoelectric layer were made using adhesive conductive silver paste (42469, Alfa Aesar). The resistance between the electrodes for Sb_2Te_3 films and Bi_2Te_3 films ranged from $6\text{-}12 \Omega$ and $2\text{-}10 \Omega$, respectively.⁽³⁴⁾

Fig 1 The SEM images of: **a)** Sb_2Te_3 film on Al_2O_3 **b)** Bi_2Te_3 film on Al_2O_3 . **c)** 1. The XRD pattern of Sb_2Te_3 before thermopower wave propagation, 2. after adding nitrocellulose and 3. after thermopower wave propagation **d)** 1. The XRD pattern of Bi_2Te_3 before thermopower wave propagation, 2. after adding nitrocellulose and 3. after thermopower wave propagation.

2.2 Preparation of fuel

The fuel used for the exothermic reaction was a combination of nitrocellulose ($\text{C}_6\text{H}_8(\text{NO}_2)_2\text{O}_5$) and sodium azide (NaN_3).

Nitrocellulose was used due to its large enthalpy of reaction ($4.75 \times 10^6 \text{ J kg}^{-1}$). It was prepared by dissolving millipore nitrocellulose membranes (N8645, Sigma Aldrich) in acetonitrile (15g/L). This solution was then dropcast on the Bi_2Te_3 and Sb_2Te_3 films using a pipette and left to dry. The acetonitrile evaporated leaving an adhesive and solid nitrocellulose layer deposited on top of the thermoelectric materials. NaN_3 was used to serve as a primary igniter due to its low activation energy (40 kJ mol^{-1} for NaN_3 , compared to $110\text{--}150 \text{ kJ mol}^{-1}$ for nitrocellulose). NaN_3 (14314, Alfa Aesar) in aqueous solution (50 mg mL^{-1}) was then added on top of the nitrocellulose layer using a pipette. The total thickness of the fuel layer was $\sim 240 \mu\text{m}$.

2.3 Characterization of films

Figures 1(a,b) show scanning electron micrographs (SEM) of Sb_2Te_3 and Bi_2Te_3 films, respectively. The images clearly show the difference between the structures of the two films with Bi_2Te_3 exhibiting a more irregular structure compared to Sb_2Te_3 . This confirms the polycrystalline nature of Bi_2Te_3 . The SEM images also show that Bi_2Te_3 films are more porous than Sb_2Te_3 , when deposited on Al_2O_3 substrates under identical conditions.

A surface profilometer (Ambios Technology XP-2) was used to further investigate the morphology of the Sb_2Te_3 and Bi_2Te_3 surfaces and that of the substrate [see ESI†, Figs. S1(a,b)]. The average surface roughness of Sb_2Te_3 and Bi_2Te_3 was determined to be 0.4 and $0.7 \mu\text{m}$, respectively. This indicates that the Bi_2Te_3 films are more porous; thereby, confirming the earlier observation made from the micrograph in Fig. 1(b). A greater porosity enables more fuel to be placed on the surface of the films. Additionally, a higher porosity is also important to facilitate increased thermal conduction between the fuel and thermoelectric films, resulting in a more sustained reaction propagation.

X-ray diffraction (XRD) measurements were carried out on the Sb_2Te_3 and Bi_2Te_3 samples in order to assess the crystallinity of the films. The XRD was conducted using a Bruker D8 DISCOVER microdiffractometer fitted with a GADDS (General Area Detector Diffraction System). The XRD pattern for Sb_2Te_3 [Fig. 1(c), 1] shows three well defined peaks, which indicate its crystalline structure. In case of Bi_2Te_3 , the XRD pattern reveals a more polycrystalline structure. On addition of nitrocellulose different effects on the XRD patterns of Sb_2Te_3 and Bi_2Te_3 are observed [Fig. 1(c), 2 and Fig. 1(d), 2]. For Bi_2Te_3 the peak at $18.1^\circ 2\theta$ (006) plane for rhombohedral Bi_2Te_3 – ICDD No. [1-75-0921] is no longer exhibited in the samples with nitrocellulose. Possibly incoherent scattering and adsorption effects may be responsible for the disappearance of this peak. It is also probable that the nitrocellulose coating on both the Sb_2Te_3 and Bi_2Te_3 samples effectively masks the surface, thus inhibiting the penetration of X-rays to, and collection of scattering data from this

Fig 2 Schematic of the fuel/thermoelectric material/substrate thermopower wave generation system. Ignition at one end of the fuel (combination of nitrocellulose and sodium azide) layer results in a self propagating exothermic reaction. (Not to scale.)

particular peak. This effect is particularly acute for the relatively low intensity (006) Bi_2Te_3 peak, which lies in the same region as the principal XRD reflections for nitrocellulose (ICDD No. [03-0114]). Sb_2Te_3 on the other hand does not show any significant diffraction peaks in this region. Thus, the masking effect caused by the nitrocellulose layer only results in a general decrease in diffraction intensity across its entire diffraction pattern. The XRD patterns of the samples following consumption of the nitrocellulose are also shown [Fig. 1(c), 3 and Fig. 1(d), 3], and will be discussed later.

3 Results and Discussion

Figure 2 shows the schematic of the set-up [also see ESI†, Fig. S2 for the actual apparatus] used for the thermopower experiments. Ignition takes place at one end of this system and the reaction self propagates to the other end. A custom-made blow torch with a localized flame tip is used to initiate the reaction. Samples with low mass of nitrocellulose or NaN_3 (masses below 15 mg per 75 mm^2) showed no sustained wavefront propagation in any direction. Several samples using different mass of the fuels were prepared and tested. For both Bi_2Te_3 and Sb_2Te_3 systems identical masses were used to ensure statistical comparison. Ignition at one end of the system, shown in Fig. 2, results in an accelerated self propagating reaction wave that drives a simultaneous wave of free carriers. This wave of carriers results in a flow of current and voltage across the devices. The voltage signal exhibited a positive polarity for waves initiating at the positive electrode for P-type Sb_2Te_3 and the opposite polarity for the N-type Bi_2Te_3 . The duration of the output signal corresponds to the reaction time, thus enabling us to calculate the reaction propagation velocity. Even in oscillatory mode, the thermopower waves always exhibit a constant polarity during the reaction, depending on the direction of wavefront propagation. This suggests that the wave passes through the system faster than the cooling time of its posterior region.

Fig 3 Comparison of oscillations between Sb_2Te_3 and Bi_2Te_3 based thermopower devices. **a)** Oscillatory thermopower voltage signal for Sb_2Te_3 device using Al_2O_3 substrate. **b)** Oscillatory thermopower voltage signal for Bi_2Te_3 device using Al_2O_3 substrate. We can see that Bi_2Te_3 based devices exhibit amplitudes approximately 3 times larger than that of Sb_2Te_3 based devices. The difference is similar to the variation in the thermal conductivities of Bi_2Te_3 and Sb_2Te_3 .

3.1 Comparison of Sb_2Te_3 - and Bi_2Te_3 -based thermopower devices

Thermopower devices based on Al_2O_3 substrate

The thermal conductivity of Al_2O_3 is much higher than Bi_2Te_3 and Sb_2Te_3 . Both Bi_2Te_3 and Sb_2Te_3 films are comparatively thin ($<10 \mu\text{m}$ compared to the $\sim 100 \mu\text{m}$ thick Al_2O_3 substrate – see ESI† for further information). Thus we can assume that the Al_2O_3 substrate will dominate the thermal conduction process.(34)

Figures 3(a,b) show typical voltage measurements across the fuel/ Sb_2Te_3 / Al_2O_3 and the fuel/ Bi_2Te_3 / Al_2O_3 systems, respectively. The voltage profiles of Figs 3(a,b) can be divided into an initial reaction phase followed by a cooling down phase. The reaction phase consists of a rising voltage and continues until all the combustion fuel is consumed. This is followed by a region of exponential decay.(40) The voltage generated depends on the sample mass, especially the amount of fuel used (which is a combination of nitrocellulose and sodium azide). Sb_2Te_3 -based devices generated voltages as large as 200 mV with oscillations in the range of 10-40 mV. On the other hand, Bi_2Te_3 devices generated voltages as large as 140 mV with larger oscillations generally within the range of 40-140 mV. The Bi_2Te_3 -based devices occasionally showed voltages as high as 300-400 mV; however, due to lack of consistency these results are not included. Figures 3(a,b) also show the difference in the oscillation amplitudes of the Sb_2Te_3 and Bi_2Te_3 devices. The reason for the difference in oscillatory behaviour can be attributed to the larger thermal conductivity of Sb_2Te_3 (2.5 times that of Bi_2Te_3). Oscillation amplitude difference between the two devices corresponds to the difference in thermal conductivities of the two materials. A larger effective thermal conductivity means that the reaction will propagate faster potentially affecting the oscillation amplitude. Previously, we have theoretically demonstrated that a change in thermal conductivity affects the rate of heat loss and hence the oscillations.(34) For simplification in our theoretical analysis, we started with a one-dimensional space model and transformed it to a non-dimensional system to predict the occurrence of oscillatory combustion linked to the heat losses. As a first step towards analysing the combustion, we applied the model presented by Mercer *et al.*(41) Assuming that the solid fuel combustion is exothermic, oxygen is supplied in excess, and governed by

Arrhenius kinetics with volumetric heat loss and defining the non-dimensional temperature u to be:

$$u = \frac{RT}{E} \quad (1)$$

The non-dimensional equations for the conservation of heat and mass are:

$$\frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial x^2} + we^{-1/u} - \ell(u - u_a) \quad (2)$$

$$\frac{\partial w}{\partial t} = -\beta we^{-1/u} \quad (3)$$

in which T and w denote the temperature and concentration of the fuel, respectively, and x and t are the defined space and

time variables. We define as $\beta = \frac{Ec_p}{QR}$, a parameter related to

the properties of the fuel, and the non-dimensional volumetric

heat transfer, $\ell = \frac{S}{V} \frac{hE}{\rho RQA}$ where, $\frac{S}{V}$ is the surface-area-to-

volume ratio of the fuel (m^{-1}) and h is the heat transfer coefficient from the fuel to the quiescent surroundings ($\text{J s}^{-1} \text{m}^{-2} \text{K}^{-1}$), ρ is the density of the fuel (kg m^{-3}), c_p is the specific heat of the fuel ($\text{J kg}^{-1} \text{K}^{-1}$), Q is the heat of reaction (J kg^{-1}), A is the pre-exponential rate constant (s^{-1}), E is the activation energy (J mol^{-1}), R is the universal gas constant ($8.314 \text{ J mol}^{-1} \text{K}^{-1}$). The heat transfer coefficient h is an estimate for all the losses in the averaged model that we have used. For the purpose of gaining an understanding of the process, we estimated the heat losses to the substrate and the environment and lumped them together in h . Heat is transferred from the fuel layer to the (Bi_2Te_3 or Sb_2Te_3) thermoelectric/substrate layer and to the surroundings. Depending on the value of system parameters, we can predict the oscillatory behaviour of the thermopower waves. The determination of all the necessary parameter values is the key to being able to apply the model to a real system. The solution of the model presented by Mercer *et al.*, (41) assumes that the ambient temperature is absolute zero ($u_a=0$). Ambient temperatures are very small relative to the reaction temperature, and hence, this inference has little effect on the overall behaviour of the solutions to this model. For the fuel, the value of the parameters is as follows: $S/V=1/240 \mu\text{m}^{-1}$, $\rho = 1600 \text{ kg.m}^{-3}$, $c_p = 1596 \text{ J.kg}^{-1}.\text{K}^{-1}$, $Q = 4.75 \times 10^6 \text{ J.kg}^{-1}$, $E = 1.26 \times 10^5 \text{ J.mol}^{-1}$, $A = 10^5 \text{ s}^{-1}$ and $h=2 \times 10^{-3} \text{ J.s}^{-1}.\text{m}^{-2}.\text{K}^{-1}$. In all cases the β value is 5.09 for the nitrocellulose fuel layer. The oscillation period is directly related to β and ℓ . For $\beta = 5.09$, the critical heat loss value for the onset of oscillations can be estimated as being smaller than $\ell/\beta = 0.000035$ (using Fig. S3, see ESI†). The change in thermal conductivity affects the heat loss parameter. The higher thermal conductivity of Sb_2Te_3

Fig 4 a) Comparison of voltage generated as a function of mass (of fuel and thermoelectric material) for Sb_2Te_3 and Bi_2Te_3 based thermopower devices. **b)** Power as a function of mass. **c)** Comparison of the reaction propagation velocity for Sb_2Te_3 and Bi_2Te_3 based devices.

compared to Bi_2Te_3 will enhance heat transfer (see ESI†, Section S5). Hence, we would predict faster combustion velocities for Sb_2Te_3 -based devices and that the system would be closer to the parameter values where oscillations in the combustion occur. However, as the model is based on a one-dimensional average over all the layers, the resultant changes predicted are not as dramatic as observed in the experiments. Abrahamson et al.(40, 42) have employed a similar averaged one-dimensional model. They have solved their model numerically (rather than use the asymptotic results

that we have) with a set of estimated parameter values and found corresponding results to those reported in this work, albeit for carbon nanotube guided thermopower waves, which are on a smaller scale than our experiments.

The analysis of voltage and power as a function of mass shows an optimal mass range for both Sb_2Te_3 - and Bi_2Te_3 -based devices. A very low amount of fuel does not generate enough heat to sustain the propagation of the exothermic reaction. Too much fuel, on the other hand may provide more heat energy, but the unreacted accumulated fuel mass renders the reaction unsustainable.(34) As a result, we observed an optimal mass range (22-26 mg for $\sim 75 \text{ mm}^2$ samples) that sustained the wavefront propagation (see ESI†, Section S4.1 for detailed information on mass of substrates and the films). Almost both type devices exhibited well-matched trends in terms of the relation between voltage and mass [Fig. 4(a)].

Comparison of peak power vs. mass for the two different materials is shown in Fig. 4(b). The peak power was

determined using the equation: $P = \frac{V^2}{R}$ in which P is the peak power in watts, V is the peak output voltage in volts and R the resistance in Ω .

The resistance of the Sb_2Te_3 films ranged from 6-12 Ω , while Bi_2Te_3 films resistances were between 2-10 Ω . The difference between the resistances can be attributed to the larger electrical conductivity of Bi_2Te_3 (see ESI†, Section S6). Hence, we expect the power output from Bi_2Te_3 -based devices to be higher. Experimental observations show that the peak power generated by Sb_2Te_3 -based devices is $\sim 6 \text{ mW}$, while Bi_2Te_3 -based devices on an average generate 67% larger power with the peak value of $\sim 10 \text{ mW}$. The peak specific powers of the Sb_2Te_3 and Bi_2Te_3 devices were 0.6 kW kg^{-1} and 1.0 kW kg^{-1} , respectively. This difference can be mainly attributed to the higher electrical conductivity of Bi_2Te_3 . The resistance of the thermoelectric films remains almost the same even after the end of the reactions. The resistance dependence on temperature is defined by the following equation:

$$R(T) = R_0[1 + \alpha(T - T_0)] \quad (4)$$

where T is the temperature, T_0 is the reference temperature, R_0 is the resistance at T_0 and α is the temperature coefficient of resistivity of the material. Since, Sb_2Te_3 and Bi_2Te_3 are semiconductors they exhibit negative temperature coefficient of resistivity. (43-44) Hence, it is expected that the resistance actually decreases during the reaction propagation in our case. Additionally, the XRD pattern of the samples after the thermopower wave propagation, confirm that the structures of Sb_2Te_3 and Bi_2Te_3 remain unchanged [Fig. 1(c), 3 and Fig. 1(d), 3], as the peaks remain dominantly unaltered. The average wavefront propagation velocity for Sb_2Te_3 -based devices was approximately 0.7 m/s compared to $\sim 0.3 \text{ m/s}$ average propagation velocity for Bi_2Te_3 -based devices. Propagation velocities for Sb_2Te_3 -based devices were generally 2-2.5 times higher than the velocity for Bi_2Te_3 -based devices [Fig. 4(c)], which nicely corresponds to the difference

Fig 5 Comparison of thermopower voltage signals on terracotta substrates for: **a)** Sb_2Te_3 films, **b)** Bi_2Te_3 films.

in the thermal conductivities of the two thermoelectric materials. A higher thermal conductivity of the thermoelectric material provides an extra path for heat conduction (*i.e.* the surface of the Sb_2Te_3 film) in addition to the path provided by the thermally conductive Al_2O_3 substrate. This will consequently cause the wave to travel faster on Sb_2Te_3 , as the thermal wave generated from the exothermic reaction travels approximately 2.5 times faster than on Bi_2Te_3 (see ESI† Section S5 for details).

Thermopower devices based on terracotta substrate

The oscillatory behaviour and wave front velocities strongly depend on the thermal conductivity of the substrate. Terracotta has an identical thermal conductivity to Bi_2Te_3 . However, thermal conductivity of Sb_2Te_3 is 2.5 times higher. Consequently, we expect the rate of heat conduction for Sb_2Te_3 -based devices to be approximately 2.5 times faster than Bi_2Te_3 -based thermopower devices (see ESI† Section S5).

Figures 5(a,b) show the voltage signals obtained for Sb_2Te_3 /terracotta and Bi_2Te_3 /terracotta systems, respectively. In line with our expectation, we can see that the reaction propagation for the Sb_2Te_3 /terracotta device is ~ 3 times faster than the Bi_2Te_3 /terracotta device. This endorses our explanation that the reaction propagation velocity depends on the thermal conductivity of the material (see ESI† for comparison). The voltages generated by both devices are comparatively low. The reason is that a high thermal conductivity is essential for a sustained thermopower wave propagation and to maintain a high reaction temperature.⁽³⁴⁾

4 Conclusions

This work demonstrates a performance comparison of thermopower wave systems based on two complementary thermoelectric materials, Bi_2Te_3 and Sb_2Te_3 , which are N- and P-type semiconductors, respectively. Incorporating both N- and P-type materials resulted in output signals with opposite polarities, which is crucial for developing alternating signal sources. We have experimentally investigated the coupling of an exothermic reaction of fuel (combination of nitrocellulose and sodium azide) to layers of Sb_2Te_3 and Bi_2Te_3 in order to generate output power at high rates. A highly thermally conductive substrate of Al_2O_3 accelerated the thermopower waves, which entrain free electrical carriers to produce high energy discharge rates. Our work demonstrates a new class of micro-power sources and shows that alternating output signals

with opposite polarities can be obtained. This is an important milestone towards making efficient thermopower wave systems for future industrial applications.

Notes and References

- ⁵⁵ †Electronic supplementary information (ESI) available: Experimental measurements of thermal and electrical properties of the substrates and the thermoelectric films, additional data on calculation of specific power. See DOI: <https://doi.org/10.1039/C9TA00000A>
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