

## SUPPLEMENTARY INFORMATION

### Bottom-up engineering of thermoelectric nanomaterials and devices from solution-processed nanoparticle building blocks

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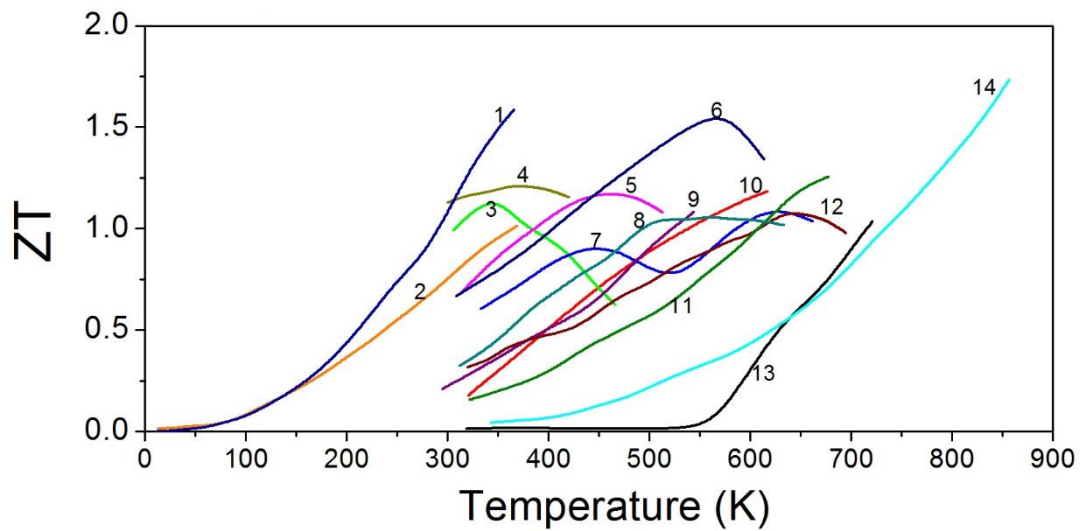
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## 1. State-of-the-art solution-processed ZT materials



**Figure S1.** State-of-the-art ZT values obtained from solution-processed nanoparticle-based thermoelectric materials. **p-type:** **1.**  $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ <sup>1</sup>, **4.**  $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ <sup>2</sup>, **6.**  $(\text{Ag}_2\text{Te})_5(\text{Sb}_2\text{Te}_3)_5$ <sup>3</sup>, **8.**  $\text{PbTe-BiSbTe}$ <sup>4</sup>, **9.**  $\text{AgBi}_{0.5}\text{Sb}_{0.5}\text{Se}_2$ <sup>5</sup>, **11.**  $\text{Cu}_3\text{Sb}_{0.88}\text{Sn}_{0.10}\text{Bi}_{0.02}\text{Se}_4$ <sup>6</sup> and **12.**  $\text{AgSb}_{0.98}\text{Bi}_{0.02}\text{Se}_2$ <sup>7</sup>. **n-type:** **2.**  $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ <sup>1</sup>, **3.**  $\text{K}_{0.06}\text{Bi}_2\text{Te}_{3.18}$ <sup>8</sup>, **5.**  $\text{Bi}_2\text{Te}_{2.5}\text{Se}_{0.5}$ <sup>9</sup>, **7.**  $\text{PbTe}_{0.66}\text{Se}_{0.33}$ <sup>10</sup>, **10.**  $\text{PbTe-Bi}_2\text{Te}_3$ <sup>11</sup>, **13.**  $(\text{PbTe})_{0.72}(\text{PbS})_{0.28}$ <sup>12</sup> and **14.**  $\text{PbS-Ag 4.4\%}$ <sup>13</sup>

## 2. Solution-processed nanoparticle-based thermoelectric devices

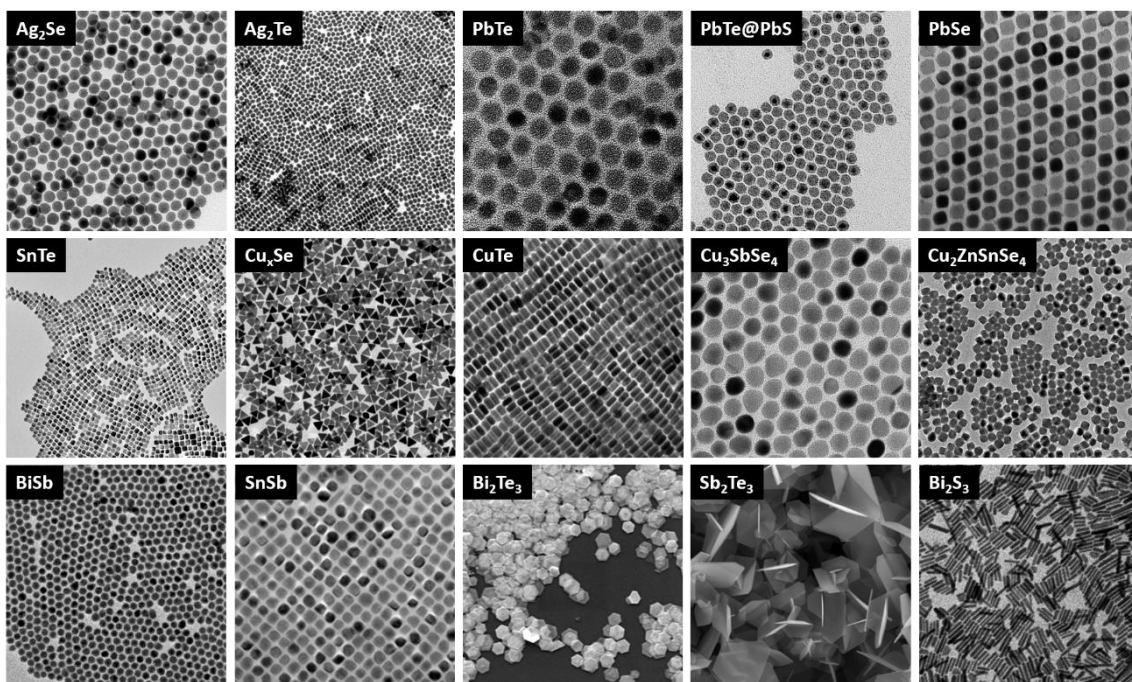
**Table S1.** Key parameters from solution-processed nanoparticle-based thermoelectric devices.

| Reference / Material  | Deposition Method  | Substrate             | n° pairs & architecture             | Output Power  | Flexible? |
|---|--------------------|-----------------------|-------------------------------------|---|-----------|
| <sup>14</sup><br><b>Bi<sub>2</sub>Te<sub>3</sub>-epoxy</b><br><b>Sb<sub>2</sub>Te<sub>3</sub>-epoxy</b>                         | Dispenser Printing | Polyimide             | 50 in-plane                         | 171.6 mV, 10.5 μW,<br>75 μW/cm <sup>2</sup> @ ΔT= 20 °C   | Yes       |
| <sup>15</sup><br><b>ZnSb</b><br><b>CoSb<sub>3</sub></b>   | Screen Printing    | Alumina               | 2 through-plane                     | 27 mV, 0.1 mW/cm <sup>2</sup><br>@ ΔT= 50 °C  | No        |
| <sup>16</sup><br><b>PbTe</b>  | Dip Coating        | Glass Fibers          | -                                   | 1.7 mV @ ΔT= 58 °C  | Yes       |
| <sup>17</sup><br>-  | Dispenser Printing | PDMS                  | - through-plane                     | 7 mV, 2.1 μW @ ΔT= 19 °C  | Yes       |
| <sup>18</sup><br><b>Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub>/Te-epoxy</b>   | Dispenser Printing | Polyimide             | 60 in-plane                         | 152 μW/cm <sup>2</sup> @ ΔT= 20 °C  | Yes       |
| <sup>19</sup><br><b>Sb<sub>1.5</sub>Bi<sub>0.5</sub>Te<sub>3</sub></b><br><b>Bi<sub>2</sub>Te<sub>2.7</sub>Se<sub>0.3</sub></b> | Inkjet Printing    | Polyimide             | 3 in-plane                          | PF @ 75 °C<br>p-type 77 μWm <sup>-1</sup> K <sup>-2</sup><br>n-type 183 μWm <sup>-1</sup> K <sup>-2</sup> | Yes       |
| <sup>20</sup><br><b>Bi<sub>2</sub>Te<sub>3</sub></b><br><b>Sb<sub>2</sub>Te<sub>3</sub></b>                                     | Screen Printing    | Glass Fabric          | 11 through-plane<br>8 through-plane | 2.9 mV, 3 μW @ ΔT= 20 °C<br>90 mV, 3.8 mW/cm <sup>2</sup><br>@ ΔT= 50 °C                                  | Yes       |
| <sup>21</sup><br><b>I-doped PbTe</b>  | Dip Coating        | Glass                 | 2 in-plane                          | 43 mV @ ΔT= 27 °C   | No        |
| <sup>22</sup><br><b>Bi<sub>2</sub>Te<sub>3</sub></b><br><b>Sb<sub>2</sub>Te<sub>3</sub></b>                                     | Screen Printing    | Polyimide             | 8 in-plane                          | 36.4 mV, 40.3 nW @ ΔT= 20 °C  | Yes       |
| <sup>23</sup><br><b>Bi-epoxy</b><br><b>Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub>/Te-epoxy</b>                              | Dispenser Printing | Polyimide             | 10 in-plane                         | 1230 μW/cm <sup>2</sup> @ ΔT= 70 °C   | Yes       |
| <sup>24</sup><br><b>Bi<sub>2</sub>Te<sub>3</sub></b><br><b>Sb<sub>2</sub>Te<sub>3</sub></b><br><b>PEDOT:PSS</b>                 | Screen Printing    | Polyimide             | 7 in-plane                          | 85.2 mV, 1.22 mW/cm <sup>2</sup><br>@ ΔT= 50 °C   | Yes       |
| <sup>25</sup><br><b>Bi<sub>2</sub>Se<sub>3</sub> nanoplates/ PVDF</b>   | Drop-casting       | Free-standing TE foil | N/A                                 | 90 mV @ ΔT= 1.2 °C  | Yes       |
| <sup>26</sup><br><b>Ag<sub>2</sub>Te/</b>   | Dip Coating        | Nylon fibers          | 2 in-plane                          | 3.5 mV, 5 nW, 0.6 μW/cm <sup>2</sup><br>@ ΔT= 20 °C   | Yes       |

| PEDOT:PSS   |  |                         |                   |  |        |
|---|--|-------------------------|-------------------|--|--------|
| <sup>27</sup><br><b>Bi<sub>2</sub>Te<sub>3</sub></b><br><b>Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub></b>   | Dispenser Printing                     | Polyimide               | 25 in-plane       | 33 μW, 2.8 W/m <sup>2</sup> @ ΔT= 20 °C  | Yes    |
| <sup>28</sup><br><b>Cu<sub>1.75</sub>Te NWs /PVDF</b>   | Vacuum Filtration                      | Free-standing TE foil   | N/A               | PF = 23 μWm <sup>-1</sup> K <sup>-2</sup> @ 25 °C                                    | Yes    |
| <sup>29</sup><br><b>WS<sub>2</sub> NSs</b><br><b>NbSe<sub>2</sub> NSs</b>   | Vacuum Filtration/<br>Contact Printing | PDMS                    | 100 through-plane | 38 nW @ ΔT= 60 °C  | Yes    |
| <sup>30</sup><br><b>Bi<sub>2</sub>Te<sub>3</sub></b><br><b>Sb<sub>2</sub>Te<sub>3</sub></b>   | Dispenser Printing                     | Polyimide               | 1 through-plane   | 1.54 nW @ ΔT= 20 °C  | Yes    |
| <sup>31</sup><br><b>Ca<sub>3</sub>Co<sub>4</sub>O<sub>9</sub></b>   | Screen Printing                        | Alumina                 | 10 in-plane       | PF = 0.16 mWm <sup>-1</sup> K <sup>-2</sup> @ 300 °C                                 | No     |
| <sup>31</sup><br><b>(ZnO)<sub>5</sub>In<sub>2</sub>O<sub>3</sub></b>  |  |                         | 10 in-plane       | PF = 1.4 μWm <sup>-1</sup> K <sup>-2</sup> @ 300 °C                                  | No     |
| <sup>32</sup><br><b>Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub></b>  | 3D Printing                            | Free-standing TE pellet | N/A               | ZT = 0.12 @ 43 °C  | No     |
| <sup>33</sup><br><b>Bi<sub>2</sub>Te<sub>2.8</sub>Se<sub>0.2</sub></b>  | Screen Printing                        | Polyimide               | 5 in-plane        | ZT = 0.43 @ 175 °C<br>6.1 μW/cm <sup>2</sup> , 4.1 mW/cm <sup>2</sup><br>@ ΔT= 60 °C | Yes    |
| <sup>34</sup><br><b>TiS<sub>2</sub>/hexylamine</b>  | Drop-casting                           | Free-standing TE foil   | N/A               | 32 μW/cm <sup>2</sup> @ ΔT= 20 °C  | Yes    |
| <sup>35</sup><br><b>PbS QDs</b><br><b>Bi-doped PbTe QDs</b>   | Dip Coating                            | Cellulose paper         | 3 in-plane        | 21.1 mV @ ΔT= 33 °C  | Yes    |
|   |  |                         | 3 through-plane   | 14.2 mV @ ΔT= 33 °C  | Yes    |
| <sup>36</sup><br><b>Bi<sub>2</sub>Te<sub>3</sub></b><br><b>Sb<sub>2</sub>Te<sub>3</sub></b>   | Dispenser Printing                     | Silk Fabric             | 12 through-plane  | 10 mV, 15 nW @ ΔT= 35 °C   | Yes    |
| <sup>37</sup><br><b>Bi<sub>2</sub>Te<sub>3</sub>/Sb<sub>2</sub>Te<sub>3</sub> ChaM</b>  | Brush-painting                         | Polyimide or Glass      | 5 in-plane        | 2.43 mW/cm <sup>2</sup> @ ΔT= 50 °C  | Yes/No |
|   |  | Alumina                 | 1 through-plane   | 4 mW/cm <sup>2</sup> @ ΔT= 50 °C   | No     |
| <sup>38</sup><br><b>Ag</b><br><b>Ni</b>   | Screen Printing                        | Polyimide               | 15 in-plane       | 22 mV, 14.6 μW @ ΔT= 113 °C  | Yes    |
| <sup>39</sup><br><b>Bi<sub>1.8</sub>Te<sub>3.2</sub></b><br><b>Sb<sub>2</sub>Te<sub>3</sub></b>   | Screen Printing                        | Polyimide               | 8 in-plane        | 26.6 mV, 455.4 nW @ ΔT= 20 °C  | Yes    |
|   |  | Glass fibers fabric     | 8 in-plane        | 42 mV, 2.3 μW @ ΔT= 20 °C  | Yes    |
| <sup>40</sup><br><b>(Bi<sub>0.98</sub>Sb<sub>0.02</sub>)<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub></b><br><b>(Bi<sub>0.25</sub>Sb<sub>0.75</sub>)<sub>2</sub>(Te<sub>0.95</sub>Se<sub>0.05</sub>)<sub>3</sub></b> | Dispenser Printing                     | Polyester fabric        | 12 through-plane  | 23.9 mV, 3.11 nW @ ΔT= 22.5 °C   | Yes    |
| <sup>41</sup>   | Screen Printing                        | PDMS                    | 72                | 4.78 mW/cm <sup>2</sup> @ ΔT= 25 °C  | Yes    |

|  |                   |                       |               |   |     |
|--|-------------------|-----------------------|---------------|---|-----|
| <b>Bi<sub>0.3</sub>Sb<sub>1.7</sub>Te<sub>3</sub></b><br><b>Bi<sub>2</sub>Se<sub>0.3</sub>Te<sub>2.7</sub></b><br>42 |                   |                       | through-plane |   |     |
| <b>Bi<sub>2</sub>Te<sub>2.7</sub>Se<sub>0.3</sub></b><br><b>Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub></b><br>43 | Brush-painting    | Alumina               | 4 in-plane    | $\Delta T = 3.7\text{ }^\circ\text{C}$ @ 0.3 A cooling performance          | No  |
| <b>Bi<sub>1.8</sub>Te<sub>3.2</sub></b><br><b>Sb<sub>2</sub>Te<sub>3</sub></b><br>44                                 | Screen Printing   | Polyimide             | 8 in-plane    | 32 mV, 444nW @ $\Delta T = 20\text{ }^\circ\text{C}$                        | Yes |
| <b>TiS<sub>2</sub>/organic</b><br><b>PEDOT:PSS</b><br>45   | Contact Printing  | PET                   | 5 in-plane    | 33 mV, 2.5 W/m <sup>2</sup> @ $\Delta T = 70\text{ }^\circ\text{C}$         | Yes |
| <b>Bi<sub>2</sub>Te<sub>3</sub> NWs</b><br><b>Te-</b><br><b>PEDOT:PSS</b><br>6                                       | Vacuum Filtration | Free-standing TE foil | 6 in-plane    | 56 mV, 32 $\mu\text{W}/\text{cm}^2$ @ $\Delta T = 60\text{ }^\circ\text{C}$ | No  |
| <b>Sn- Bi-doped</b><br><b>Cu<sub>3</sub>SbSe<sub>4</sub></b>   | Filling           | Cu rings              | N/A           | 20 mV, 1 mW @ $\Delta T = 160\text{ }^\circ\text{C}$                        | No  |

### 3. Electron microscopy micrographs of solution-processed nanoparticle building blocks



**Figure S2.** Selection of solution-processed nanoparticles produced by the authors of this review following reported synthesis procedures:  $\text{Ag}_2\text{Se}$ ,<sup>46</sup>  $\text{Ag}_2\text{Te}$ ,<sup>46</sup>  $\text{PbTe}$ ,<sup>12</sup>  $\text{PbTe@PbS}$ ,<sup>12</sup>  $\text{PbSe}$ ,<sup>12</sup>  $\text{SnTe}$ ,<sup>47</sup>  $\text{Cu}_x\text{Se}$ ,<sup>48</sup>  $\text{CuTe}$ ,<sup>49</sup>  $\text{Cu}_3\text{SbSe}_4$ ,<sup>6</sup>  $\text{Cu}_2\text{ZnSnSe}_4$ ,<sup>50</sup>  $\text{BiSb}$ ,<sup>51</sup>  $\text{SnSb}$ ,<sup>51</sup>  $\text{Bi}_2\text{Te}_3$ ,<sup>2</sup>  $\text{Sb}_2\text{Te}_3$ ,<sup>2</sup>  $\text{Bi}_2\text{S}_3$ .<sup>52</sup>

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