Lattice Architectures for Thermoelectric Energy Harvesting

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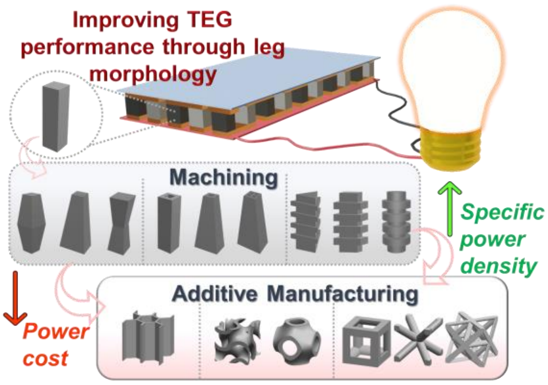
Abstract

Tapping on waste heat for green electricity is a welcomed goal especially in an energy-scarce era. For more than 200 years, the optimization of thermoelectric modules has been either through materials efficiency or device topology (fill factor, aspect ratio, *etc*.). Now, there is a pressing need for a re-examination of its design for higher power performance modules. Specifically, there have been recent reports on the potential of leg geometries for increased output power, but progress is hampered by current leg fabrication methods. In this perspective, we offer a fresh take on how lattice architectural thermoelectric legs are a cost-effective strategy that can drastically improve power output. We will discuss the extent to which lightweight lattice legs can improve output power, its trade-offs with mechanical performance and conclude with strategies to realize them for industrial applications.



TOC

Energy Poverty and Opportunities

The historic Paris Agreement during the 2015 United Nations Climate Change Conference (COP 21) was an agreement that charted a new course in global climate efforts. It called for countries to recognize and respond to the rising global average temperatures by keeping a global temperature rise well below 2^{o}C compared to pre-industrial levels and to pursue efforts in limiting the temperature increase even further to 1.5^{o}C. To achieve this, countries must reduce greenhouse gas emissions significantly. Renewable energy technologies are now more important than ever in their quest towards sustainable low-carbon transportation, heating and electrification. These technologies have been attracting research attention from academia and industries alike to increase their device efficiencies and cost of production. Leading the charge for net zero emissions, the European Union (EU) has been shifting away from coal and towards greener renewable energy sources like hydropower, wind and solar (**Fig. 1a**) [1]. This shift is evident from the steep rise in the contribution of renewables as electricity for EU, especially after 2010. The consumption of renewable energy reached 38 % in 2020 for the EU's total electricity production (**Fig. 1b**) [1]. However, intermittent supply of solar and wind limits the dependency on these sources. To strengthen and expand the supply network for renewable energy, alternative technologies must be examined. One of these technologies is through thermoelectrics materials which inter-converts heat to electricity through a thermal gradient. They are therefore useful solid-state devices for thermal management (cooling) and energy harvesting applications. In 2018, 126 Twh of thermoelectricity was supplied for Italy's energy consumption. [2] This is approximately 6% of the total energy consumed in the country that year. [3] This is an encouraging sign for the uptake and development of thermoelectrics as a steady source of renewable energy for countries, especially with the tremendous amount of industrial waste heat lost to the environment.

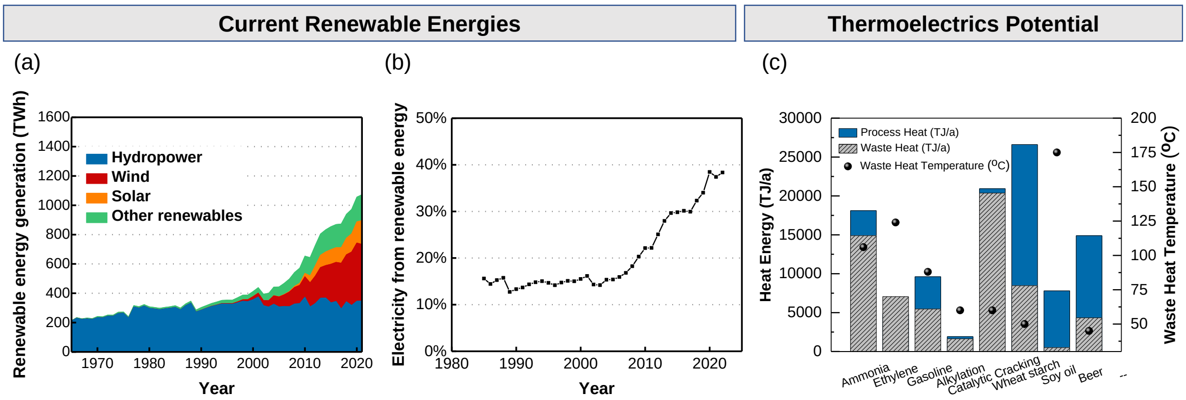


Figure 1. Renewable Energy generation in EU and the potential of thermoelectrics for waste heat harvesting. (a) Stacked area chart of renewable energy generation in EU. Hydropower contributes the highest percentage of renewable energy generation. Other renewables include geothermal, biomass etc. (b) Around 40% of electricity supplied in EU comes from renewable energy. (c) Waste heat generated in EU's chemical, refinery and food processes as a comparison to heat energy used in the processes, per production quantity. Majority of the waste heat temperature of these processes are below 200°C. (a-b) Data obtained from Our World in Data. Accessed date: 01 Feb 2024. (c) Data obtained from Industrial process and waste heat data for EU28, Mendeley Data (2020) Accessed date: 01

Thermoelectrics for Waste Heat Recovery

Despite thermoelectric generators (TEGs) having an efficiency range of 5-15%, which pales in comparison to the efficiencies of steam engines, thermoelectrics is still an attractive energy harvesting material candidate. With industrial waste heat producing approximately 304 terawatt-hours per year (TWh/yr) of energy lost to the environment in the EU alone [4], energy generation through thermoelectrics has caught the attention of the United Kingdom [5] and USA [6]. The vast amount of untapped heat released into the environment allows thermoelectrics to be a low-cost sustainable clean energy solution candidate, with low-grade heat accounting for > 50% of total heat dissipated from industries [7, 8] (**Fig. 1c**) [9]. In addition, sunlight can also be converted into electricity through photothermal materials applied on TEGs [10], which is suitable for outdoor portable power generation.

For large-scale commercial TEG development, the barriers of increasing thermoelectric conversion efficiency and decreasing thermoelectric power costs must be resolved. In the development of efficient thermoelectric materials (quantified through a figure-of-merit zT), nano-structuring and low-dimensional TE materials have been utilized to improve electronic and transport properties compared to bulk counterparts through increased phonon mean-free-path range, quantum confinement and spatial confinement of energy carriers. [11] Apart from the development of low thermal conductivity thermoelectric materials, the other focus on reducing thermal resistance is via the overall device assembly. This has been covered by several research groups through the minimization of interfacial thermal resistances [12, 13], thermal impedance matching [14], leg aspect ratio [15, 16] and fill factor [17, 18] (**Fig. 2**).

Besides the improvement of zT, output power can be maximized by increasing the temperature gradient across the leg. The maximum output power (P_{max}) of TEGs is defined as:

$$P_{\mathrm{max}} = \frac{S_{\mathrm{pn}}^2 \Delta T^2}{4R_{te}} \tag{1}$$

In which the external load impedance (R_{L}) equals electrical resistance (R_{te}) (*i.e.*, the impedance ratio $\lambda = R_{\mathrm{L}}/R_{\mathrm{te}}$ and $\lambda_{P_{\mathrm{max}}} = 1$). [17] S_{pn} is the sum of the Seebeck coefficients of p-and n-type TE legs, and ΔT denotes the temperature gradient across the hot and cold sides of thermoelectric legs. From **Eq. 1**, the improvement of ΔT can significantly increase output power P_{max}. The geometric design of the thermoelectric legs is an important aspect that can be optimized for ΔT for increased electric potential and output power across the legs. [19, 20] However, the topic of thermoelectric leg geometry design is less investigated and is rarely reported despite its potential for cost-effective power generation via an analysis of its power-cost ratio (PCR). This lack of development is due to current manufacturing limitations which have been concorded by researchers. [19-21]

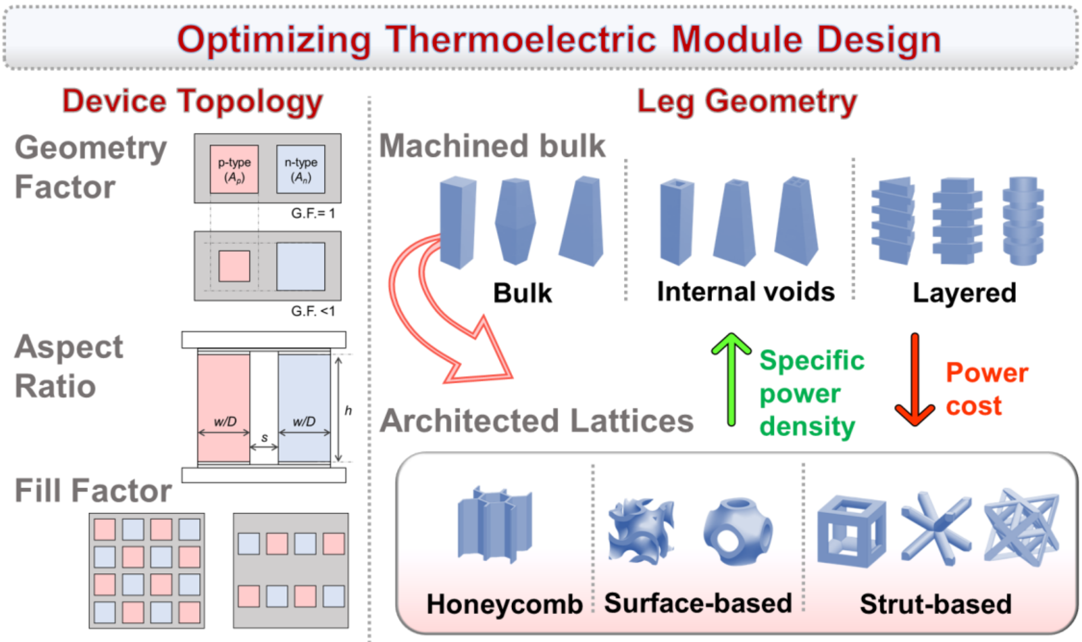


Figure 2. Strategies to optimize thermoelectric module design through device topology and leg geometry. Altering leg geometry from machined bulk designs to architected lattices can increase specific power density and lower power cost.

Lattice Architectures for Energy Harvesting

Engineered lattices are designed to improve application-specific material properties. Their topologies are often inspired by naturally occurring cellular materials that possess architected structures such as truss lattices and Triply Periodic Minimal Surfaces (TPMS) (**Fig. 3**). The exoskeleton of deep-sea hexactinellid sponges [22] and bamboos [23] possess truss-based microstructures while TPMS-based porous structures can be found in butterfly wings and the exoskeletons of weevils [24]. Other forms of bio-inspired lattices such as honeycombs and nacre can also give rise to re-entrant honeycomb metamaterials [25] or nacre-like composites metamaterials [26]. These lattice structures create opportunities for lightweight, high-energy absorption, high strength and sound absorption engineered materials. [27-29] These metamaterials serve to increase the mechanical characteristics of engineered tools and have been reviewed by several groups. [30-32]

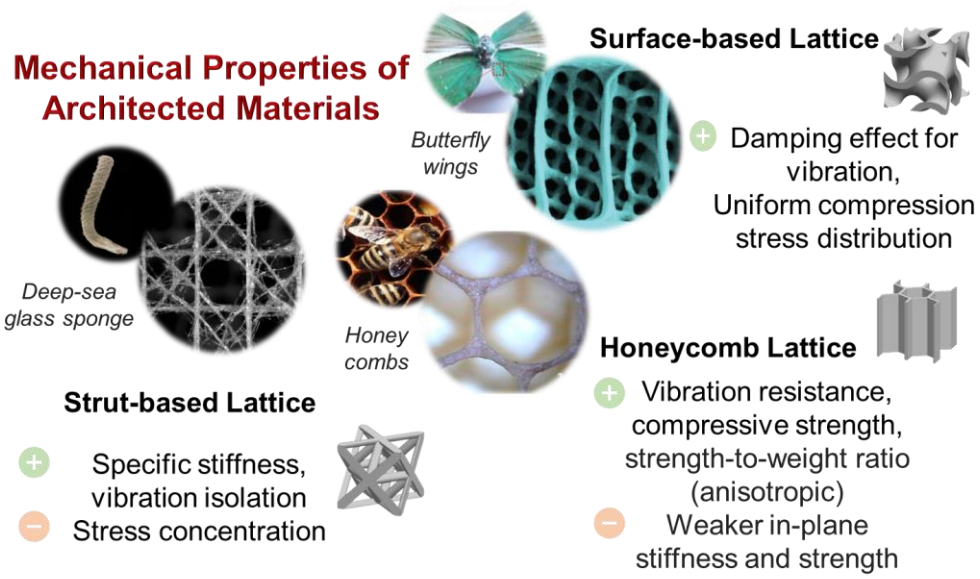


Figure 3. Mechanical properties of lattice structures. Surface-based, honeycomb and strut-based lattices with improved mechanical properties has been engineered to mimic natural occurring structures.

However, with the demands for device integration, there is a large and exciting field of multifunctionality for lattices, beyond just the improvement of mechanical properties.

Emerging research directions of actuators and sensing have shown great potential in the field of functional lattice architectures.[32-34] In particular, the emerging research field of additively-manufactured thermoelectrics, made popular again in the 2000s since the discovery of thermoelectrics in 1821, brings about the potential for energy harvesting through architected lattices.[35-37] In recent years, 3D printed periodic lattice structures has been explored for thermoelectrics applications. Honeycomb architecture legs have shown to possess a larger plastic deformation region and higher fractural strain as compared to typical cuboid legs. [37]By further altering the material properties of the lattice struts, specific strength and ductility can be improved as compared to cuboid legs. [38] The complexity of lattices also widens the areas for heat harvesting, from exterior surfaces to interior of heat pipes. [36] The new world of complex functional lattices made possible through additive manufacturing technology opens up a research area in the balance in thermoelectric device design, where the merits of power and device efficiency must be weighed against the structural performance of thermoelectric lattices and the overall manufacturing cost. Therefore, in this era of increased energy demand and limited material resources, a fresh approach to the need for functional energy harvesting lattices is required.

Improvement in Cost-Effectiveness

The central importance in the consideration of TEG designs is in their cost-effectiveness, *i.e.*, the power generated per thermoelectric fabrication costs. With the rising fabrication costs due to scarce thermoelectric elements such as Te, Ga and Bi [5] and the low energy conversion efficiencies, thermoelectric module designs need to take on a new perspective. Analytical frameworks proposed by Yazawa [18] and Yee [39] examine power output via variance in fill factors and leg lengths of present-day TEG devices with typical cuboid legs. Beyond this, another factor for consideration is the design of thermoelectric legs - from current bulk rectangular legs into lattice thermoelectric legs. To warrant the merits of lattices for cost-effectiveness, we have to dissect it into two main questions – 1. Do lattices increase the thermal gradient across the TEG, and to what extent and 2. The power cost (i.e., price per watt) ($/W) trend for lattices compared to conventional bulk legs for efficient material usage. To demonstrate the feasibility and advantages of power generation from lattice legs, COMSOL Multiphysics® software was used to simulate the thermoelectric properties of Bi_2Te_3 legs with a fix hot end temperature of 400 K. The constant hot end temperature was applied on a leg with an area of 1 cm^2 and a height of 2 cm. The cold end is subjected to an outwards convective heat flux by which the effect of electrical conductivity and thermal conductivity of the 'lattice-air' composite can be observed. Due to the advantages of the open cellular architecture for the reduction of thermal conductivity, we define effective thermal conductivity (κ_{eff}) as the steady-state Fourier conduction of the lattice-air composite. It is worth noting that the configuration modeled here is based on finite heat flux, as opposed to constant ΔT assumed in many literatures. Such finite-heat flux model can be more representative of real-life applications.

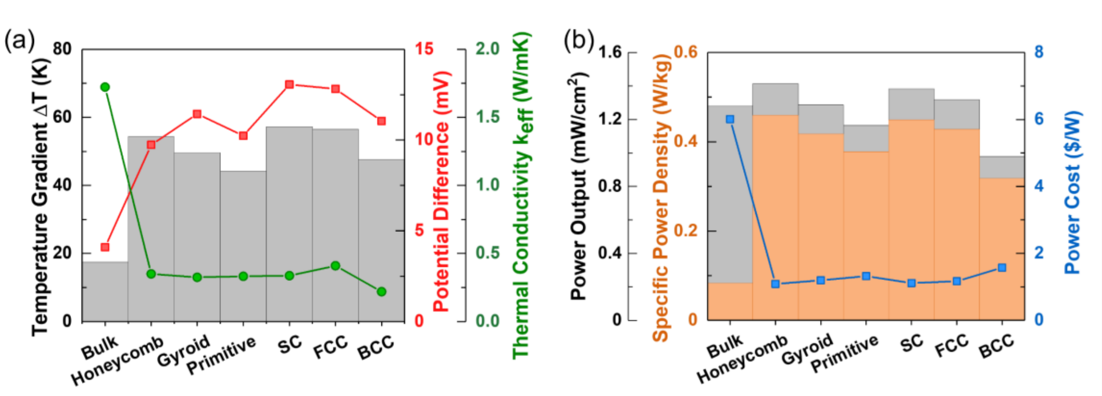


Figure 4. Comparison of thermoelectric properties with bulk and lattices. (a) A higher temperature gradient can arise from lattice legs compared to bulk thermoelectric legs, allowing a higher potential difference generated. The 'lattice-air' composite allows the thermal conductivity to decrease across the leg. (b) The increased in potential difference and reduced thermal conductivity and mass of the leg allows lattice-designed legs to have a higher specific power density and lower power cost, even though power output per area is comparable.

To illustrate the potential roles of lattices in thermal gradient across TEG, lattices of different architectures with a relative density of 0.2 were compared to conventional bulk legs (**Fig. 4**). The advantages of a 'lattice-air' composite design can be seen from the simulation results. Due to the influence of air within the lattice legs and an increased surface area for heat dissipation, there was a remarkable 5-fold decrease in κ_{eff} by altering bulk cuboid legs to lattice-designed ones. This resulted in a 3-fold increase in temperature differential across the hot and cold ends, leading to a higher potential difference generated, as compared to conventional bulk legs (**Fig. 4a**).

Seemingly, the power output $P_{\max}$ of bulk cuboid and lattice legs are comparable with no significant influence by leg design (**Fig. 4b**). To accurately quantify the effectiveness of leg designs, specific output power density (W/kg) and power cost ($/W) (*i.e.,* energy harvested per material weight and cost) should instead be used as metrics. In a bulk leg, 0.08 W was produced for every 1 kg of Bi_2Te_3 used. By changing the leg design into a lattice architecture, a higher output power of 0.46 W is produced for every 1 kg of Bi_2Te_3. This is approximately 6-times the power generated for every 1 kg of Bi_2Te_3 used in a TEG. For thermoelectric systems under constant heat-in flux, the power generation efficiency is proportional to zT/κ_{eff}. [40] As zT is proportional to $1/\kappa_{eff}$, efficiency is thus proportional to $1/\kappa_{eff}^2$. As such, the significant decrease in κ_{eff} through lattice legs proves to be an effective strategy for power generation efficiency and enhanced specific output power. The power cost of TEGs comprises of material, manufacturing and heat exchanger costs, with material costs bearing a significant portion of the overall cost. With the scarcity and thus high costs of inorganic thermoelectric materials, reducing material without compromising specific power output can significantly alleviate the overall power cost. As a basis for material cost-saving comparison between bulk and lattice structures, we will take the material cost of Bi_2Te_3 (*i.e.,* \$500/kg). [18] For bulk cuboid legs, the cost to produce 1 W is approximately \$6. Comparatively, the cost to produce 1 W for lattice-designed legs averages about \$1.20. Honeycomb lattice legs for TEG devices can produce the highest cost-efficient energy at \$1.09/W. These results convince us that lattice thermoelectric legs are approximately 5-times more cost-effective than bulk legs. Thus, the customizability of legs through 3D printing has the potential to increase the overall cost-effectiveness of TEG fabrication through optimized leg design with minimal material wastage.

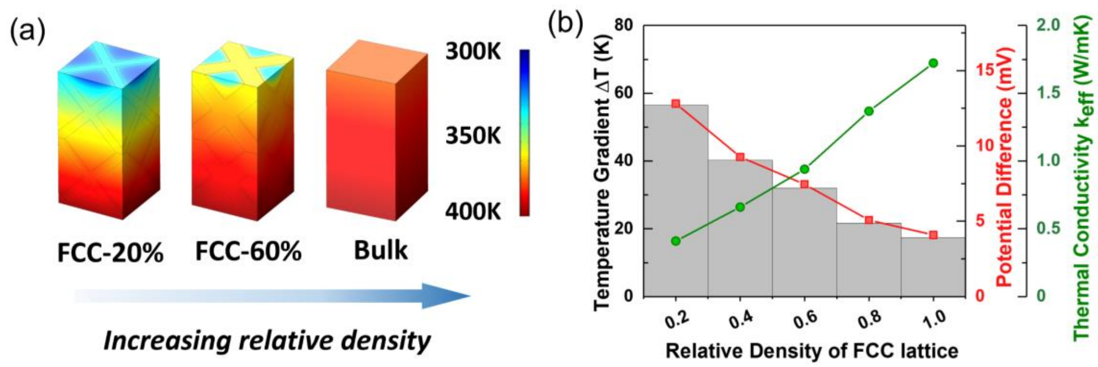


Figure 5. Effects of temperature gradient with relative density of thermoelectric leg. (a) Using FCC lattice as an example, simulation results show the drastic difference in temperature gradient when lattice-designed legs are used instead of bulk legs. (b) The corresponding increase in potential difference and reduced thermal conductivity is evident when relative lattice density decreases.

Trade-offs with Mechanical Robustness

As with many machines in the transport and manufacturing industries, weight is a key factor in lowering fuel consumption and operational costs. The change in subsystems and mechanisms to wireless systems through solid-state TEG can reduce the overall mass. By further substituting bulk thermoelectric legs with lightweight cellular lattice architectures, energy conversion efficiencies can be increased with reduced costs. Besides cost-efficiency, reliability is also an important aspect for the implementation of TEGs in the industry. TEG device failure often occurs at the interface between the thermoelectric materials and electrodes. During operation, thermal stresses will be generated at the connection of the thermal insulating plates, electrode, barrier layer and thermoelectric legs. Usually, the corners between the legs and electrodes experience high thermal stress resulting in micro-crack initiations.[16, 41] These cracks will ultimately increase the contact electrical and thermal resistance leading to a decrease in thermoelectric output performance. To reduce interfacial resistance and improve the device's overall mechanical performance, leg geometry can play an important contributing factor. Simulations have shown that geometric optimization of thermoelectric legs such as asymmetric legs is able to alleviate the concentration of thermal stress at the corners. [41]

The effects of relative lattice density to temperature gradient between the hot and cold ends of the legs are examined in **Fig. 5**. Keeping the hot end temperature constant at 400 K, reducing the relative density will reduce κ_{eff} of the 'lattice-air composite' leg. Thus, a lattice with a smaller relative density can sustain a significantly higher temperature gradient, generating a higher voltage and thus specific power density. However, in exchange for the characteristic lightweight properties of lattices and their potential for reduction of device thermal stresses, the mechanical properties may be compromised (**Fig. 6a**). Mechanical parameters (*e.g.*, fracture toughness, hardness, Young's modulus and compressive strength) for bulk thermoelectric legs as well as strengthening strategies have been reported and consolidated. [41, 42] Besides these material properties, physical properties such as macro design play a role in the overall mechanical properties. While all three lattice structures present some degree of vibration attenuation better than a bulk material due to their high specific stiffness, it is particularly impressive in the honeycomb and surface-based lattices because of the mechanisms present to dissipate energy [43, 44] and the structural design resulting in a low natural frequency, respectively [45]. Under compression, the honeycomb and strut-based lattice display anisotropic properties with the strut-based lattice being prone to plastic buckling and strain localization. With the elimination of joints in surface-based lattices and thus minimization of stress concentration points [29], they possess higher resistance against compressive deformations as compared to strut-based lattices. However, surface-based lattices such as gyroids may require longer slicing times for 3D printing due to the complexity of the interlocking channels. Our preliminary simulation results also showed that lattice Bi_2Te_3 legs of 0.2 relative density possess between 10-20% of the Young's modulus and flexural strength of bulk legs, with honeycomb lattices having the highest Young's modulus and flexural strength at a mere 20% that of bulk legs. To improve the device's mechanical properties, strategies such as encapsulation of the lattice legs in an insulating polymer or increasing the lattice relative density can be considered. These strategies have to be delicately caried out, especially via choosing encapsulant material with ultra-low thermal conductivity. Otherwise, it may compromise power generation due to a smaller thermal gradient across the legs. However, we must not isolate the lattice legs' mechanical properties from the overall TEG device's mechanical performance. The ends of the thermoelectric legs are attached to two thermally insulating face sheets which provide an overall improved torsional rigidity. [46] Similar to sandwich panels used as lightweight structures in airplane wings and sound adsorption panels, the concept of lightweight thermoelectric modules that can withstand bending loads while harvesting energy is feasible. In all, the imperative of improving the device's mechanical properties must be weighed against the application requirements. In most applications, priority is on the net weight of device in which the mechanical strength of bulk legs may be excessive. Heavy impact areas of applications include aircraft fuselage, radioisotope power systems and exhaust systems of gasoline engines in vehicles. Devices that experience minimal impact include medical devices and wireless sensor networks. By identifying the areas in which thermoelectric materials can be applied, we can tailor the material and structural architecture to suppress the performance-limiting mechanisms of mechanical and thermal stresses while maximizing power generation.

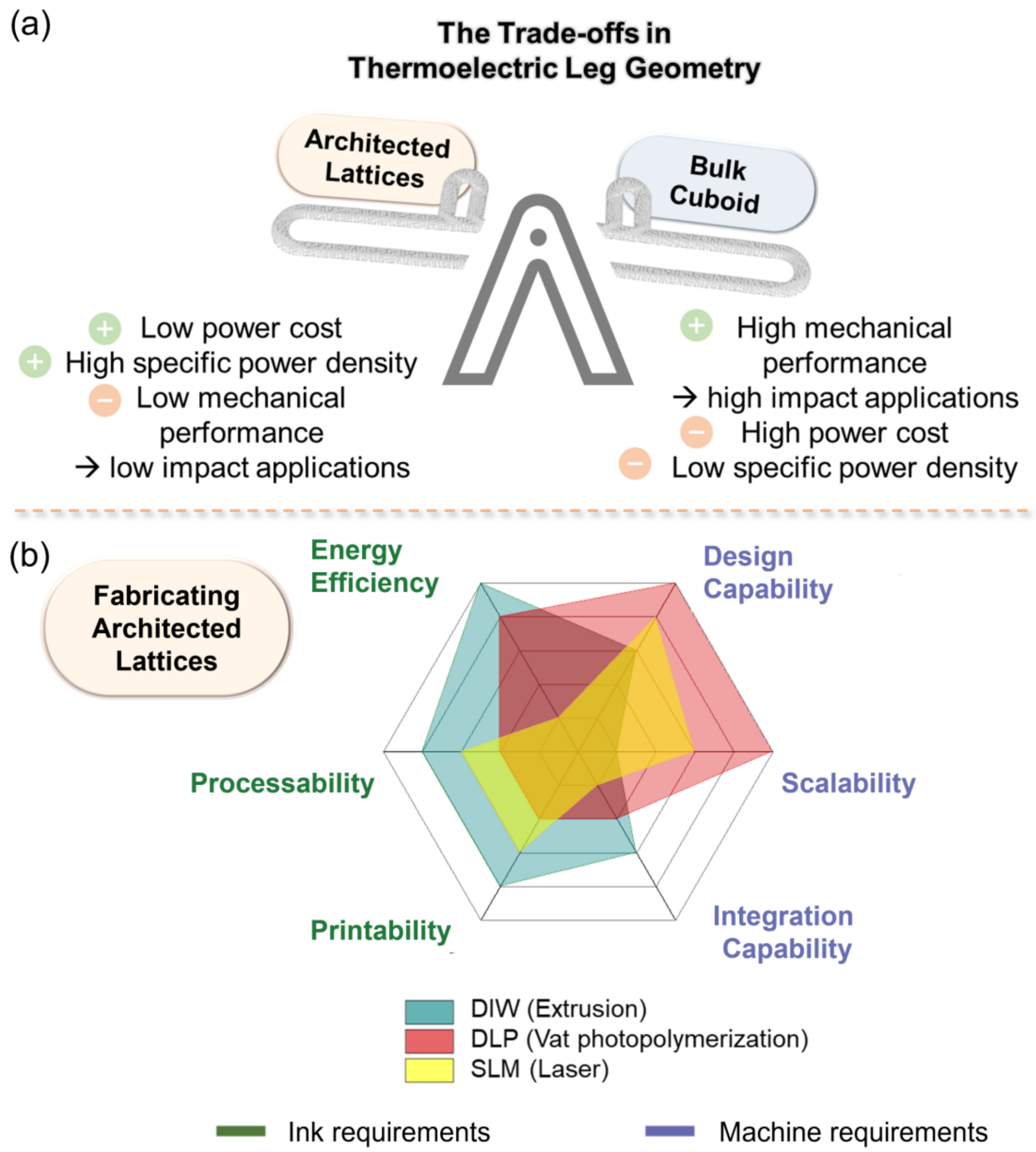


Figure 6. Trade-offs in thermoelectric leg geometry and variables to consider for lattice legs fabrication. (a) Lattice legs offer a lightweight solution for high specific power density with low costs but suffers in mechanical performance. (b) To fabricate architected lattices, various 3D printing techniques can be used. A radar chart is used to weigh the variables required for a successful lattice leg fabrication from 3 commonly used 3D printing techniques.

Integration into Devices and Scaling Up

Therefore, to obtain cost-efficient lightweight thermoelectric modules that can prevent stress and cracking, thermoelectric legs should be redesigned. However, complex lattices are impossible to fabricate using conventional machining. Comparatively, additive manufacturing matches up to its fabrication demands and intricacy. The ease of vertical TEG lattice fabrication will be evaluated based on 3 commonly used 3D printing techniques – direct ink writing (DIW), digital light processing (DLP) and selective laser melting (SLM). Factors classified according to the machine and ink requirements will thus aid readers on the technology most suitable for lattice intricacy, mass production and materials research purposes (**Fig. 6b**). Apart from these 3D printing techniques, inkjet printing and screen printing are useful for the fabrication and customization of thin film TEGs.

Design capability refers to the ease of printing complex structures. DIW technique is less versatile for the direct printing of overhangs. Comparatively, the powder bed in a SLM printer functions as the support bed, while photopolymerized layers in DLP systems prevent overhang segments from collapsing. *Integration capability* refers to the technique's capabilities for multi-material and device printing in the same print. The multi printheads in DIW systems give them an advantage. Multi-material segmented legs that offer efficiency across a wide temperature range [47] can be 3D printed with different printheads. Multi-material 3D printing also offers the possibility of reducing contact resistance between the legs and electrode through the formation of a thin diffusion layer when different materials undergo co-sintering. However, the quest to integrate multi-material printing into fully 3D printed thermoelectric devices has been rather sluggish due to the main challenge of thermal expansion difference between materials, which easily result in cracks and lamination between materials. This domain of functional multi-material printing has also been an ongoing research area in the wider 3D Printing field for electrochemical energy devices and electronic devices. The understanding of reaction kinetics, thermal stress and design fabrication are thus important research aspects to the successful fabrication of multi-material structures. Next, we define s*calability* as the ability for high printing throughput in a single print job while maintaining consistency in the material's properties from batch to batch. DLP systems require the least time for multiple prints due to the simultaneous photocuring of an entire layer. This stands in comparison to SLM and DIW systems which require laser rastering and extrusion respectively, through each layer of a single leg.

With regards to ink requirements, *printability* refers to the ink's ability to achieve successful prints. In SLM printers, powders are required to flow for recoating the powder bed. In a similar vein, inks for DLP are required to have low viscosities (< 5 Pa.s) for the recoating of layers. For materials extrusion, it is determined by the ease of slurry extrusion ($<10^3$ Pa.s) through the nozzle without clogging and maintain the filament shape without sagging. In this regard, low viscosity-inks with high inorganic thermoelectric filler content, especially for DLP systems, are more challenging to formulate. *Processability* is defined as the ease of feedstock preparation and the requirements for post-processing (binder burn-off). The need for large volumes of powder in powder bed systems makes them cost-inefficient, especially in preliminary materials research stages. Due to high binder content in low-viscosity DLP inks, binder burn-off is typically a long process with slow ramping rates. To tackle the standing issue of densification in 3D printed thermoelectrics, inorganic sintering aids [37, 48, 49], lower-dimensional thermoelectric materials [50], organometallic precursor solutions [51, 52] or high laser power melting [53, 54] can be utilized. It should be noted that different AM techniques and printing directions may result in varying densities and mechanical properties for the same lattice design, and it is yet a research area to be explored. Varying the annealing process parameters can also modulate the amount and type intrinsic defects to achieve optimal carrier mobility and thermoelectric performance. [55] Lastly, e*nergy efficiency* (*i.e*., the inverse of energy intensity) relates the amount of energy input from the ink preparation stage to the printing process and finally to obtaining the resultant inorganic thermoelectric structure. SLM printers are energy intensive compared to DIW and DLP systems due to requirements of large powder volume and high energy laser during printing. Comparatively, DIW and DLP systems are less energy intensive as their systems use compressed air for extrusion and ultraviolet (UV) irradiation for photocuring respectively. Inks for DIW and DLP are often homogenized using planetary centrifugal mixers. Even though their printed parts also often require a post-printing step of debinding and sintering to obtain the inorganic thermoelectric structure, these are often done in large batches, increasing the energy efficiency of their processes.

Overall, with its ease of ink preparation and printing, DIW systems are highly suitable for thermoelectric 3D printing materials research. This is evident from the number of literatures on DIW for 3D printed thermoelectrics. Despite its merits, the technology lacks the potential for scaling up to industrial demands in the future. On the other hand, DLP systems have the potential to produce higher complexity thermoelectric lattices in a shorter amount of time. The focus and development of DLP-based 3D printed thermoelectric structures is thus believed to have the potential for the industrial adaptation of 3D printed thermoelectric devices.

Outlook and Roadmap

Waste heat is a longstanding issue to tackle, yet an abundant commodity to leverage upon. To accelerate the practical utilization of thermoelectrics for industrial applications, a fresh approach to the development of cost-effective yet efficient and mechanically robust thermoelectric devices must be taken. The energy cost needs to be analyzed, beyond the standard engineering practice of increasing the efficiency of energy conversion *zT*. The seemingly paradoxical challenge of balancing the trade-offs between manufacturing cost, energy conversion efficiency and structural performance requires urgent consideration given the deepening energy crisis. Regarding the lattice design, the relative density, geometry and variation of unit cells are factors that require investigation for its efficacy to further increase specific power density whilst providing sufficient mechanical stability for its end use. Recent advances in machine learning in energy research has accelerated the research from bottom-up materials designs to top-down system optimization for effective utilization of thermal energy, and should be tapped on. [56] Together with the optimization at the material level (*i.e.,* energy conversion efficiency) and device level (*i.e.,* fill factor, leg geometry, and thermal and electrical impedance matching), thermoelectric devices can finally take root for as a sustainable, low-cost energy solution. Specifically, by drawing on the mechanical strengths of bioinspired lattice structures and fabrication cost-effectiveness from additive manufacturing, the solution to practical large-scale energy harvesting through lattice-designed thermoelectrics might be one step closer to reality.

Conflicts of interest

There are no conflicts to declare.

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Quotes for highlight

- By further substituting bulk thermoelectric legs with lightweight cellular lattice architectures, energy conversion efficiencies can be increased with reduced costs.
- To warrant the merits of lattices for cost-effectiveness, we have to dissect it into two main questions – 1. Do lattices increase the thermal gradient across the TEG, and to what extent and 2. The power cost (*i.e.,* price per watt) ($/W) trend for lattices compared to conventional bulk legs for efficient material usage.
- By identifying the areas in which thermoelectric materials can be applied, we can tailor the material and structural architecture to suppress the performance-limiting mechanisms of mechanical and thermal stresses while maximizing power generation.