




Review

Advances in Thermo-Electrochemical (TEC) Cell Performances for Harvesting Low-Grade Heat Energy: A Review

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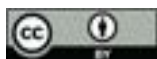
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Abstract: Thermo-electrochemical cells (also known as thermocells, TECs) represent a promising technology for harvesting and exploiting low-grade waste heat (<100–150 °C) ubiquitous in the modern environment. Based on temperature-dependent redox reactions and ion diffusion, emerging liquid-state thermocells convert waste heat energy into electrical energy, generating power at low costs, with minimal material consumption and negligible carbon footprint. Recent developments in thermocell performances are reviewed in this article with specific focus on new redox couples, electrolyte optimisation towards enhancing power output and operating temperature regime and the use of carbon and other nanomaterials for producing electrodes with high surface area for increasing current density and device performance. The highest values of output power and cell potentials have been achieved for the redox ferri/ferrocyanide system and $\text{Co}^{2+}/^{3+}$, with great opportunities for further development in both aqueous and non-aqueous solvents. New thermoelectric applications in the field include wearable and portable electronic devices in the health and performance-monitoring sectors; using body heat as a continuous energy source, thermoelectrics are being employed for long-term, continuous powering of these devices. Energy storage in the form of micro supercapacitors and in lithium ion batteries is another emerging application. Current thermocells still face challenges of low power density, conversion efficiency and stability issues. For waste-heat conversion (WHC) to partially replace fossil fuels as an alternative energy source, power generation needs to be commercially viable and cost-effective. Achieving greater power density and operations at higher temperatures will require extensive research and significant developments in the field.

Keywords: thermo-electrochemical cells; energy harvesters; low grade waste heat; wearable electronics; micro supercapacitors; thermoelectrics

1. Introduction

The conversion of energy from primary energy sources to their final use is accompanied by several losses in the form of waste heat. Forman et al. [1] have estimated the waste heat potential in transport, commercial, industrial and residential sectors. Nearly 72% of the primary energy consumed was found to be lost as waste heat, of which ~63%

of waste heat streams occurred at temperatures below 100 °C. In the USA, conventional industries and power plants are known to annually waste over 8000 TWh as low-grade heat [2]. This ubiquitous waste heat energy in the form of vehicle exhaust, industrial waste heat, geothermal heat, body heat, etc., is found distributed almost everywhere. However, vast amounts of low-grade heat is mostly discarded and rarely exploited commercially due to its intrinsic low temperatures, space-time variations and the lack of cost-effective and efficient energy-recovery technologies [3]. Renewable energies such as solar, wind or nuclear, which have a negligible carbon footprint compared to non-renewable fossil fuels, [4] are seeing a great resurgence. The recent UN Climate Change Conference, held at Glasgow, UK [5] was focussed on the threat of climate change through mitigation and conservation efforts, deforestation, methane abatement and reducing the carbon footprint and emissions from energy production. Due to the vast and renewable nature of waste heat energies and their huge energy potential, harvesting low-grade waste heat is beginning to attract a great deal of attention as a promising zero-carbon source of electricity.

The transformation of thermal gradients into electrode potentials for generating electricity has been investigated for a long time [6]. The generation of electric potential in the presence of a temperature gradient between different electrical conductors/or semiconductors was discovered by Thomas Johann Seebeck in 1821. Seebeck coefficients, representing the potential difference generated per unit temperature difference, are typically in the order of few $\mu\text{V}/\text{K}$ for devices based on semiconductor materials [7]. Early research was mainly in the field of solid-state thermoelectric generators (TEGs) consisting of n- and p-type semiconductors connected in series as modules and then connecting a number of modules in parallel between the heat source and a cold sink. Under the influence of thermal gradients, mobile charge carriers ‘electrons/holes’ diffused from the hot electrode to the cold, building up charges and a small potential difference [8]. Solid-state semiconductor thermoelectrics have long been investigated for the conversion of thermal to electrical energy, and several exciting advances have been made [9–11]. Such devices are, however, not really suitable for low-grade heat harvesting, due to their relatively low efficiencies at ambient temperatures [12].

Early studies, which later led to thermoelectrochemical converters, were used for applying thermal corrections to electrochemical processes in the field of current sources and in the production of galvanic coatings. However, the gradual dissolution of anodes was found to limit their commercial application [13]. Landry [14] suggested the integration of low- and high-temperature processes to minimize heat losses. Wakao and Nozo [15] developed a process for recovering thermal energy from low-level heat generated during the mixing of nitric acid with water. The advent of nano-structured thermoelectrics has led to a great deal of interest in waste-heat conversion (WHC) to electricity [16]. Although early research was dominated by thermoelectrics, a number of articles have been published in leading journals on new types of WHC devices using a wide range of phenomena. These include thermomagnetic generators [17], ionic heat-to-electricity conversion [18], thermo-osmotic systems [2], liquid-state thermocells [19], high temperature pyroelectric systems [20,21] and organic Rankin cycles [22] for applications in industries, construction, transportation and energy sectors. These devices are used for harvesting energy from a heat source such as automotive exhaust systems [23], fuel cells [24] or hypersonic engines [25]. Such energy harvesting can reduce the charging requirements on batteries while eliminating wired power connections. Most R&D in this field is focussed on developing better n-type and p-type thermoelectric materials.

Thermoelectrochemical (TEC) cells or ‘thermocells’ are an alternative device design based on redox-active electrolytes which can produce much higher potential differences (mV/K vs. $\mu\text{V}/\text{K}$ for solid-state semiconductor devices). A thermocell has two identical electrodes in contact with a redox couple electrolyte in the cell and an external connection [26]. Under a thermal gradient, the redox reaction causes the oxidation of the redox couple at the anode and reduction at the cathode. The reduced species are transported back to the anode through diffusion, convection and migration in the electrolyte, creating a

continuous reaction and current flow. Within the degradation constraints of cell materials, thermocells reaction and current flow slowly generating electricity with units of cooling materials, thermal producing emissions. Ghum and Gentry (2012) have also discussed an alternative thermally regenerative Electrochemical cycling (TEC) design where electrochemical materials are used to convert heat into electricity (TEC) within the constraints of Carnot-cycle efficiency.

A significant step in the development of the electrochemical systems was the study of TEC of the gas-based electrochemical cells where reactions at the electrodes are redox with the release of gas or uptake of gas (e.g., HNO_3 (HNO_2), N_2O and OHNO_2 ; H_2O_2 , Br_2 and Br^-) released or absorbed at the electrodes. These reactions are reversible and the reaction is reversible. Representative examples of such thermocells are shown in Figure 1. Figure 1a shows a KBr/Br_2 -based TEC cell. When the intercalation of lithium gas (Br_2) graphite electrode plays a key role in the cell operation. At the anode (cathode) (anode), Br_2 is oxidized to Br^- and Br^- is reduced to Br_2 and dissociates into Br^- and Br_2 . These ions diffuse through the bridge to the hot electrode (anode) and transform into Br_2 and Br_2 releasing. These electrons travel from the anode to the cathode in the outer loop, completing the circuit. Figure 1b shows a cell based on an organic reaction, propene and propanone. Reaction at the anode involves $\text{CH}_3\text{CH}(\text{OH})\text{CH}_3 \rightarrow \text{CH}_3\text{C}(\text{O})\text{CH}_3 + \text{H}^+$ and the reverse reaction occurs at the cathode. Such processes allow the relatively long and continuous generation of potential difference and provide higher output power values. However, these systems are unstable, and their operation is accompanied by the release of toxic products such as bromine, hydrogen or nitrogen oxides.

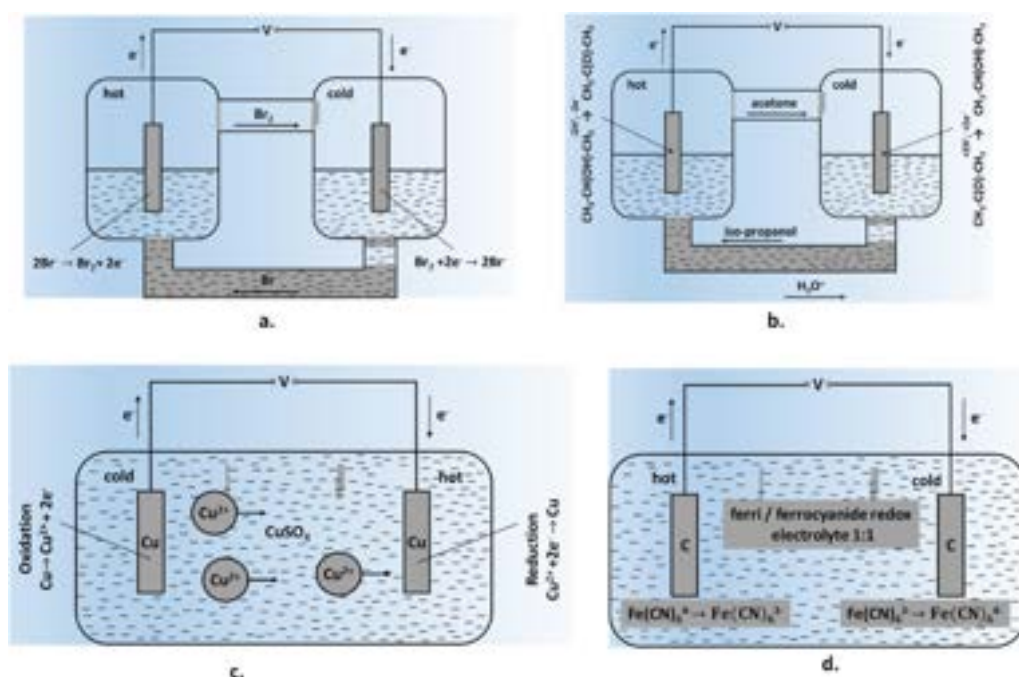


Figure 1. Schematic representations: (a) KBr/Br_2 -based TEC cell, (b) acetone-isopropanol thermocell, (c) Cu^{2+}/Cu system in CuSO_4 solution with soluble electrodes, (d) a ferri/ferrocyanide redox couple couple $[\text{Fe}(\text{CN})_6]^{3-}/[\text{Fe}(\text{CN})_6]^{4-}$ with inert electrodes.

Electrolyte-based TEC systems are preferable to solid-state thermoelectrics due to several factors. Firstly, the higher values (by about 100 times) of the Seebeck coefficient allow their application at low temperatures. Secondly, their high flexibility, using liquid systems and flexible electrodes, simplifies a large number of usage options without additional costs for the development and just adjustment of the production equipment. The overall simplicity of manufacturing and low material costs ensure relatively lower associated costs. However, TECs also have some disadvantages. The fundamental difficulty in the practical application of TECs is the high internal resistance in the electrolyte (especially

non-aqueous ones [31]) and low rates of electrode reactions, leading to low values of the exchanging current and, correspondingly, to the low output powers of the device. The most promising solutions to these problems are associated with the use of aqueous alkaline electrolytes with low resistance [32], applying a catalyst to the electrode material [33,34] and increasing the specific surface area and structuration of the electrodes [33,35].

If we consider thermocells based on metals in solutions of their own salts, then the period of their operation is limited by the consumption of the electrode or they become quasi-continuous if it is possible to change the polarity of hot and cold heat sources. Attention also needs to be paid to the finite operating life, which depends only on the degradation of the electrodes or electrolyte or both, especially when considering cells based on cyanoferrate/ferrites. Another important limitation of electrochemical converters is the significant differences in operating modes, depending on the relative location of the cell and the source of heat flow. Attempts are being made to use membranes, separators and gel electrolytes, which significantly mitigate the results of this effect.

Growing interest in energy recovery from low-grade heat (<100 °C, or even <150 °C/200 °C) is reflected in the number of research publications in the field increasing from 50 articles/year in 2010 to over 200 articles/year in 2019/2020 [36]. In addition to the magnitudes of the Seebeck coefficient, the efficiency of thermocells is affected by several factors. The conversion of heat to electricity in liquid thermocells occurs via two main processes: redox reactions at the electrodes and mass transport in the electrolyte. This process involves three key parameters: thermopower (S_e), representing reaction thermodynamics, which is dependent on the thermopower of redox species in the electrolytes; effective electrical conductivity (σ_{eff}), representing reaction kinetics, which is dependent on electrode resistance and diffusion/migration resistance in the electrolytes; and effective thermal conductivity (k_{eff}), which is based on thermal conduction/convection in electrolytes under thermal gradients [37]. While high Seebeck coefficients are an important requirement for thermocells, the maximum power output is limited by several overpotentials such as ohmic, charge-transfer and mass-transport overpotentials within the cell and their temperature dependence [38]. The performance of thermocells is defined by a unitless parameter ‘Figure of Merit (ZT)’ in terms of potential (ΔV) and thermal (ΔT) gradients:

$$ZT = \left[\frac{\Delta V}{\Delta T} \right]^2 \frac{\sigma}{\kappa} \quad (1)$$

Considering the influence of mass transport, a modified ‘Figure of Merit (ZT*)’ can be written as:

$$ZT^* = \left(\frac{z^2 F^2}{R} \right) \frac{S_e^2 D_{lim} c}{R} \quad (2)$$

where F and R are the Faraday and gas constants, respectively, z the charge on the ion, c the concentration of the redox couple and D_{lim} the limiting diffusion coefficient [39]. Currently, the application of thermocells is limited by their low power output and conversion efficiency. Typical thermocell devices have a conversion efficiency of <1% relative to the Carnot cycle [40]; however, conversion efficiencies up to 3.95% have been achieved in thermocells based on carbon nanotube electrodes [41]. Being a complex and interdependent issue, multifaceted approaches are required for increasing the efficiency of thermocells towards becoming commercially viable. Several strategies are being employed for enhancing S_e (new redox couples, ionic concentration gradients, ionic solvation structures), enhancing σ_{eff} (optimization of electrolytes and electrodes), reductions in k_{eff} (thermal separators, gel based electrolytes), etc. [42,43].

In this article, we present an in-depth overview of the advances made in TEC design and performance, focussing on different aspects, new growth areas and emerging concepts in the choice of redox couples, electrolytes, electrode design and configurations. The article is organized as follows. A brief summary of historical developments in the field is provided in Section 2. Sections 3 and 4 highlight developments in redox couples, electrolytes and

electrodes, spanning several research directions. Emerging applications of thermocells in the fields of wearable and portable devices, energy storage and associated applications are presented in Section 5. The article concludes with highlighting economic concerns and future perspectives.

2. Historical Background

Some of the early-generation TEC cells involved the gradual dissolution of electrodes, limiting their long-term prospects. One such example is shown in Figure 1c, consisting of soluble metal (Cu) electrodes placed in CuSO_4 solution at two different temperatures. Such a cell will work until there is a certain degree of electrode consumption, then require a reversal of the temperature gradient for operating in the opposite direction. A significant development was the use of inert electrodes, providing surfaces for released gaseous products such as $\text{NO}/\text{N}_2\text{O}_4/\text{HNO}_2$ or Br^- ; the released volatile products were condensed at the counter electrode, where the reverse reaction occurred [29,44]. While such processes permitted relatively long and continuous generation of the potential difference as well as higher output power values, these systems had limited stability along with the continuous release of noxious gases. The next stage involved the utilization of redox electrolytes, providing relatively fast kinetics of the electrode process and high values of the hypothetical Seebeck coefficient. Systems based on potassium hexacyanoferrates/ferrites were the most widespread [45]; a schematic representation of this cell is given in Figure 1d. As this system has a negative Seebeck coefficient (-1.4 mV/K), oxidation takes place at the hot electrode and reduction at the cold electrode. At the hot electrode, $\text{Fe}(\text{CN})_6^{4-}$ loses an electron and transforms into $\text{Fe}(\text{CN})_6^{3-}$ which diffuses back towards the cold electrode. At the cold electrode, $\text{Fe}(\text{CN})_6^{3-}$ picks up an electron to transform into $\text{Fe}(\text{CN})_6^{4-}$, which then diffuses towards the hot electrode thereby setting up a perpetual motion of ions and the generation of electric current. It can be seen that the electrodes remained inert during the process and were not consumed. There has been a significant progress in the cell design and yields since early studies on this system.

A large number of TEC converters, electrode materials and electrolytes have been investigated in recent years. An ideal TE material should have high values of S_e to improve energy conversion, high electrical conductivity σ to minimize joule heating and low thermal conductivity k to maintain the thermal gradient and a high figure of merit. Improvements in ZT are often restricted by the interdependence of S_e , σ and k . Basic characteristics of some of these TE materials are summarized in Table 1.

Table 1. Properties of thermoelectric materials, thermogalvanic cells and organic polymers at room temperature.

| TEC Devices | Seebeck Coefficient (mV/K) | Electrical Conductivity (S/cm) | Thermal Conductivity (W/(cm·K)) | ZT |
|--|----------------------------|--------------------------------|---------------------------------|---------------|
| Thermoelectric Semiconductor Materials | | | | |
| Heusler alloy [46] | -0.1 – 0.6 | 1000 – 2000 | 2.7 – 3.0 | 1 – 6 |
| Copper chalcogenides Cu_{2-x}E (E = S, Se, Te) [11,47] | 0.15 – 0.2 | 1500 – 4000 | >0.009 | 1 |
| Bi_2Te_3 alloys [48] | 0.18 – 0.25 | ~ 1000 | ~ 0.02 | 0.5 – 1.5 |
| Thermogalvanic Cells Based on Ferri/Ferrocyanide | | | | |
| Organic solvent [49] | 1.5 – 10 | <0.01 | 0.002 | 0.002 |
| Aqueous solvent [50] | 1 – 13 | 0.6 | 0.006 | 0.03 |

Table 1. Cont.

| TEC Devices | Seebeck Coefficient (mV/K) | Electrical Conductivity (S/cm) | Thermal Conductivity (W/(cm·K)) | ZT |
|--|----------------------------|--------------------------------|---------------------------------|-------|
| Organic and organometallic polymers | | | | |
| polyaniline/graphene/ polyaniline/ double-walled carbon nanotube [50] | −1.14 | 8 | ~0.006 | 1.05 |
| poly (3,4-ethylenedioxy thiophene)/carbon nanotube [51] | 0.13 | 10.8 | - | 1.825 |

In addition to the cells mentioned above, several other types of thermoelectric cells have been developed. These are briefly listed below:

- TEC cells with phase transitions during mass transfer have been studied for a relatively long time (the first publications date back to the 1970s), but these have not found practical application due to their complexity of design and rapid degradation of components. Wang et al. [52] have recently developed electrochemical sodium heat engines based on phase-change reactions. Nevertheless, their important advantage is their continuous operation, which does not require changing hot and cold sources.
- Typical TECs are non-isothermal electrochemical cell systems consisting of two electrodes, an electrolyte and a separator. The conversion of thermal energy into electrical energy involves electrode kinetics, thermodynamics, and heat and mass transfer. TEC cells involve changes in the aggregate state of oxidized and reduced forms (nitric acid and nitrogen oxides or KBr and Br₂) during continuous operation [13,35].
- Thermally regenerative electrochemical cycles (TREC) consist of two electrodes with opposite thermopowers; anodes and cathodes generally have positive and negative thermopowers, respectively. For negative thermopower, the cycle involves cooling, discharging, heating and charging; for positive thermopower, the cycle involves heating, charging, cooling and discharging [53].
- Other cell configurations include thermogalvanic cells (TGC) based on soluble, reversible metal electrodes in solutions of their own salts at different temperatures—TECC cells, using inert electrodes placed in a redox electrolyte or ionic liquid; TGC-Li cells, based on a Li⁺/Li redox system; TRABs (thermally regenerative ammonia-based batteries); DTCCs (direct thermal charging cells); etc. [54–57].

3. Redox Couples and Electrolytes

3.1. Redox Couples

There are two key research areas for increasing the current-generating efficiency of thermocells, namely increasing the potential difference in the cell and the current density at the electrodes. Cell potential can be enhanced by developing combinations of redox couples and electrolytes, increasing the maximum cell temperature through optimized design, solid electrolytes, separators, boiling electrolytes, etc. Current density can be improved using electrodes with large surface areas as well as through electrolytes with high rates of mass transfer. These aspects will be discussed in the following sections as potential strategies to improve the efficiency of thermocell devices.

The Seebeck coefficient of a redox couple, which determines the maximum potential difference generated in a thermocell, is a measure of the entropy change during oxidation or reduction. Structural changes in the redox species, solvent shells and solvent interactions are known to play key roles [58]. Although Seebeck coefficients have been measured/computed for a number of redox couples, their application in thermocells depends on additional factors such as thermal stability, electro-chemical reversibility and avail-

ability [19]. Migita et al. [59] have reported on thermo-electromotive forces and reaction entropies for a number of transition-metal redox couples in an amide-type room temperature ionic liquid using a non-isothermal cell. The highest Seebeck-coefficient values were reported for $[\text{Fe}(\text{CN})_6]^{3-/4-}$ as -1.49 mV/K and Fe(III/II) as 0.96 mV/K . Laux et al. [60] have reported on the iodide/triiodide (I^-/I_3^-) redox couple in aqueous as well as non-aqueous solutions and in ionic liquids. The Seebeck coefficient in this system showed a strong dependence on the electrolyte concentration, changing threefold between 0.01 M and 2 M ethyl ammonium nitrate solutions; a maximum value of 0.97 mV/K was recorded at 0.01 M .

Abraham et al. [39] have reported on cobalt-based redox couples: $\text{Co}^{2+/3+}(\text{bpy})_3(\text{NTf}_2)_{2/3}$, ($\text{bpy} = 2,2'$ -bipyridyl; $\text{NTf}_2 = \text{bis}(\text{trifluoromethanesulfonyl})\text{amide}$ solutions, recording a maximum Seebeck coefficient of 2.19 mV/K . This high Seebeck coefficient was attributed to changes in the spin state of $\text{Co}^{2+/3+}$, providing additional electronic contributions. Anari et al. [61] combined ferrocene and iodine results for enhancing thermoelectrochemical waste-heat harvesting abilities in terms of Seebeck coefficient and power output. These two couples were found to interact even in dilute solutions, and displayed a mixture of ferrocene, ferrocenium, iodine and triiodide ions; the highest Seebeck coefficient of 1.67 mV/K was observed by combining dibutanoylferrocene and iodine. Zinovyeva et al. [62] have reported on an organic thiolate/disulfide redox couple (McMT^-/BMT), derived from 2-mercapto-5-methyl-1,3,4-thiadiazole (McMT); the thermoelectric power was found to depend strongly on the concentrations of the redox couple as well as the ionic liquid. Seebeck coefficient data for a number of redox couples are given in Table 2.

In combined TEC systems, the unidirectional nature of mass and charge transfer in electrochemical systems creates difficulties in the design and manufacture of ready-made converters. Conductor arrangement can reduce the effective electrode surface area relative to the area of the TEC device by connecting cells in series or parallel. Kim et al. [63] showed that the Seebeck coefficient for $\text{Fe}^{2+}/\text{Fe}^{3+}$ transition can change signs for different aqueous systems. There are also reports of hypothetical Seebeck coefficients of up to 7 mV/K measured in electrolytes without a redox couple. The temperature-dependent potential in these systems was attributed to a combination of differences in Eastman entropies and charge-density changes at the electrode surfaces due to the Soret effect [64,65].

Table 2. Seebeck coefficients for several redox couples [13,66].

| Redox Couples | Seebeck Coefficient (mV/K) |
|--|----------------------------|
| $\text{Np}^{3+} \rightarrow \text{Np}^{4+} + \text{e}$ | 2–2.25 |
| $\text{Fe}(\text{CN})_6^{3-} + \text{e} \rightarrow \text{Fe}(\text{CN})_6^{4-}$ | 1.5 |
| $\text{Hg} + 4(\text{CN})^- \rightarrow \text{Hg}(\text{CN})_4^{2-} + 2\text{e}$ | 1.65 |
| $\text{H}(\text{g}) \rightarrow \text{H}^+ + \text{e}$ | 1.38 |
| $\text{Zn} + 4(\text{CN})^- \rightarrow \text{Zn}(\text{CN})_4^{2-} + 2\text{e}$ | 1.19 |
| $\text{Fe} \rightarrow \text{Fe}^{2+} + 2\text{e}$ | 0.93 |
| $\text{Cu} \rightarrow \text{Cu}^{2+} + 2\text{e}$ | 0.879 |

3.2. Electrolytes

Early thermocell research was primarily carried out on redox couples in aqueous electrolytes. High power output in aqueous ferri/ferrocyanide cells was attributed to the fast diffusion of ions due to the low viscosity of water [34]. As the low boiling point of water limits these cells to operating below 100°C , research has been carried out on a range of ionic liquids, organic solvents for extending the thermal operating range and for water-insoluble or other redox couples with stability issues [67]. Ionic liquids are generally characterized by high boiling points, low vapor pressure, high ionic conductivity and low thermal conductivity, which in turn could lead to high Seebeck coefficients

through higher entropy changes [68]. Detailed data on several redox couples and ionic liquids, TFSA[−] (bis(trifluoromethylsulfonyl)amide, EMI⁺ (1-ethyl-3-methylimidazolium), PP13⁺ (1-methyl-1-propylpiperidinium, BMP⁺ (1-butyl-1-methylpyrrolidinium) and their blends, have been provided by these authors.

Redox reaction entropies in ionic liquids were found to depend strongly on Coulombic interactions between electrolyte ions and the redox couple. Abraham et al. [33] investigated the behaviour of an iodide/triiodide (I[−]/I₃[−]) redox couple in 1-ethyl-3-methylimidazolium tetrafluoroborate; the temperature of the hot electrode was 130 °C. A Seebeck coefficient of 0.23 mV/K and power output of 29 mW/m² were reported. Jiao et al. [69] have reported on the use of [Co(bpy)]^{+3/+2} redox couples in several ionic liquids (cation: 1-ethyl-3-methylimidazolium; anions: bis(trifluoromethylsulfonyl)imide, tetracyanoborate, tris(pentafluoroethyl) trifluorophosphate etc.). The TEC response was found to depend linearly on ΔT (up to 140 °C) with Seebeck coefficients in the range 1.44 to 1.88 mV/K. Due to the generally high viscosity of ionic liquids, mass transport becomes a power-limiting factor, leading to slower rates of diffusion for redox ions. Kim et al. [53] have reported some of the highest Seebeck coefficients (2.9 mV/K) in mixed organic solvent electrolytes (20% methanol in water). The mass transport in electrolyte can also be improved by the addition of other species; the addition of MWCNTs to imidazolium-based ionic liquids lowered the ohmic resistance of the solution due to increased dissociation of ionic pairs and the formation of percolated networks [70]. However, the addition of CNTs to electrolytes containing Co^{2+/3+} (bpy)₃ in [C2mim][NTf2] (see Dupont et al. [26] for nomenclature) showed reduced power with increases in viscosity outweighing the contributions from increases in conductivity [71]. Lithium-based ionic liquids, lithium bis(trifluoromethylsulfonyl)imide (Li[NTf2]) tetraglyme and a thiolate/disulfide redox organic couple in a mixture with 1-ethyl-3-methylimidazolium tetrafluoroborate/acetonitrile have also been tested to produce Seebeck coefficients of 1.4 and 0.6 mV/K, respectively [50,57].

While liquid electrolytes ensure fast ion diffusion, mass transport and high power density, possible leakage and low mechanical strength limit their application for wearable or flexible thermocells. Several quasi-solid-state electrolytes, as blends of polymeric solids with liquids, have been fabricated using a number of gelling agents such as polyvinyl alcohol, agar, poly(sodium acrylate) beads and cellulose [72]. Jin et al. [73] have reported on a quasi-solid-state electrolyte containing a cellulose-polymer matrix and a redox-active (ferri/ferrocyanide) aqueous phase. Organic gel electrolytes show excellent performance due to lower electronic conductivity, high ionic conductivity and good interfacial characteristics [74]. Yang et al. [75] fabricated two types of thermogalvanic gel electrolytes using two redox couples, aqueous ferri/ferrocyanide and ferric/ferrous chloride, with negative and positive Seebeck coefficients (−1.21 and 1.02 mV/K, respectively) gelled together using polyvinyl alcohol. This electrolyte was sandwiched between two polyamide electrodes and was connected in series to generate a larger potential difference; an output voltage up to 1 V could be generated in this system utilizing body heat.

4. Electrode Materials and Designs

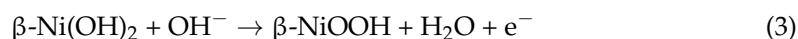
Due to their non-reactive and thermally stable nature, platinum electrodes were used during early studies on thermocells. The electrolyte had to be stirred continuously to achieve Carnot efficiencies of 1.2% [76]. High power generation in thermocells requires large current densities, which may be achieved through high concentrations of redox mediators, increasing the number of sites for redox reactions and exposing electrodes to large thermal gradients [37]. Different types of thermocell electrodes and materials are detailed next.

Reversible metal electrodes: Thermocells using ‘reversible electrodes’ consist of soluble metal electrodes placed in solutions of corresponding salts at two different temperatures (Figure 1c). This cell can only work until a certain level of electrode consumption is reached; the direction of the temperature gradient is then reversed for the cell to work in the opposite direction. Most of the studies have reported on the Cu²⁺/Cu system due to its simple

design and good reproducibility. Seebeck coefficients for this system could be modified from hypothetical values of 0.879 mV/K to 0.75–1.32 mV/K by changing experimental conditions and electrolyte additives [77]. Natural convection was found to have a significant influence on the efficiency of Cu/Cu²⁺-based thermogalvanic cells. Burmistrov et al. [78] have reported on the efficiency of thermocells based on copper, zinc and nickel metallic electrodes. While the efficiency parameters for Cu electrodes agreed well with the theoretical data and published results, the efficiency of zinc-based TEC cells was somewhat lower than theoretical expectations. The thermogalvanic system with nickel electrodes in an aqueous solution of nickel sulphate exhibited an unusual behaviour. The dependence of open-circuit voltage on temperature gradient exhibited an inflection point, with a Seebeck coefficient of 1.05 mV/K for $\Delta T < 30$ °C and a Seebeck coefficient of 2.83 mV/K for $\Delta T = 30$ –60 °C. This result indicates the need for further research in the field towards improving thermocell efficiency.

Inert carbon electrodes: Carbon-based electrodes are becoming increasingly important as a promising and affordable alternative to platinum electrodes. Nanostructured carbon materials, such as single (SWNT) and multiwalled carbon nanotubes (MWNT) and graphene, have a large surface area, which helps in increasing the number of reaction sites. Depending on the number of nanotube walls, the specific surface area of CNTs lies in the range 50–1315 m²/g [79]. These materials also show fast electron-transfer kinetics for the ferrite/ferrocyanide redox couple. Both of these properties can increase the current density achieved with the thermocouple [80]. Most research in this area has been focused on composite materials involving nanostructured materials and additional treatment with additives or alloying [35]. The internal resistance of these electrodes fluctuates due to the porous structure; the highest power density (6.8 W/kg at a temperature difference of 20 °C) has been achieved by electrodes based on purified single-layer carbon nanotubes [37]. Similarly, carbon-nanotube (CNT) aerogel electrodes provide at their highest an efficiency of 3.95% in an aqueous ferri/ferrocyanide system. Hu et al. [81] have reported on a thermocell using 0.4 M ferri/ferrocyanide electrolyte, multiwalled nanotubes (MWNT) electrodes (10 nm dia., 0.5 cm² area, 5 cm apart) and ΔT (61 °C), achieving an aerial power density of 1.36 W/m². Composites of SWNTs with reduced graphene oxide in a 9:1 proportion, 0.45 µm thick, were able to generate an aerial power density of 0.46 W/m².

Hollow nickel microsphere electrodes: Burmistrov et al. [82] have shown that hollow Ni microspheres can be an effective electrode material for thermoelectrochemical cells and provide extremely high values of the hypothetical Seebeck coefficient and open circuit voltage. Electrodes were prepared by pressing tablets of nickel microspheres and reduced at 250 °C. The most effective composition, a KOH-based alkaline electrolyte, was chosen based on the influence of the alkali content on the hypothetical Seebeck coefficient value and specific power as determined by the number of charge carriers in the electrolyte and the intensity of reactions at the electrodes [83,84]. The basic electrode process in this system is described as



This reaction proceeds on the hot electrode followed by electron release, resulting in the formation of a potential difference between the hot and cold electrodes (Figure 2). The accumulation of excess charge or heating on the nickel-oxide electrode results in the side reaction of $\beta\text{-NiOOH}$ transforming into $\gamma\text{-NiOOH}$; the unstable $\gamma\text{-NiOOH}$ gets hydrolysed to $\alpha\text{-Ni(OH)}_2$, which later transforms into $\beta\text{-Ni(OH)}_2$. These reactions can lead to the absorption of electrons at the hot electrode and a lower thermocell potential. The Seebeck coefficient of these thermocells was found to reach 4.5 mV/K (ΔT up to 35 °C), one of the highest values reported for aqueous electrolytes.

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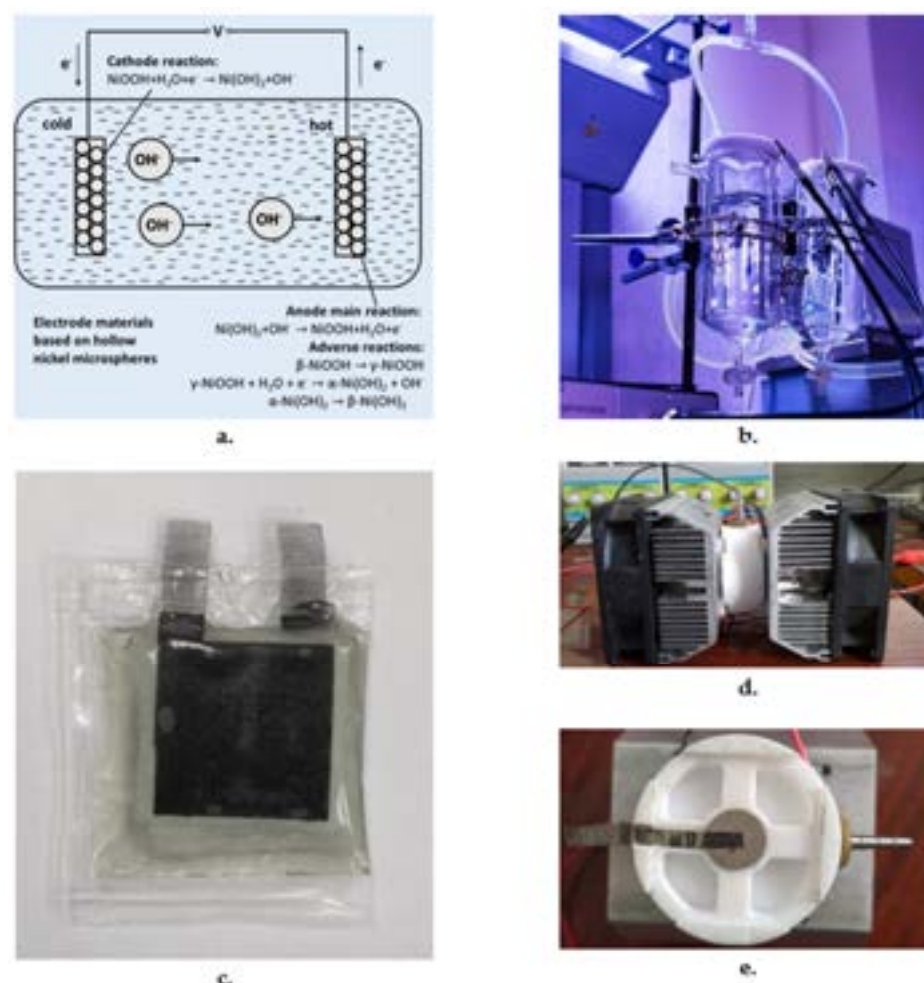


Figure 2. (a) A schematic representation of Ni(OH)_2 redox reactions mediated by Ni hollow microspheres, (b) and assembled cells with water heating/cooling and salt bridge, (c) polymer film body, (d) cell with adjustable distance between electrodes heating/cooling by Peltier elements, (e) structure view of the adjustable cell.

Additional electrode designs: The flexible nature of nano-carbons has led to new thermocell designs suitable for wrapping around curved or irregularly shaped surfaces. The thermocell design is suitable for wrapping around curved or irregularly shaped surfaces such as a vehicle exhaust pipe to support cooling/heating lines in power plants and a distance of 2 mm, these a Carnot engine. This increased the relative efficiency of the cell threefold to 1.4% relative to a Carnot engine. The thermocells have been developed using chemical vapor deposition by producing a catalyst with layers of Ti (30 nm), Al (10 nm) and Fe (2 nm) and using it to grow 100 μm tall MWNT forests on the thermocell casing. Nomex HT 4848 separators impregnated with 0.4 M ferri/ferrocyanide solution kept the electrodes apart; an aerial power density of 0.98 W/m² was obtained for ΔT of 60 °C [30]. A stacked electrode configuration using SWNT and rGO (reduced graphene oxide) (in 9:1 proportion) consisting of layers of stainless-steel mesh as separators between 4.5 μm electrode films maintained a conductive path between individual films. A 10-stack configuration attained an efficiency of 2.63% relative to a Carnot engine. Marquardt et al. [85] have reported on a thermocell based on a proton-exchange membrane with hydrogen electrodes; the maximum open-circuit voltage with a power density of 45.3 mW/cm² was observed at a humidifier temperature of 323 K. A Seebeck coefficient of 1.75 mV/K was observed for ΔT of 35 °C.

5. Emerging Applications

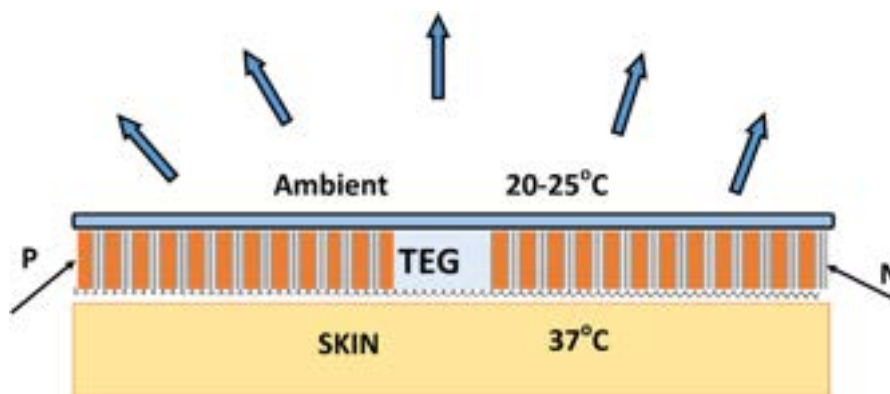


Figure 3. A schematic representation of a TEG wristband for harvesting body heat.

In rigid TECs, n- and p-legs and interconnects are typically affixed to the substrate by conductive wire or substrate (plate) (aluminum/alumina) or flexible (TECs) (gallium). In interdigital devices, however, the base film is a flexible metal or metal interconnects to a certain interdigital configuration. Koppert et al. [96] reported a p-type thin-film thermoelectric material (Bi₂Te) on a flexible substrate (polyimide) and a p-type thin-film thermoelectric material (Bi₂Te) on a flexible substrate (polyimide) [95]. Typical power output range of 0.1–0.2 mW/cm² [95]. Liu et al. [97] have presented several different designs for wearable devices for harvesting body heat in a range of applications.

Organic materials have also been investigated as flexible TEGs due to lower prices, light weight, low thermal conductivity and high flexibility. While significant progress has been made, high-performance organic TEGs still lag behind inorganic chalcogenide BiTe-based alloys. One of the strategies is to fabricate flexible TE devices from composites of organic and polymeric materials with micro/nanomaterials [98]. The TE properties of sev-

eral p-type and n-type coordination polymers have been tabulated by Masoumi et al. [98]. π -conjugated conducting polymers are another group of organic TEs, with a capacity for doping with a wide variety of elements and adjustable doping levels [95]. Some of key conducting polymers include polyacetylene, polyvinylidene fluoride, polyphenylenevinylene, poly(3-hexylthiophene) and polypyrrole. A power factor of $401 \mu\text{W}/\text{mK}^2$ and a Seebeck coefficient of $43.5 \mu\text{V}/\text{K}$ has been achieved with some conducting polymers [99]. Other TE materials include carbon nanotubes, graphene, fullerenes, carbon nanodots, small molecules and organic composites with inorganic fillers. [94].

Energy storage devices: The integration of energy-harvesting and storage devices has been extensively investigated for emerging self-powered electronic devices [100]. Thermoelectric generators can be used to convert excess heat generated during the operation of electronic devices into electricity [101]. Using the Soret effect, ionic thermoelectric supercapacitors utilize ionic electrolytes to produce charges and energy storage in a single device; the operation of these devices is, however, limited by long charging and discharging times and a rigid configuration [102,103]. Yang et al. [104] have reported on a TEG device with n- and p- type modules consisting of Ag_2Te - and Ag_2Se -nanoparticle thin films. This device was directly linked with a planar micro supercapacitor. A Seebeck voltage of 82 mV was generated for ΔT of 15.8 K and a charging efficiency of 98% . Wu et al. [105] have reported on a novel conjugated conducting polymer (PDAQ-BC) [DAQ: 2,6-diaminoanthraquinone; DBC: 3,6-dibromo-9-(4-bromophenyl)carbazole], which showed a specific capacitance of 180.5 F/g for a current of 1 A/g . This polymer retained up to 95% coulombic efficiency and 85% capacitance after 5000 cycles of operation.

Park et al. [106] have reported an all-in-one energy system consisting of a TEG on one side of the substrate and a micro supercapacitor (MSC) on the other side. The TEG was constructed from screen-printed p- and n- modules and a p- type TE film for alignment with electrodes. An MSC was fabricated on the other side using rGO/CNT electrodes and Au current collectors. Electrodes were positioned above and below the TEG-substrate-MSC system; the electrode on the TEG side was connected externally to the current collector of MSC. This system was able to generate 10.8 V of electrical energy for thermal differences up to 10 K and store it without loss.

Liu et al. [102,107] have reported on coupling thermoelectricity and electrocatalysis for hydrogen production via a $\text{PbTe-PbS}/\text{TiO}_2$ heterojunction involving a cathode, an anode and an electrolyte operating at the hot end and in situ endothermic production of electrochemical hydrogen on the cold end. At 70°C and a bath voltage of 1.0 V , hydrogen was generated at a rate of $6.1 \text{ mL}/\text{cm}^2/\text{h}$ with energy and heat efficiencies reaching 88% and 49.9% , respectively. Liu et al. [108] have reported on an advanced Li ion battery with charging based on a coupled thermoelectric approach. Richards et al. [109] have explored solid-state electrochemical heat engines (EHE) for generating electric power from available thermal energy based on reversible redox reactions. EHEs control and utilize the electrochemical potential of molecules undergoing redox reactions with temperature, composition and pressure playing an important role.

Liao et al. [110] have reported on a hybrid active/passive battery thermal management system in combination with thermoelectric elements (TEE) and phase-change materials for the management of Li ion batteries operating in extreme environments. TEEs were used to provide refrigeration at high temperatures and heating to preheat the batteries in cold environments. Nguyen and Shabani [111] have discussed capturing the heat produced by proton-exchange-membrane fuel cells, heat recovery solutions and opportunities for integration with TEGs and thermally regenerative electrochemical cycles for power cogeneration applications.

6. Conclusions and Future Perspectives

The development of thermoelectrochemical technologies for harvesting low-temperature waste heat opens the prospect of increasing the efficiency of various devices and mechanisms operating in exothermic mode or creating systems for generating electricity based on

natural heat sources. Any process that can partially replace fossil fuels as a prime energy source will be used only if it is attractive to industry; an alternative energy source such as waste-heat conversion (WHC) has to be cost effective to become commercially viable. Geoffroy et al. [112] have shown that current WHC heat engines are not economically viable below 100 °C and require temperatures above 150 °C coupled with 100–1000 kW power outputs to be economically competitive. Studies in recent years have shown the possibility of significant increases in the power and conversion efficiency of TEC cells. The highest values of output power and cell potentials have been achieved for the redox ferri/ferrocyanide system and $\text{Co}^{2+}/^{3+}$, which offers great opportunities for further development and research in both aqueous and non-aqueous solvents.

Our results show the pathways to overcome the key fundamental limitations of thermocell performance and set new tasks for fundamental research and the further development of electrodes, electrolyte materials and cell design. One of the key tasks in thermocell development is to investigate mechanisms of entropy change via new redox couples and electrolytes. This will be related to increasing the hypothetical Seebeck coefficient, as well as improving the properties of electrodes and solvents to increase the mass transfer rate (diffusion capacity) towards increasing exchange currents and output power values.

One of the most promising thermoelectric power-generation applications involves using vehicle waste-heat recovery to improve fuel economy, wherein waste heat from the exhaust is redirected to produce electricity. Other applications include harvesting industrial waste heat (from incinerators, cement, steel mills, etc.), or combining geothermal power generation, fuel-oil-fired furnaces or gas water heaters with this technology. Despite extensive research, WHC technology has yet to achieve significant market penetration. For WHCs to become a serious contender, they have to compete with solar, wind and geothermal technologies in terms of capacity factors, capital costs and operational as well as maintenance costs. Significant developments are therefore required on multiple fronts to achieve greater power density performances, especially at higher temperatures. It is possible that TECs might have major advantages over solar cells and semiconductor thermoelectrics, in particular in the Wh/dollar efficiency [81]. Achieving industrial production volumes of MWCNTs and reducing their cost, we believe, can make TECs competitive devices.

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