



## Multi-cell thermogalvanic systems for harvesting energy from cyclic temperature changes



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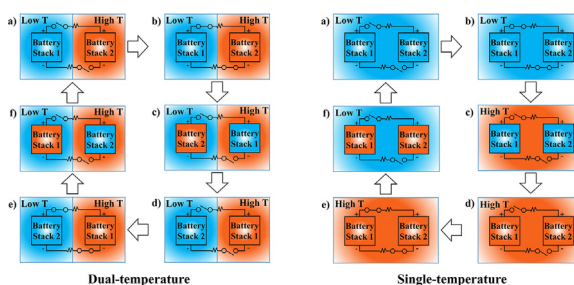
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### HIGHLIGHTS

- New schemes for harvesting energy from cyclic temperature changes are demonstrated.
- Two cells alternately cycled between two temperatures provide high-efficiency conversion.
- Use of two stacks of cells cycled together between two temperatures is also demonstrated.
- Both schemes provide flexibility for materials selection.
- Optimized materials selection is expected to improve conversion efficiency.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

**Keywords:**  
Thermal energy harvesting  
Batteries  
Thermogalvanic  
Temperature cycle  
Lithium-ion  
Charge-free

### ABSTRACT

Two types of electrochemical systems for harvesting energy from cyclic changes in environmental temperature using the thermogalvanic effect are demonstrated. Both systems are based on two battery stacks which function in either a dual-temperature or single-temperature configuration. In the dual-temperature configuration, two LiCoO<sub>2</sub>/Li cells were separately cycled between two temperatures to achieve an energy conversion with an efficiency of 0.22%, free of heat recuperation, and a peak output power of 0.4 μW when cycled between 20 °C and 50 °C. This energy conversion efficiency is comparable to other proposed systems for thermal energy harvesting such as charge-free thermally regenerative electrochemical cycle (TREC) devices, thermocapacitive devices and ionic thermoelectric supercapacitors. However, this system can function with a broader range of materials, which will likely allow further increases in efficiency and power. A single-temperature configuration in which two stacks of cells were cycled together between two temperatures was also demonstrated. Both systems are attractive for harvesting thermal energy for self-powered sensor networks, especially in remote areas.

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## 1. Introduction

Technologies for the Internet of Things (IoT) are being developed for a vast number of networking applications, monitoring many aspects of life and environmental conditions [1]. An IoT network consists of large quantities of networked sensors that are often in remote or difficult to access locations, which drives the need for self-powered systems. Research on self-powered systems harvesting mechanical energy [2–5], solar energy [6] and other energies [7] has been explored. Among these energy sources, low-grade thermal energy ( $< 100\text{ }^{\circ}\text{C}$ ) has the advantage that it is ubiquitous, which makes it a good candidate energy source for self-powered systems. Previous research on thermal-electrical energy conversion has focused on three types of devices: thermoelectric devices, thermogalvanic devices and thermally regenerative electrochemical cycle (TREC) devices.

Thermoelectric devices take advantage of the Seebeck effect, in which an electromotive force is produced by a spatial temperature gradient [8–14]. So far, the figure of merit of thermoelectric materials is limited to 1.5 below  $100\text{ }^{\circ}\text{C}$  [15], which corresponds to an absolute energy conversion efficiency of 2.1% if the cold side is at  $20\text{ }^{\circ}\text{C}$  and the hot side is at  $50\text{ }^{\circ}\text{C}$ .

Thermogalvanic devices have similar architectures as thermoelectric devices; they utilize the thermogalvanic effect and a change in temperature causes a change in a redox couple's electrochemical potential [16–21]. If a material serves as an anode and cathode in an electrochemical cell and these electrodes are held at different temperatures, the resulting thermogalvanic voltage can drive an ionic current in an electrolyte and an external electrical current. Although thermoelectric and thermogalvanic devices generally have similar temperature coefficients  $\alpha$ , poor ionic conductivity through the electrolyte causes the energy conversion efficiency,  $\eta$ , of thermogalvanic devices to be lower than thermoelectric devices. Here the temperature coefficient is defined as the output voltage when the temperature difference across the device is one degree C. In addition to thermoelectric and thermogalvanic devices, there are also other devices that can convert thermal energy into electricity, such as alkali metal thermal electrochemical converters [22].

Both thermoelectric and thermogalvanic devices must be used within temperature gradients; they cannot take advantage of cycles between spatially uniform temperatures. While temperature gradients are useful for converting energy from continuous waste heat streams, many other thermal energy sources are intermittent, such as diurnal cycles (alternating night and day), banks of generators (some switch on and off depending on loads), revolving doors, hot water pipes, electrical heaters and air conditioning ducts. Additionally, the requirement to maintain a gradient across a cell requires heat sinks and makes decreasing the size of any system difficult, and practically impossible if moving to system sizes below a few millimeters. Given these limitations, work to discover practical uses of the thermogalvanic effect for energy harvesting from temperature cycles has begun.

Thermally regenerative electrochemical cycle (TREC) devices use temperature cycles to generate electricity [23]. Recent work has explored the potential these systems hold [24–26]. Lee et al. demonstrated an energy conversion efficiency (defined as the total energy output per cycle divided by the total amount of heat absorbed by the system per cycle) of 3.7% for cycling between  $10\text{ }^{\circ}\text{C}$  and  $60\text{ }^{\circ}\text{C}$ , with no heat recuperation, using a TREC device with CuHCF as the positive electrode and Cu/Cu<sup>2+</sup> as the negative electrode [24–26]. Yang et al. demonstrated a TREC device with a NiHCF cathode and an Ag/AgCl anode and reported an energy conversion efficiency of 1.6% between  $15\text{ }^{\circ}\text{C}$  and  $55\text{ }^{\circ}\text{C}$ , with no heat recuperation [25]. Hartel et al. applied the TREC approach to supercapacitors, which gave an energy conversion efficiency of 0.38% between  $0\text{ }^{\circ}\text{C}$  and  $65\text{ }^{\circ}\text{C}$ , with no heat recuperation [26]. However, a conventional TREC-based system requires an external charge source to complete the cycle, which prevents its application in self-powered systems. Yang et al. further developed self-

powered TREC-based systems with electrode materials whose potentials cross in the middle of the temperature range [27]. In other words, at low temperatures ( $20\text{ }^{\circ}\text{C}$ ), the potential of electrode #1 (ferri/ferrocyanide) is higher than the potential of electrode #2 (Prussian blue), while at high temperatures ( $60\text{ }^{\circ}\text{C}$ ), the potential of electrode #2 is higher than the potential of electrode #1. This specific combination of electrode materials allows the system to spontaneously discharge at both temperatures of the cycle. This system offered an energy conversion efficiency of 0.68% between  $20\text{ }^{\circ}\text{C}$  and  $60\text{ }^{\circ}\text{C}$  with no heat recuperation. However, unlike a conventional TREC cell that only requires different temperature coefficients, self-powered systems also require that the electrodes have similar electrochemical potentials. Because of this additional constraint, the electrode materials require a more careful selection process.

In this paper we report two types of dual-stack self-powered electrochemical systems that can generate electrical power through temperature cycles. In the first, the dual-temperature system, two temperature environments are required at any given time around the system. The second, the single-temperature system, only requires a single temperature environment around the system to cycle.

## 2. Experimental procedures

### 2.1. Principles of operation

The dual-temperature dual-stack self-powered electrochemical system is depicted in Fig. 1. Two stacks, connected electrically but operating in different temperature environments, can convert thermal energy into electrical work. This system uses two identical electrochemical stacks. Here we use “stack” to refer to not only multiple batteries connected in series, but also a single battery, to keep generality. However, each electrochemical stack is held at a different temperature. At the start of the cycle (Fig. 1a), the voltage between the stacks is the product of the temperature coefficient of the full stack and the temperature difference. When the switches are closed (Fig. 1b), current flows through the load from battery stack 1 to battery stack 2 and causes a change in the state of charge in each of the electrodes until the cells reach equilibrium. After that, the temperatures around the two stacks are changed (Fig. 1c) and held until thermal equilibrium is reached (Fig. 1d). Due to the thermogalvanic effect, a voltage will again develop within the circuit so that when the switches are closed, current will flow from battery stack 2 to battery stack 1 (Fig. 1e) until equilibrium is again achieved. Finally, the two stacks are returned to their original position (Fig. 1f) and the system also reaches its original state. A schematic showing the voltages of the stacks and the difference in potential across the load is provided in Fig. 1g.

The single-temperature system works similarly, with the electrochemical stacks having similar operating potentials but oppositely signed temperature coefficients. Its operation is illustrated in Fig. 2. At the start of the cycle (Fig. 2a), a potential difference exists between the two battery stacks. When the switches are closed (Fig. 2b), current flows through the load from battery stack 1 to battery stack 2 and causes a change in the state of charge in each of the electrodes until the cells reach equilibrium. After that, the entire system is heated (Fig. 2c) and held until thermal equilibrium is reached (Fig. 2d). Due to the different temperature coefficients, a voltage will develop within the circuit so that when the switches are closed, current flows from battery stack 2 to battery stack 1 (Fig. 2e) until equilibrium is reached. Finally the two stacks are cooled down to the original temperature (Fig. 2f) and the system reaches its original state before the cycle. A schematic showing the voltages of the stacks and the difference in potential across the load is provided in Fig. 2g.

The dual-temperature system relies on cells with the greatest absolute value of temperature coefficient. It does not matter whether the temperature coefficient is positive or negative. Since a crossover of potentials between electrode materials is not required, a broader library

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