

A revisit of thermo-electrochemical techniques for low-temperature waste heat recovery

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In recent years, various thermo-electrochemical techniques have been proposed to recover low-temperature waste heat ($<100^{\circ}\text{C}$), including thermogalvanic cell (TGC), thermally regenerative electrochemical cycle (TREC), direct thermal charging cell (DTCC), and thermally regenerative batteries (TRBs). Despite the spring up of innovative concepts and extensive efforts toward performance improvement, the thermo-electrochemical community still face great challenges in advancing any technique to commercialization due to a lack of consistent framework for performance evaluation and big gaps between practical demonstration and laboratory work due to questionable performance when scaling. In this context, we propose multi-level performance evaluation criteria and an integrated design approach for comprehensive development of high-performance practical devices, while also addressing the specific challenges of each technology.

STATE-OF-ART TECHNIQUES

Figure 1A shows the working principle of the state-of-the-art thermo-electrochemical systems. TGC with electrodes at different temperatures generates a potential due to thermogalvanic effect, thermodiffusion effect or a combination of both effects. The highest efficiency of 1.3% ($20\text{--}60^{\circ}\text{C}$) and power density of 17.7 W/m^2 ($20\text{--}70^{\circ}\text{C}$) were reported.¹

TREC leverages a thermogalvanic cycle in isothermal cells to generate thermal potentials, operating in either intermittent (charging/discharging at different temperatures) or continuous charging-free mode. The highest efficiency of 4.1% ($10\text{--}60^{\circ}\text{C}$) was reported with a temperature coefficient of -3 mV/K .²

DTCC is configured with a capacitor-type cathode and a battery-type anode in aqueous electrolyte that can generate voltage from pseudocapacitive effect and thermogalvanic effect. The highest efficiency of 3.59% and power density of 0.68 W/m^2 ($25\text{--}75^{\circ}\text{C}$) were reported with a thermopower of 3.7 mV/K .³

TRB operates isothermally by using chemical potential difference between two metal electrodes, generated through metal-ligand complex formation exclusively in anolyte. An efficiency of 1.9% is achieved, which can be enhanced to 5.3% ($25\text{--}95^{\circ}\text{C}$).⁴

MULTI-LEVEL PERFORMANCE EVALUATION

Thermodynamic performance is typically characterized by three metrics: thermopower, energy efficiency, and power/energy density. Thermopower quantifies the thermal-induced voltage in electrochemical cells. Energy efficiency, the ratio of electric work output to heat input, is often challenging to compare fairly across studies due to differing assumptions in estimating heat input. Power and energy density, expressed per unit mass or area, depend heavily on the chosen reference—such as cathode's active material, both cathode and anode, or the total system mass, and whether projected membrane area or electrode area is used. Additionally, average and peak values must be clearly distinguished to enable meaningful evaluation.

In addition to thermodynamic performance, multiple critical factors must be introduced. By integrating assessments of reliability, complexity, scalability, safety, and economic viability, this study proposes a six-level evaluation framework (Figure 1B) to enable systematic comparison across different technologies.

Reliability is another critical criterion, as a system with a limited lifetime is impractical regardless of its short-term performance. We propose two complementary metrics for its assessment: cyclability (the number of cycles before performance degrades to a threshold) and lifetime (the total stable operation time). Cyclability alone is insufficient, as a high cycle count with

short cycle duration (e.g., 500 cycles of $<1\text{ min}$ each) yields a short total lifetime. Therefore, a joint evaluation of both metrics is essential for a comprehensive reliability analysis.

System complexity is frequently overlooked in existing literature, yet practical implementation critically depends on size, volume, weight, and compactness. For instance, a device generating 1 kW peak power with 1 W/m^2 power density and 1 mm thickness would require 1 m^3 volume, demonstrating poor compactness. Scaling such systems introduces additional challenges associated to performance degradation and auxiliary components. Non-isothermal configurations present additional constraints, typically requiring either increased electrode spacing or enlarged flow channels. These design requirements inevitably compromise system compactness, resulting in bulkier implementations than idealized laboratory-scale prototypes might suggest.

Scalability represents a critical requirement for transitioning technologies from laboratory prototypes to practical applications. Current limitations of single-cell configurations include insufficient voltage/current output for powering external devices, necessitating series/parallel cell integration. However, performance degradation would occur when scaling single cell with increased cell dimensions or excessive mass-loading and adopting multi-cell arrays, which requires careful characterization and experiment evaluation.

Safety considerations remain critically under-addressed. Aqueous electrolytes exhibit $\sim 4\%$ volume expansion when heated from 10 to 90°C ,⁵ potentially causing electrolyte leakage and accelerated cyclability degradation. It is necessary to employ cell packaging with effective sealing using materials with low heat capacity and matched thermal expansion coefficient. Some current efforts in the pursuit of high energy efficiency have led to unsafe practices including adopting organic solvent, strong acid electrolytes, and toxic polymer components. We advocate for comprehensive safety protocols utilizing stable, non-flammable and non-toxic electrode materials, electrolytes, and separators/membranes.

Economic viability represents a critical yet underexplored dimension for commercializing emerging thermal-electrochemical energy conversion systems. The cost of any system should include capital expenditures of materials and essential components such as heat exchangers, and operation and maintenance costs. Systematic reporting of comprehensive cost estimates is necessary for economic optimization.

SYSTEM DESIGN FRAMEWORK

We propose an integrated system design framework that synergizes top-down (application-driven) and bottom-up (material-centric) approaches to optimize electrochemical energy harvesting systems (Figure 1C). This dual-strategy methodology enables performance-targeted design through thermal source characterization, fundamentally-optimized materials via multiscale modeling and AI-driven discovery, and seamless scalability from nanoscale mechanisms to macroscale implementation.

The design framework begins with a top-down analysis of performance requirements. A thorough heat source assessment is critical to define application-specific needs, including heat carriers, temperature ranges, and accessibility. This evaluation helps researchers identify design constraints for tailored thermo-electrochemical systems, optimize thermal components (e.g., heat exchangers) to minimize complexity, cost, and exergy losses, and determine feasibility of heat-to-electricity conversion based on source characteristics. For dissipation-oriented applications (e.g., solar panels, data centers), rapid heat removal often outweighs energy conversion. This assessment ensures alignment with system objectives.

Afterwards, the thermo-electrochemical system development follows a

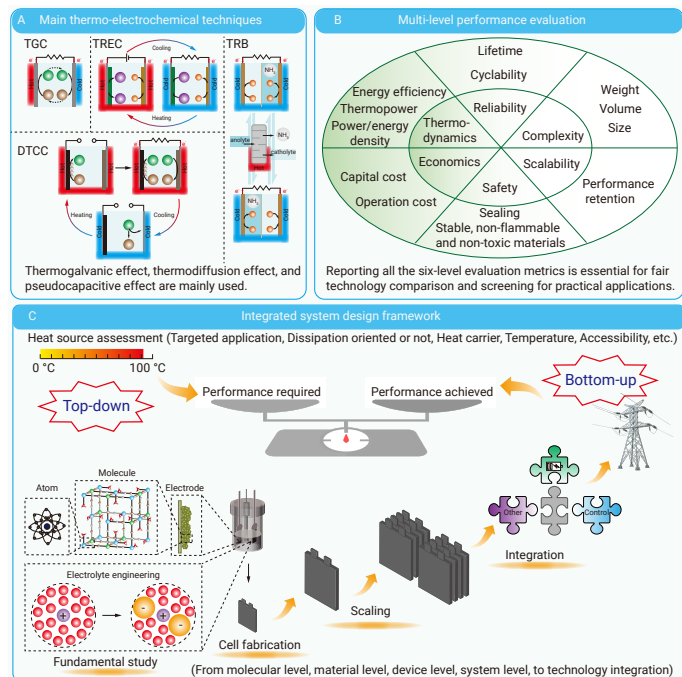


Figure 1. Thermo-electrochemical techniques for low-temperature waste heat recovery (A) Working principle of the state-of-the-art techniques. (B) Multi-level performance evaluation. (C) System design framework.

bottom-up approach through materials development, cell fabrication, and scaling. For materials development, the key focus is to establish structure-property relationships by addressing the correlation between materials structures and reaction entropy change and the influences of active sites (vacancies, functional groups, or dopants) on the charge transfer. A combined methodology is suggested by trial-and-error experimental tests, in-situ characterization, multiscale modeling (molecular dynamics and density functional theory), and machine learning assisted innovative design. For cell fabrication and scaling, the integration of optimized materials into functional devices and systematic scale-up protocols are required.

For each technique, the remaining opportunities are summarized below.

TGC suffers from low energy efficiency due to the interdependence of thermopower (α), thermal conductivity (k_{eff}), and electrical conductivity (σ_{eff}). Recent advances demonstrate that thermal-sensitive crystallization can enhance α and suppress k_{eff} without sacrificing σ_{eff} .¹ Further improvements require coordinated electrode design (guided by atomic-scale reaction kinetics), electrolyte engineering (developing redox couples with high α , solubility, and optimal thermomdiffusion properties), and advanced configuration optimization (e.g., adopting thermal separators with 3D-printed ordered porous structures). Machine learning-assisted optimization may be integrated with these approaches to enable effective design of materials and configuration to significantly boost TGC performance for practical applications.

For TREC, three critical issues need to be addressed for high efficiency and power density: (1) large sensible heat of cells. Developing materials with low heat capacity and implementing heat recuperation are recommended; (2) large hysteresis. Reducing hysteresis is necessary to reduce energy loss. We recommend integrating fast-kinetic electrodes, low-resistance membranes, highly conductive electrolytes, and low-internal resistance configurations; and

(3) intermittent power generation. Redox-flow battery based TREC sacrifices energy efficiency. More advanced solutions need to be explored to address this challenge.

TRBs face two main challenges of low efficiency and inferior durability. Key research priorities include innovating regeneration processes to improve efficiency, fundamental studies to overcome electrode irreversibility (especially anode mass loss), and essential experimental validation—moving beyond simulation-based studies—using cycled electrolytes rather than fresh solutions to obtain realistic performance data and accurately assess performance and longevity.

DTCC has limitations on power density and energy density due to low current density and high hysteresis. Developing anode with low onset potential may facilitate spontaneous discharging. Exploring materials with high temperature coefficient and low hysteresis to reduce voltage drop at the beginning of discharging is essential for high energy output. Besides, the stability of materials is also important for cell durability. Current research reported graphene oxide as cathode, which however has cyclability issue due to reduction of functional groups. Similar to TREC, the efficiency can be improved by reducing cell heat capacity and introducing heat recuperation.

Besides, new high-performance approaches are expected. System integration with other techniques (e.g., energy storage) has great potential for applications where heat dissipation is not required—including exhaust pipes, human body, and sunlight.

Overall, it is believed that there is a big challenge for low-temperature waste heat recovery, but there is also a substantial market if cost-effective techniques can be developed. Multidisciplinary studies across material science, chemistry, chemical engineering, mechanical engineering, and computational science will lead to more innovative systems for technology advancement in this field in the future.

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DECLARATION OF INTERESTS

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