https://doi.org/10.3311/PPme.43893

Periodica Polytechnica Mechanical Engineering

Dynamic Thermally Regenerative Electrochemical Cycle System Concentration Tracking for Efficient Control

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Received: 04 March 2026, Accepted: 24 March 2026, Published online: 22 April 2026

Abstract

Particularly in the transportation sector, waste heat originating from ultra-low-temperature sources (25–80 °C) represents a significant but largely untapped energy resource. With conventional heat recovery solutions, this type of heat loss is either not or only poorly exploitable. Therefore, thermally regenerative electrochemical cycles offer a promising solution, as they can directly convert low-temperature thermal energy into electricity. In this research, the dynamic behavior of a system using iron- and iodine-based redox pairs was investigated. In the model, the temporal variations of reactant concentrations were simulated over a 24-hour period, along with the electromotive force. Based on our results, the system's electromotive force closely follows the concentration changes, which means that the regeneration process can be effectively controlled solely based on voltage. This enables the optimization of pump operation using voltage as the input parameter for pump control, avoiding unnecessary pump cycling. The method may be particularly promising for dynamic applications in vehicles, where concentration measurement is difficult to implement.

Keywords

TREC, waste heat, concentration, electricity

1 Introduction

Waste heat originating from heating systems represents a substantial and often underutilized source of global thermal pollution [1]. In the mobility sector, two major contributors can be identified: the waste heat associated with the manufacturing of structural materials and components [2], and the thermal losses related to the production and operational use of energy carriers and auxiliary materials [3]. According to published data, more than 96% of the waste heat generated within the European Union lies in the temperature range between 25 °C and 80 °C, which is classified as "ultra-low-grade waste heat" [4]. Conventional heat recovery technologies, such as Organic Rankine Cycle (ORC) systems [5] and absorption cooling systems [6], achieve high efficiencies primarily at temperatures above 80 °C. However, in transportation applications, numerous heat sources operate below 80–100 °C, including batteries [7], engine cooling circuits [8], fuel cells, and metallic structural components. Consequently, there is a clear need for heat recovery solutions capable of converting sub-80 °C thermal energy into electrical power with minimal additional heat losses.

One promising approach is the thermally regenerative electrochemical cycle (TREC) [9]. A typical TREC configuration consists of two electrochemical cells: a high-temperature (hot) cell and a low-temperature (cold) cell [10]. The hot cell is maintained at elevated temperature by an external heat source, while the cold cell is cooled either actively or by exposure to a lower-temperature environment [11]. The temperature difference between the two cells establishes a thermal gradient. Because electrochemical reaction equilibria are temperature dependent, the two cells reach distinct equilibrium states. This difference in chemical potential drives the system toward equilibrium through coupled electron and ion transport. Electrons flow through the external circuit—following the path of lowest resistance—thereby generating electric current, while ion migration within the electrolyte enables electrode regeneration during charge–discharge processes. The electrical energy produced is harvested through an external load connected between the electrodes. A key limitation of TREC systems is the inherently slow natural diffusion process responsible for regenerating concentration differences [9]. To enhance regeneration,

Cite this article as: Lévai, E., Bereczky, Á. "Dynamic Thermally Regenerative Electrochemical Cycle System Concentration Tracking for Efficient Control", Periodica Polytechnica Mechanical Engineering, 2026. https://doi.org/10.3311/PPme.43893

pumps are typically installed between the hot and cold compartments to circulate the electrolyte; however, their power consumption reduces the overall system efficiency [12]. In most implementations, these pumps operate continuously, regardless of whether the concentration gradient is already sufficient to sustain the desired electrical output. As a result, energy is often expended unnecessarily.

A more efficient strategy would involve regulating pump operation according to the actual concentration state of the system. In this research, we propose a control methodology for TREC heat exchangers operating under dynamic load conditions—such as those encountered in vehicular applications—where pump actuation is governed exclusively by the output voltage signal. This approach eliminates the need for separate concentration measurements while enabling adaptive and energy-efficient system control. The system not only offers direct savings at the user level, for example to a vehicle operator, but also supports the European Union's energy and climate objectives. According to Article 26 of the Energy Efficiency Directive (EED 2023), efficient district heating systems must progressively integrate renewable energy and waste heat into their energy supply, and an efficient system must include at least 50% renewable energy, 50% waste heat, or 75% cogeneration (CHP) heat (or combinations thereof) by the end of 2027, with higher shares phased in later. In the context of transport, waste heat can originate only from the system itself as the source [13]. Another EU directive promotes the increased adoption of electric and hydrogen-powered vehicles in the coming years [14]. Since these drivetrains operate at lower temperatures, their integration does not reduce the share of waste heat required under the above targets. As a result, the utilization of low grade waste heat is becoming increasingly valuable from the perspective of EU regulations, particularly in the transport sector. Furthermore, the Renewable Energy Directive (RED III) [15] explicitly recognizes waste heat recovery as an eligible contribution toward Member States' renewable energy share in heating and cooling, thereby linking the exploitation of waste heat with broader EU renewable energy targets.

Overall, it can be said that the TREC system supports the European Union's global efforts in the field of transport energy globally. However, this requires that the system is also locally profitable, which will be discussed later in the calculations of the manuscript.

2 Materials and methods

The tests were performed with an iron-complex-based catholyte formed from an aqueous solution of erythritol and erythritol, and an anolyte formed from an aqueous solution of iodine. The concentrations were adjusted according to the experiment of Qian et al. [16] to make the results comparable. It means 1M I_2 + 0.1M KI as catolyte, 0.375 M $K_4Fe(CN)_6$ + $K_3Fe(CN)_6$ as anolyte. The mixtures were prepared using distilled water at room temperature for 3 hours using a magnetic stirrer. The mixtures were prepared with a precision of 0.001 g using a balance. During the simulations, the parameters of the TREC system were set based on Table 1.

The basis of the Nernst equations in the computational model is the Nernst equation, which in this case is in specific form:

$$E = E^0 + \frac{RT}{zF} \ln \frac{[\text{oxidized}]}{[\text{reduced}]} \tag{1}$$

specifically:

$$E_I = E^0_{I_3} / I^- + \frac{RT}{zF} \ln \frac{[I_3^-]}{[I^-]^3} \tag{2}$$

$$E_{Fe} = E^0_{Fe(CN)_6^{3-}} / Fe(CN)_6^{4-} + \frac{RT}{zF} \ln \frac{[Fe(CN)_6^{3-}]}{[Fe(CN)_6^{4-}]}. \tag{3}$$

Table 1 Used terms and values (if fixed)

Title	Note	Value	Unit
Electromotive force (general)	E^0	-	V
Electromotive force (iodinge reaction)	$E^0_{I_3/I^-}$	-	V
Electromotive force (feroocianide - reaction)	$E^0_{Fe(CN)_6^{3-}/Fe(CN)_6^{4-}}$	-	V
Faraday constant	F	96485	C/mol
Universal gas constant	k	8.314	J/mol K
Hot side temperature	T_h	323	K
Cold side temperature	T_c	293	K
Number of electrones	z	1	-
Volume of electrolyte	V	0.25	l
Electric load	R	200	Ω
Concentration	C	-	mol/L
Time	t	-	s
Damköhler number	Da	-	-
Current density	i	-	A/m^2
Cell distance	L	-	m
Transferred electrons in a unit reaction	n	-	-
Diffusion coeffitient	D	-	-
Starting concentration	C_0	-	mol/L

The concentration change can be calculated according to Faraday's law as a function of concentration:

$$\frac{dC}{dt} = -\frac{\mathrm{I}}{zVF} \,. \tag{4}$$

Based on these equations, the time dependence of the concentrations can be written for a system of known size and initial composition, since the process generating the voltage is always chemically identical. In addition, it can be plotted where we are in a concentration space in time, i.e., without indirectly measuring the concentration, we can get an idea of how much reactant we currently have and where locally (on the hot or cold side).

It should be emphasized that, although the TREC system is highly scalable, the results presented in the manuscript refer to diffusion-limited devices. When modeling a TREC system, it is necessary to determine the limiting current at which diffusion at the electrode interface is sufficiently fast, allowing the use of a volumetric model to assess depletion. Beyond this point, the reaction rate exceeds the diffusion rate, and the system requires the assistance of pumps. In this case, instead of a volumetric model, a more complex approach must be considered, incorporating an active boundary layer and an electrochemically less active internal electrode volume.

In practice, for example in vehicles, the heat transfer surface of a fuel cell or a battery pack may reach the geometric limit where this calculation becomes necessary. This calculation also helps determine the method for increasing system performance: if the current remains within the limiting value, the current can be increased by enlarging the surface area of a single cell. However, if the current exceeds this limit, higher voltage—and thus higher power—can only be achieved by adding additional cells in parallel.

The limiting current of the diffusion model is given by the Damköhler equation:

$$\mathrm{Da} = i \cdot L / n \cdot F \cdot D \cdot C_0 \,, \tag{5}$$

where

- $\mathrm{Da} < 1$: diffusion is fast compared to the reaction rate, volumetric used,
- $\mathrm{Da} \cong 1$: the system operates on the limiting-current point,
- $\mathrm{Da} \leq 1$: diffusion is slow relative to the reaction rate, volumetric model is not usable.

If the goal is to shape the TREC module (by increasing the surface area or by parallel connection) so that it is at the diffusion limit, the limiting current can be determined knowing the initial concentration:

$$i_{LIM} = n \cdot F \cdot D \cdot C_0 / L \,\cdot \tag{6}$$

3 Results and discussion

Based on the experimental results, the concentration–time profiles of the iodine and iron complexes are presented in Fig. 1.

The depletion rates of the ions differ significantly. This can be attributed to two main factors: first, the initial concentrations of the species are not identical, and second, the molecular sizes of the catholyte and anolyte solutes differ substantially. Fig. 2 illustrates how the reactant ratio in the system can be monitored, while the scale on the right indicates the corresponding electromotive force (EMF) measured at a given ratio. The total EMF output is defined as the difference between the cold and hot sides and can be measured directly at the TREC terminals. Using the hot-side iron complex as an example, the expected time dependence of the electromotive force (Fig. 2) can be derived from the time-dependent concentration equations (Fig. 1) combined with the concentration dependence of the EMF over the 24-hour measurement period (Fig. 2). Within this concentration-monitoring framework, the instantaneous local concentration can be calculated from the measured cell potential at any given time. The temporal trajectory of this process, based on experimental data, is shown in Fig. 3. It can be seen from the image that the diagram describes a very large time scale for the life cycle, as only the series of points marked in yellow take place during a 24-hour week. This also gives an idea of the stability of the system, as its passive depletion is also slow.

4 Conclusion

The system was tested 24 hours using 0.25 L of electrolyte and a 200 Ω external load, without applying external pumping. During the test period, the concentrations of $[\mathrm{Fe(CN)}_6]^{3-}$ and $[\mathrm{Fe(CN)}_6]^{4-}$ in the TREC system, as well as those of the iodine complexes, were determined from the measured electromotive force (EMF). For both the iodine and iron redox couples, a strong correlation was observed between the EMF and the concentration of the electrochemically active species. This demonstrates that the system state can be reliably monitored solely through voltage measurements. Consequently, direct concentration measurements become unnecessary for subsequent energy-efficient pump control, while the system operation remains concentration-driven. This finding enables simpler and more energy-efficient control strategies

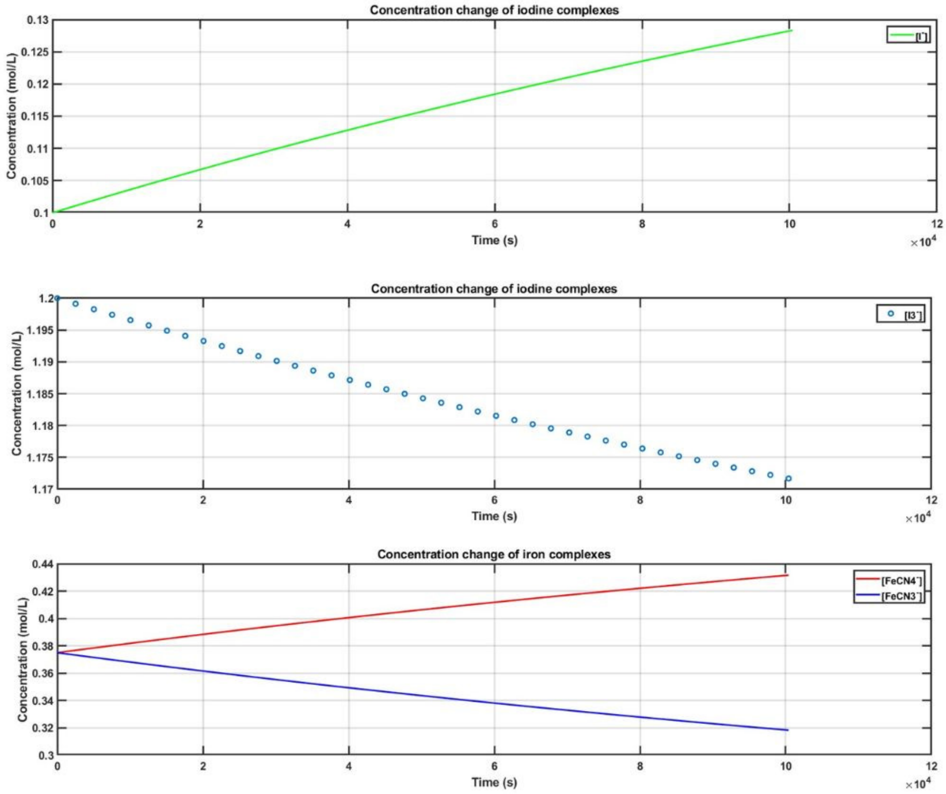
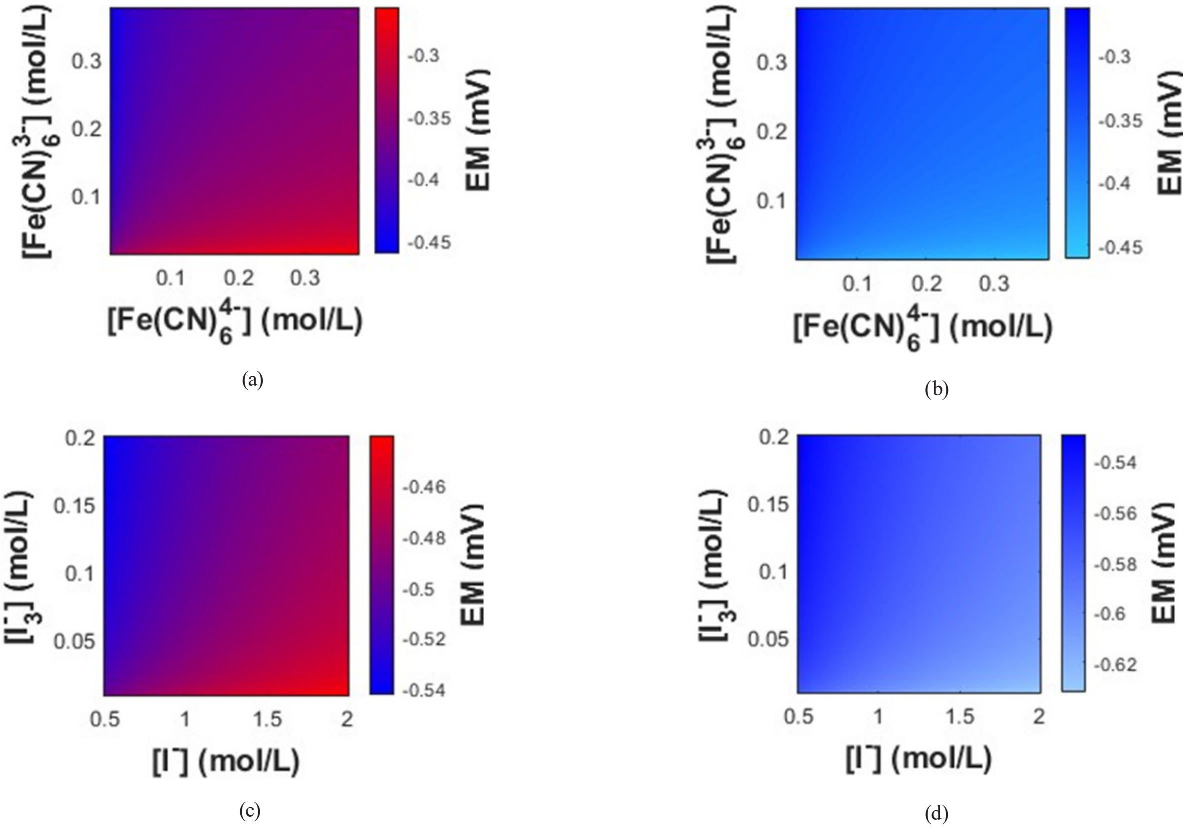


Fig. 1 Time dependence of concentration in case of all species (hot side)



(a)

(b)

(c)

(d)

Fig. 2 Full concentration maps on the hot and cold sides: (a) Hot side of Fe complexes; (b) Cold side of Fe complexes; (c) Hot side of iodine complexes; (d) Cold side of iodine complexes

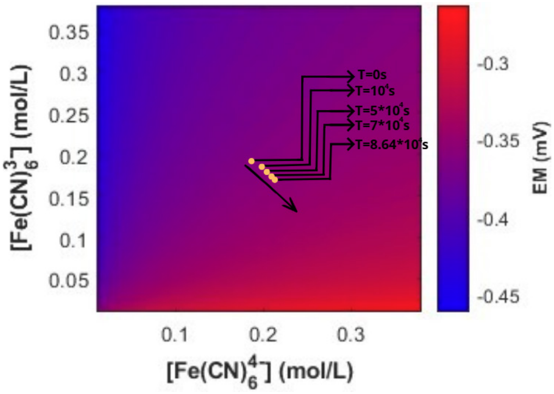


Fig. 3 Electromotive force variation during 24-hour operation on a concentration map (using the example of the Fe complex sample of the hot side)

in future developments. Under laboratory conditions and in the generation of industrial response functions, it is particularly significant that concentration measurement instruments can be replaced by a computational evaluation protocol. This reduces operational costs and minimizes uncertainty. The experimental results can be described analytically using a diffusion-based model, or alternatively, by fitting a predictive model to the measured data. The manuscript also presents a method for calculating the limiting current associated with the diffusion boundary. Although pump control based solely on EMF is feasible, the dynamic response of the system should be considered. Transient voltage fluctuations (e.g., due to load changes) may induce undesired pump oscillations. A detailed dynamic and control analysis is left for future work.

Acknowledgement

Prepared with the professional support of the co-operative doctoral program of the University research scholarship program of the Ministry of Culture and Innovation financed from the National Research, Development and Innovation Fund.

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