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Review Article

Distribution of relaxation times: Foundations, methods, diagnostics, and prognosis for electrochemical systems

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The distribution of relaxation times (DRT) has become an indispensable technique for interpreting electrochemical impedance spectroscopy. This review traces the evolution of DRT from a powerful deconvolution tool for gaining mechanistic insights into a predictive engine for diagnostics and state estimation in fields such as batteries and fuel cells. The technique's intuitive appeal is challenged by its mathematically ill-posed nature, creating a "credibility gap" where subjective choices can yield misleading artifacts. Recent methodological advances in Bayesian and entropy-based frameworks provide greater robustness and uncertainty quantification. The path forward requires establishing a comprehensive analytical ecosystem built on community standards, benchmark datasets, and transparent reporting. This current opinion urges the community to embrace rigor and transform DRT from a specialized, expert-level tool into a reliable and reproducible cornerstone of electrochemical analysis.

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Introduction

Electrochemical impedance spectroscopy (EIS) is a mainstay in electrochemistry for probing charge transfer kinetics and transport [1]. Equivalent-circuit fits are often nonunique, as distinct circuits can fit the same data, and elements may lack a clear physical meaning [2]. The distribution of relaxation times (DRT) offers an alternative by transforming $Z(\omega)$ into a time-constant domain representation $\gamma(\log\tau)$ [3]. In an ideal DRT, distinct processes appear as separate peaks as a function $\log\tau$, where the peak position indicates the characteristic timescale, and the peak area, integrated over $\log\tau$, is proportional to that process's contribution to the total polarization resistance [4]. Because the DRT is agnostic to circuit topology, it yields a compact fingerprint of system dynamics. Its interpretative and visualization power increases when spectra are collected under controlled variations (*e.g.* temperature, state of charge [SOC], and partial pressure), letting correlated peak shifts be mapped to specific mechanisms through coupling with prior work or companion experiments (e.g. thermally activated processes shift to shorter times with increasing temperature [5]).

Despite its intuitive appeal, DRT is not a push-button method. It rests on the basic assumption that a linear, time-invariant electrochemical system responds as a superposition of elementary relaxations. Under this assumption, the impedance and the DRT are related by a Fredholm integral equation of the first kind, which is written as follows[3]

$$Z(\omega) = R_\infty + i\omega L_0 + \frac{1}{i\omega C_0} + \int_{-\infty}^{+\infty} \frac{\gamma(\log\tau)}{1+i\omega\tau} d\log\tau \quad (1)$$

where ω is an angular frequency ($\omega = 2\pi f$ with f being a frequency), R_∞, C_0, and L_0 are the series resistance (*e.g.* electrolyte resistance), capacitance, and inductance, respectively, and i is the imaginary unit. The forward DRT operator is linear in that linear combinations of DRTs map to linear combinations of impedances. Although the forward map is functional and, thus, nonparametric, in practice $\gamma(\log\tau)$, it is most often (but not always) approximated through a fixed and finite representation, namely a basis expansion in $\log\tau$ [6].

Recovering $\gamma(\log\tau)$ from discrete, noisy measurements of $Z(\omega)$ is an ill-posed inverse problem ($\frac{1}{1+i\omega\tau}$ is smoother), and minor experimental errors can cause large, oscillatory artifacts, resulting in DRT estimates that are not

physically plausible. Stabilization requires regularization, that is, augmenting the data-fit objective with constraints (*e.g.* smoothness, non-negativity, and sparsity) that suppress noise amplification while preserving resolvable structure. Open-source tools have accelerated the adoption of DRT across various electrochemical applications, including batteries and fuel cells, as well as corrosion, semiconductors, and bioimpedance [7]. This rapid interest, however, has at times outpaced a full appreciation of the method's mathematical details, strengths, and limitations. As the goal is to obtain a DRT that is both stable and interpretable, the resulting perception of simplicity carries a risk of overinterpretation, particularly because practical analysis introduces user choices that trade off data fidelity against solution smoothness. A critical, standard-based practice benchmarked against separate experimentation is therefore essential. This article highlights recent methodological advances that reduce subjectivity and improve robustness, and it surveys the new classes of applications enabled by these more reliable approaches. To orient the reader, Figure 1 summarizes recent developments of DRT's methodological foundations, core applications in electrochemical energy systems, and emerging technologies, providing a visual roadmap to the following sections.

From impedance to DRT: Mathematical framework and the ill-posed inverse problem

Eq. (1) defines the forward map from the DRT to the impedance. Under standard regularity conditions, each admissible $\gamma(\log\tau)$ yields a unique impedance $Z(\omega)$. Formally, one may also write the following inverse representation that expresses (under several assumptions) $\gamma(\log\tau)$ in terms of an analytic continuation of $Z(\omega)$ into the complex plane [8,9] [1] as follows:

$$\gamma(\log\tau) = -\frac{1}{\pi}\Im\left[Z\left(\frac{i}{\tau}\right) - Z\left(-\frac{i}{\tau}\right)\right] \tag{2}$$

where the latter equation has close connections to the inverse of the Stieltjes transform [10]. In practice, this expression requires the recovered impedance to be an analytic function over the complex plane, whereas EIS measurements provide noisy $Z(\omega)$ data only at discrete frequencies over the positive real axis ($\omega > 0$). Accordingly, to estimate the DRTone either (i) posits a representation for $\gamma(\log\tau)$ and fits it *via* Eq. (1) or (ii) regresses $Z(\omega)$ using an analytic model (*e.g.* an equivalent circuit) and then recovers $\gamma(\log\tau)$ using Eq. (2). This distinction is essential as the DRT is not merely a sum of RC-Voigt-like elements; it is a specific form of the Stieltjes transform between the frequency and time-constant domains [10], yet Eq. (2) is not universally applicable and must be interpreted in the sense of distributions at simple RC poles; in fact, if Eq. (2) is used naively, $\gamma(\log\tau)$ vanishes away from the pole and is undefined at the pole itself. Likewise, for unbounded elements that represent semi-infinite processes, the DRT diverges and should be treated in an *ad hoc* fashion and in the sense of generalized functions. As an illustration, the open-circuit, finite-length, blocking 1D transmission-line model cannot be used in a simple vanilla sense within the context of the DRT. The impedance of [11]

$$Z(\omega) = R\frac{\coth\sqrt{i\omega\tau_0}}{\sqrt{i\omega\tau_0}} \tag{3}$$

which is unbounded and converges to infinity, yet even a seemingly intractable element like that given in Eq. (3) admits a DRT. For instance, we can rewrite using Mittag-Leffler expansion as follows:

$$Z(\omega) = \frac{R}{i\omega\tau_{\tau_0}} + 2\,R\sum_{n=1}^{\infty}\frac{1}{1+i\omega\frac{\tau_0}{(n\pi)^2}} \tag{4}$$

where upon subtraction of the capacitive terms, the characteristic times $\tau_n = \frac{\tau_0}{(n\pi)^2}$ are the locations of the poles of $Z(\omega)$.

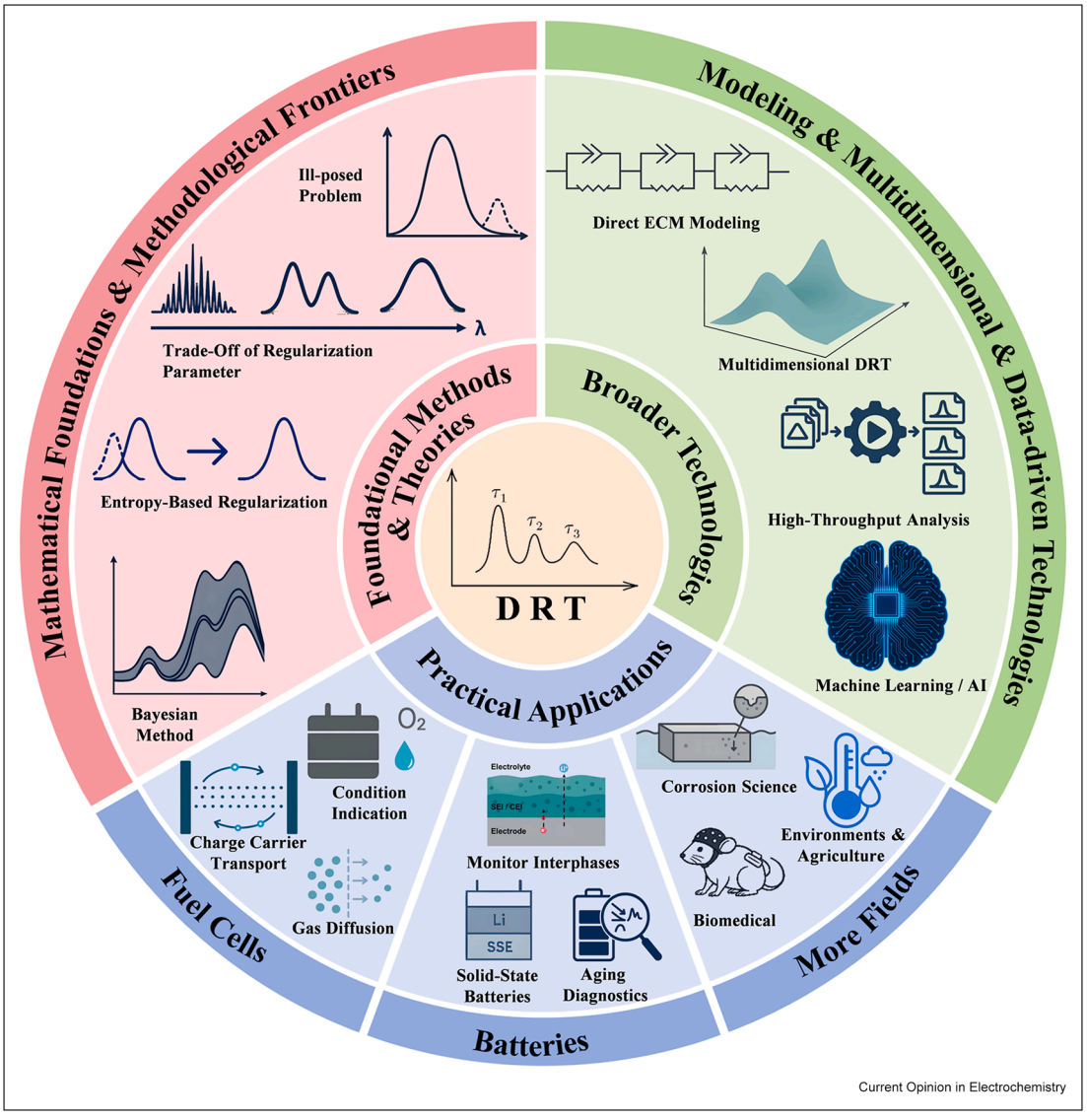
In practice, DRT deconvolution is implemented *via* surrogate models which can be (i) nonparametric, including Gaussian-process priors or nonparametric Bayesian mixtures (*e.g.* Dirichlet-process mixtures); (ii) parametric, featuring for instance structured peak families or neural networks; or (iii) semiparametric, that is, hybrids that couple a parametric core with a nonparametric component [12,13]. Physical interpretation remains delicate as identifiability limits persist; robust peak-to-mechanism assignments require support *via* complementary measurements, prior work, or mechanistic modeling.

Methodological frontiers toward robust and trustworthy DRT analysis

Inverting through Eq. (1) from noisy data is ill-posed because even tiny perturbations in data can cause large, oscillatory changes in a finite representation of data, leading to many different DRTs that fit the data within experimental errors [14]. Regularization restores stability by minimizing a weighted sum of data misfit and a penalty on the roughness or complexity of $\gamma(\log\tau)$. Tikhonov (ridge) remains widely used, typically penalizing the 0th, first, or second derivative to favor DRT simplicity or smoothness, with the regularization parameter λ governing the resolution−stability trade-off. Small λ yields high resolution but gives noise-amplifying, peak-rich solutions. In contrast, large λ suppresses noise at the risk of over-smoothing and peak merging (Figure 2(a)) [3,6]. The regularization level λ is typically chosen using methods like the L-curve, generalized cross-validation (GCV), or the discrepancy principle. While these methods are practical, they are not universally optimal. Furthermore, before adjusting λ, it is advisable to perform Kramers−Kronig (KK)/Hilbert consistency checks on the raw impedance data to assess random and systematic errors. Ignoring this step can lead to contamination from drift, lead inductance,

[1] Extensions to this formula including taking into account poles in the impedance are also available.

Figure 1



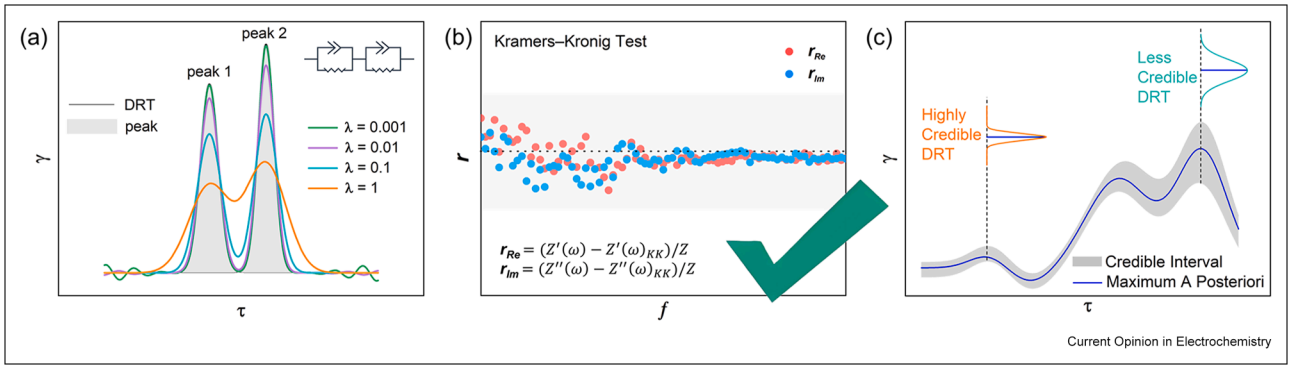
A landscape of DRT's recent progress: foundational methods, theories, practical applications, and broader technologies. DRT, distribution of relaxation times.

or nonlinearity, which may bias the DRT and distort the inversion results (see Figure 2(b)). For data consistent with KK/Hilbert relations, adaptive and hybrid techniques such as hierarchical or Bayesian λ selection, entropy penalties, and scale-dependent smoothing can enhance stability while maintaining the resolution of peaks in complex systems.

A major advance is the adoption of Bayesian methods, which move from seeking a single best-fit DRT to estimating a full posterior over admissible DRTs (Figure 2(c)) [15–17]. Treating $\gamma(\log\tau)$ as a random process and incorporating prior knowledge (*e.g.* nonnegativity and smoothness), Bayes' theorem yields credible bands that widen appropriately with noise and helps prevent overinterpretation of artifacts. Hierarchical models can learn the regularization level from the data, mitigating subjectivity. Entropy-based regularization offers an alternative to smoothness penalties by favoring the maximum-entropy distribution consistent with the data; this often suppresses unphysical peaks and avoids undue peak broadening observed with ridge regression [18] (Table 1). These frameworks also support uncertainty quantification and joint inversion of multiple spectra.

Recent work distinguishes two objectives in DRT analysis, namely (i) regression, which estimates the continuous amplitude of the DRT across time scales and (ii) process

Figure 2



Mathematical foundations and methodological frontiers. **(a)** Distributions obtained by the DRT analysis with a variation of different λ values. **(b)** An example of residual results from Kramers–Kronig (KK) tests. **(c)** An example of a Bayesian DRT analysis result. DRT, distribution of relaxation times.

Table 1

Comparison of different regularization methods.

Method	Underlying principles	Key advantages	Key limitations
Tikhonov regularization	Minimizes squared error + roughness penalty	Simple, computationally fast, and widely available	Highly sensitive to subjective choice of regularization parameter (λ); prone to artifacts
Bayesian/hierarchical Bayesian	Computes a posterior probability distribution of possible DRTs	Provides quantitative uncertainty quantification (credible intervals); can be self-tuning	Computationally more intensive than simple regularization
Entropy-based regularization	Minimizes squared error + entropy penalty (favors smoothness)	Effectively suppresses spurious peaks and noise-induced artifacts; high accuracy	Can be computationally intensive; newer and less widely implemented
Dual-regression classification	Unifies regression (magnitude) and classification (process identification) tasks	Provides novel metrics for accuracy; enables autonomous model selection	A conceptual framework with emerging algorithms; still under active development

DRT, distribution of relaxation times.

identification, a quasi-classification task that infers the number and locations (in $\log\tau$) of discrete processes. The latter can be pursued sequentially (first estimating $\gamma(\log\tau)$ *via* Eq. (1), then detecting peaks) or jointly with models that infer a finite, data-determined set of components and their amplitudes (*e.g.* Bayesian mixtures) without prespecifying the component count. Making process identification explicit enables autonomous model selection and metrics that assess both the quality of fitting and the correctness of process detection. These approaches are promising but remain at an early stage [14,19].

Machine learning models, including Gaussian processes and neural networks, tackle the ill-posed DRT inversion by embedding regularization in the prior and architecture (*e.g.* kernel choice, weight decay, and smoothness/monotonicity constraints), which stabilizes the estimate [20]. They perform best when constrained by physics (KK consistency, non-negativity, and causality) and validated against synthetic and experimental ground truth with calibrated uncertainty.

From interpreting spectra to predicting failure: DRTs expanding role

Recent advances in DRT deconvolution have broadened the scope of DRT, shifting it from primarily interpretive analysis to a practical basis for predictive diagnostics in operational systems.

Core applications: achieving deeper mechanistic insights

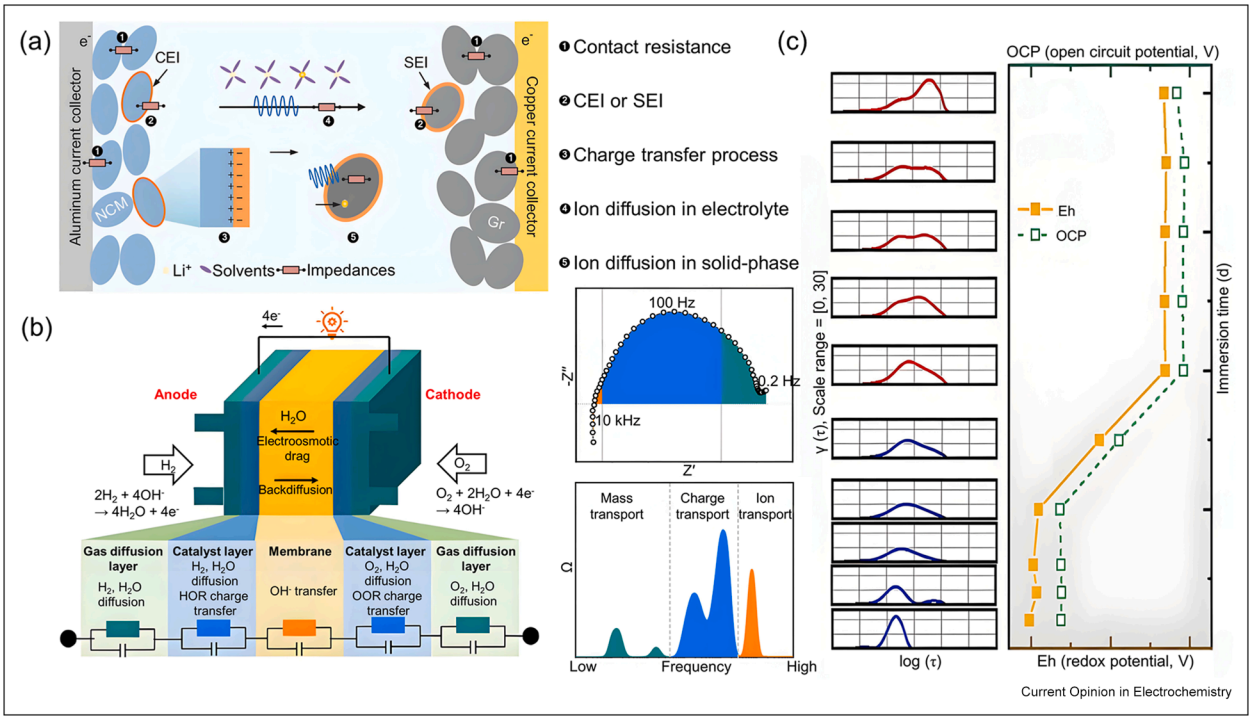
In the field of ionic materials, encompassing batteries and fuel cells, DRT is routinely used to separate impedance

contributions. For example, in battery systems, solid electrolyte interphase (SEI)/cathode electrolyte interphase formation [21,22], charge transfer kinetics [5,23,24], solid-state diffusion [25,26], and mass transport limitations [27]. In rechargeable batteries, the evolution of the DRT spectrum over thousands of cycles can be tracked. It has been shown that the growth of a mid-frequency feature often correlates with SEI thickening [28,29], while a high-frequency shift to longer times has been attributed to loss of active material or degraded contact [30,31] (Figure 3(a)). Time-resolved DRT has clarified degradation in all-solid-state batteries by linking performance loss to interfacial dynamics at solid–solid contacts under operating conditions [32–34]. In fuel cells, DRT enables mechanistic diagnosis by mapping characteristic timescales to specific phenomena, such as catalyst-layer flooding (low-frequency, mass transport peaks) [35,36], membrane dehydration (increased high-frequency proton transport resistance) [37], and anode poisoning [38] (Figure 3(b)). For instance, in protonic ceramic fuel cells, DRT has been used to deconvolve air-electrode impedance into charge transfer, ion migration, and surface exchange, thereby informing electrode design [39–41].

DRT-informed predictive diagnostics

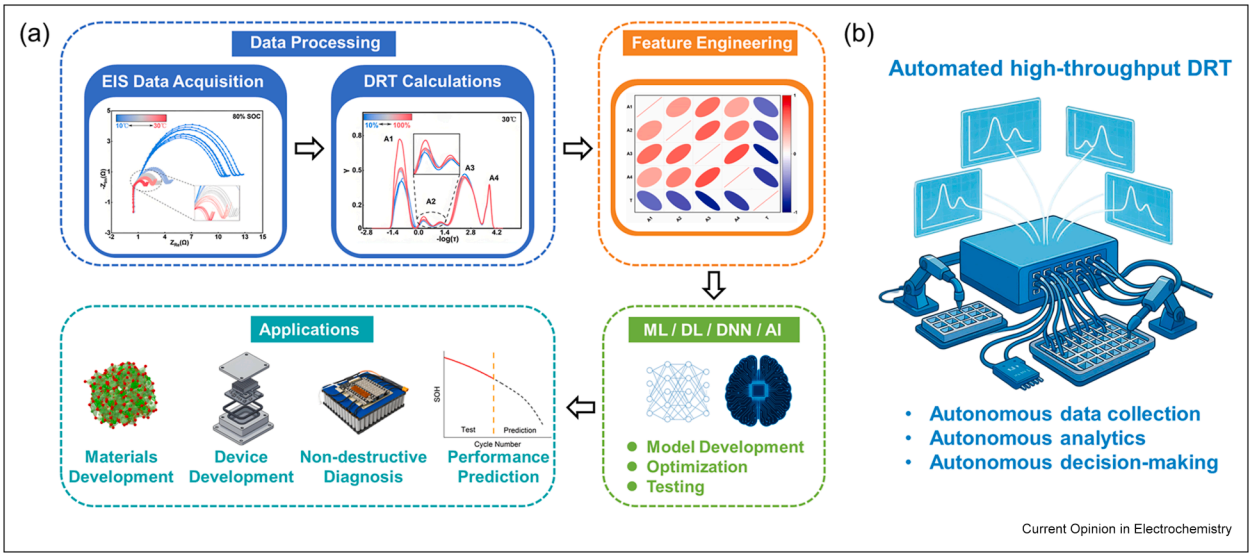
A notable trend is the use of DRT-derived features for predictive diagnostics [28,44,45]. Quantitative descriptors, including peak area, location, and width, serve as inputs to machine learning models for estimating state variables [46,47]. For example, Zhao et al. showed that incorporating DRT features in a hybrid model improved state-of-health (SOH) prediction relative to models based solely on raw impedance [46]. Because these descriptors are anchored to identifiable physical processes, shifts in peak time constants admit direct mechanistic interpretation, enabling early-stage anomaly detection, and improving remaining-useful-life forecasts. A longer-term objective is to integrate fast DRT algorithms with onboard battery management systems for real-time adaptive control and early warning, supporting digital-twin frameworks [48–51].

Figure 3



Application examples of DRT. **(a)** Mechanisms of timescale identifications of a typical Li-ion battery (Copyright 2024, Authors, Wiley, reproduced with permission) [42]. **(b)** The ion, charge, and mass transport processes are illustrated at their respective locations in the anion exchange membrane fuel cell (AEMFC) and corresponding DRT plot, divided into three regions, with ion, charge, and mass transport resistance (Copyright 2025, Authors, Wiley, reproduced with permission) [37]. **(c)** The corrosion evaluation of the aerobic–anaerobic corrosion transition of low-carbon steel under deep geological disposal conditions by DRT analysis. CEI, cathode electrolyte interphase; DRT, distribution of relaxation times; SEI, solid electrolyte interphase. (Copyright 2025, Elsevier, reproduced with permission) [43].

Figure 4



More technologies for DRT analysis. **(a)** The common workflow framework of developing machine learning/deep learning/deep neural network/artificial intelligence models for DRT analysis (Copyright 2025, Elsevier, reproduced with permission) [48]. **(b)** Schematic diagram for automated high-throughput DRT analysis. DRT, distribution of relaxation times; EIS, electrochemical impedance spectroscopy.

Extensions include internal temperature estimation and diagnosis of specific faults (*e.g.* lithium plating). A longer-term goal is to integrate fast DRT algorithms into battery-management systems to enable real-time adaptive control and early warning within digital-twin frameworks.

Unraveling complexity in emerging electrochemical systems

Improved deconvolution reliability is also enabling studies in systems with partially understood physics. In corrosion science, DRT has yielded insights into passive-film evolution [52] and alloy corrosion mechanisms [53]; for example, Wang et al. identified the aerobic-to-anaerobic transition of steel under deep geological disposal by linking a specific DRT feature to oxygen depletion [43] (Figure 3(c)). In perovskite solar cells, a dual-DRT approach separated timescales associated with fast capacitive charging, trap-mediated recombination, and ionic migration [54,55]. Applications are also emerging in bioimpedance, where DRT can resolve continuous relaxations in cells and tissues more faithfully than classical Cole–Cole models [56] and in breast tumor monitoring by impedance tomography using Gaussian relaxation-time distributions [57]. More broadly, DRT has been applied to electrolyzers [58], photoelectrochemical synthesis [59], agricultural monitoring [60], gas sensing [61], electric capacitors [62], desalination [63], construction materials [64], and photocurrent spectroscopy [65]. This breadth underscores the need for rigor: validation against complementary measurements should accompany DRT analyses in nascent applications to avoid artifact-driven interpretation.

Conclusions and perspectives

Progress on the ill-posed inverse problem has increased the reliability and utility of DRT. Methods that quantify uncertainty, suppress artifacts, and scale to large datasets have shifted DRT from qualitative interpretation to a quantitative, predictive framework.

A logical next step is multidimensional DRT, parameterizing processes over ln τ and covariates (SOH, SOC, temperature, and partial pressure) This facilitates cross-validation of peak assignments and process-level modeling. Substantial progress is likewise contingent upon standardization: the establishment of open-benchmark datasets that couple synthetic ground truth with curated measurements on canonical systems, as well as transparent reporting of code, hyperparameters, and raw data. Journals and agencies can accelerate progress by encouraging deposition of data and code and by adopting concise DRT-reporting checklists.

A pragmatic long-term objective is to integrate robust DRT into transparent, automated platforms. At the application level, models trained on high-throughput datasets that correlate experimental covariates with interpretable DRT features can facilitate precise SOH regression, mechanistic differentiation, and fault diagnosis (Figure 4(a)). These functionalities inspire the development of fully automated, closed-loop workflows capable of data acquisition, analysis with uncertainty quantification, and real-time adaptation of experimental conditions, thereby decreasing costs and enhancing reproducibility (Figure 4(b)). Transparent

implementations will expand accessibility and encourage proper usage. By emphasizing rigor, reproducibility, and judicious automation, the community can guarantee that DRT remains a dependable instrument for discovery and innovation within the field of electrochemistry.

Declaration of Generative AI and AI-assisted technologies in the manuscript preparation process

During the preparation of this work the authors used Gemini (Google) in order to improve the language and clarity of certain parts. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

Declaration of competing interest

There are no competing interests to disclose.

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List of abbreviations and symbols

EIS	Electrochemical impedance spectroscopy
DRT	Distribution of relaxation times
SOC	State of charge
SOH	State of health
SEI	Solid electrolyte interphase
CEI	Cathode electrolyte interphase
τ	Timescale
Z	Impedance
f	Frequency
ω	Angular frequency, $\omega = 2\pi f$
$\gamma(log\ \tau)$	Distribution of relaxation times
R_∞	Series resistance
i	Imaginary unit
L_0	Inductive component
C_0	Capacitive component
λ	Regularization parameter

Data availability

No data was used for the research described in the article.

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