Supplementary Information

Advancing Electrochemical Impedance Analysis: Innovating with the Distribution of Relaxation Times

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1 Tables

Table S1. Statistics of the responses regarding the usefulness of the existing DRT-deconvolution methods and pieces of software for analyzing data of electrochemical impedance spectroscopy.

Existing methods/software capability	**Years of experience**					
	1-15	16-30	31-45	46-60	61-75	Total
Yes	76 (70.4%)	2 (100.0%)	1 (50.0%)	2 (66.7%)	1 (50.0%)	82 (70.1%)
No	32 (29.6%)	0 (0.0%)	1 (50.0%)	1 (33.3%)	1 (50.0%)	35 (29.9%)
Total	108 (100.0%)	2 (100.0%)	2 (100.0%)	3 (100.0%)	2 (100.0%)	117 (100.0%)

Table S2. Statistics of the responses regarding the applicability of the DRT method in biology, medicine, and agriculture.

DRT method in biology, medicine, and agriculture	**Years of experience**					
	1-15	16-30	31-45	46-60	61-75	Total
Strongly agree	20 (18.5%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	20 (16.9%)
Agree	34 (31.5%)	1 (33.3%)	1 (50.0%)	1 (33.3%)	2 (100.0%)	39 (33.1%)
Undecided	48 (44.4%)	2 (66.7%)	1 (50.0%)	2 (66.7%)	0 (0.0%)	53 (44.9%)
Disagree	4 (3.7%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	4 (3.4%)
Strongly disagree	2 (1.9%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	2 (1.7%)
Total	108 (100.0%)	3 (100.0%)	2 (100.0%)	3 (100.0%)	2 (100.0%)	118 (100.0%)

Table S3: Statistics of the responses regarding the applicability of the DRT method for analyzing various electrochemical systems.

DRT application	Years of experience					
	1-15	16-30	31-45	46-60	61-75	Total
Batteries	37 (16.9%)	2 (40.0%)	1 (16.7%)	1 (25.0%)	1 (20.0%)	42 (17.6%)
Fuel Cells	98 (44.7%)	3 (60.0%)	3 (50.0%)	2 (50.0%)	2 (40.0%)	108 (45.2%)
Supercapacitor	7 (3.2%)	0 (0.0%)	1 (16.7%)	0 (0.0%)	0 (0.0%)	8 (3.3%)
Electrolyzer	34 (15.5%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	2 (40.0%)	36 (15.1%)
Solar Cells	4 (1.8%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	4 (1.7%)
Photoelectrolytic Cells	6 (2.7%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	6 (2.5%)
Bioelectrochemical Cells	11 (5.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	11 (4.6%)
Sensors	6 (2.7%)	0 (0.0%)	1 (16.7%)	0 (0.0%)	0 (0.0%)	7 (2.9%)
Solid state ionics	8 (3.7%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	8 (3.3%)
Biological Tissues	8 (3.7%)	0 (0.0%)	0 (0.0%)	1 (25.0%)	0 (0.0%)	9 (4.6%)
Total	219 (91.6%)	5 (2.1%)	6 (2.5%)	4 (1.7%)	5 (2.1%)	239 (100.0%)

Table S4. Statistics of the responses regarding the usefulness of the DRT method.

DRT contribution to understanding materials and systems	**Years of experience**					
	1-15	16-30	31-45	46-60	61-75	Total
Identifying multiple relaxation mechanisms	70 (30.2%)	2 (28.6%)	0 (0.0%)	1 (11.1%)	1 (33.3%)	74 (29.4%)
Characterizing electrode/electrolytes interfaces	72 (31.0%)	2 (28.6%)	0 (0.0%)	3 (33.3%)	0 (0.0%)	77 (30.6%)
Evaluating battery performance	45 (19.4%)	2 (28.6%)	1 (100.0%)	2 (22.2%)	1 (33.3%)	45 (20.2%)
Optimizing fuel cells efficiency	33 (14.2%)	1 (14.3%)	0 (0.0%)	2 (22.2%)	1 (33.3%)	37 (14.7%)
Characterizing biological processes	5 (2.2%)	0 (0.0%)	0 (0.0%)	1 (11.1%)	0 (0.0%)	6 (2.4%)
Estimating state-of-health	7 (3.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	7 (2.8%)
Total	232 (92.1%)	7 (2.8%)	1 (0.4%)	9 (3.6%)	3 (1.2%)	252 (100.0%)

Table S5. Statistics of the responses regarding the expectations about new DRT-deconvolution methods and pieces of software.

Functionalities expected from new DRT-deconvolution methods/software	**Years of experience**					
	1-15	16-30	31-45	46-60	61-75	Total
Automatic deconvolution	62 (37.1%)	4 (57.1%)	1 (33.3%)	2 (22.2%)	1 (33.3%)	70 (37.0%)
Associating peaks to electrochemical processes	66 (39.5%)	2 (28.6%)	1 (33.3%)	3 (33.3%)	1 (33.3%)	73 (38.6%)
Timescale separation	34 (20.4%)	1 (14.3%)	0 (0.0%)	2 (22.2%)	1 (33.3%)	38 (20.1%)
Robust batch processing/analysis	5 (3.0%)	0 (0.0%)	1 (33.3%)	2 (22.2%)	0 (0.0%)	8 (4.2%)
Total	167 (88.4%)	7 (3.7%)	3 (1.6%)	9 (4.8%)	3 (1.6%)	189 (100.0%)

Table S6: Statistics of the responses regarding the limitations of existing DRT-deconvolution methods and software

Limitations of existing DRT-deconvolution methods and software	**Years of experience**					
	1-15	16-30	31-45	46-60	61-75	Total
Mathematical complexity	17 (10.1%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	17 (9.1%)
Physical interpretability	77 (45.6%)	1 (25.0%)	3 (60.0%)	2 (40.0%)	1 (33.3%)	84 (45.2%)
Limited understanding of underlying mechanisms	60 (35.5%)	2 (50.0%)	2 (40.0%)	2 (40.0%)	2 (66.7%)	68 (36.6%)
Simultaneous analysis of multiple spectra	15 (8.9%)	1 (25.0%)	0 (0.0%)	1 (20.0%)	0 (0.0%)	17 (9.1%)
Total	169 (90.9%)	4 (2.2%)	5 (2.7%)	5 (2.7%)	3 (1.6%)	186 (100.0%)

Table S7. Statistics of the responses regarding the areas of future improvement for the DRT method.

Aspect needing further improvements	**Years of experience**					
	1-15	16-30	31-45	46-60	61-75	Total
Software availability	29 (14.1%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	29 (12.8%)
Stable and easily understandable method	25 (12.2%)	1 (33.3%)	2 (33.3%)	2 (33.3%)	2 (33.3%)	32 (14.2%)
Classification and standardization of methods	34 (16.6%)	0 (0.0%)	1 (16.7%)	1 (16.7%)	1 (16.7%)	37 (16.4%)
Advance AI for DRT deconvolution	65 (31.7%)	0 (0.0%)	1 (16.7%)	1 (16.7%)	2 (33.3%)	69 (30.5%)
Robust peak deconvolution	22 (10.7%)	1 (33.3%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	23 (10.2%)
Automatic multidimensional DRT fitting	18 (8.8%)	1 (33.3%)	0 (0.0%)	1 (16.7%)	1 (16.7%)	21 (9.3%)
Combined physics-based model with DRT	8 (3.9%)	0 (0.0%)	1 (16.7%)	0 (0.0%)	0 (0.0%)	9 (4.0%)
Time-dependent analysis	4 (2.0%)	0 (0.0%)	1 (16.7%)	1 (16.7%)	0 (0.0%)	6 (2.7%)
Total	205 (90.7%)	3 (1.3%)	6 (2.7%)	6 (2.7%)	6 (2.7%)	226 (100.0%)

Table S8. Statistics of the responses regarding the trends or technologies that could affect the DRT method.

Emerging trend or technology that could affect the DRT method	**Years of experience**					
	1-15	16-30	31-45	46-60	61-75	Total
Big data analytics	29 (23.4%)	1 (50.0%)	0 (0.0%)	2 (28.6%)	0 (0.0%)	32 (23.5%)
Artificial intelligence	57 (46.0%)	1 (50.0%)	1 (50.0%)	3 (42.9%)	1 (100.0%)	63 (46.3%)
High performance computing	24 (19.4%)	0 (0.0%)	1 (50.0%)	1 (14.3%)	0 (0.0%)	26 (19.1%)
Neuromorphic computing	10 (8.1%)	0 (0.0%)	0 (0.0%)	1 (14.3%)	0 (0.0%)	11 (8.1%)
Non-invasive medical diagnostic	4 (3.2%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	4 (2.9%)
Total	124 (91.2%)	2 (1.5%)	2 (1.5%)	7 (5.1%)	1 (0.7%)	136 (100.0%)

Table S9. Statistics of the responses regarding the fields where the DRT method could be more applicable.

Field needing the DRT method	**Years of experience**					
	1-15	16-30	31-45	46-60	61-75	Total
Biology	31 (62.0%)	2 (66.7%)	0 (0.0%)	0 (0.0%)	1 (50.0%)	34 (57.6%)
Agriculture	10 (20.0%)	1 (33.3%)	0 (0.0%)	2 (66.7%)	1 (50.0%)	14 (23.7%)
Electro- and photoelectron-catalysis	2 (4.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	2 (3.4%)
Sensors	4 (8.0%)	0 (0.0%)	1 (100.0%)	1 (33.3%)	0 (0.0%)	6 (10.2%)
Corrosion	3 (6.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	0 (0.0%)	3 (5.1%)
Total	50 (100.0%)	3 (100.0%)	1 (100.0%)	3 (100.0%)	2 (100.0%)	59 (100.0%)

Table S10. Comparison of various software used for DRT deconvolution and EIS analysis through Kramers-Kronig (KK) tests and equivalent circuit model (ECM) fitting.

Software name	Source	Language	Supporting OS	Source code	Functionalities
DearEIS [1]	Free and open source	Python	Any (Windows, Linus MacOS)	Yes	DRT, ECM fitting, KK
DRT-GELM [2,3]	Free and open source	Python	Any	Yes	DRT
DRT-python-code [4]	Free and open source	Python	Any	Yes	DRT
DRTtools [5]	Free and open source	Matlab	Any	Yes	DRT, KK
ED-DRT [6]	Free and open source	Matlab	Any	Yes	DRT
Impedance Analysis [7]	Commercial	Igor Pro	Windows, MacOS	No	DRT, KK
ISGP [8]	Free, not open source	Matlab	Window	No	DRT, KK
LEVM/LEVMW [9]	Free and open source	Fortran	MS-DOS, Windows	Yes	DRT, ECM fitting, KK
pyDRTtools [5]	Free and open source	Python	Any	Yes	DRT, KK
pyimpspec [10]	Free and open source	Python	Any	Yes	DRT, ECM fitting, KK
Ravdav [11]	Commercial	Python	Window	No	DRT, KK
RelaxIS3 [12]	Commercial	N/A	Windows	No	DRT, ECM fitting, KK

2 Figures

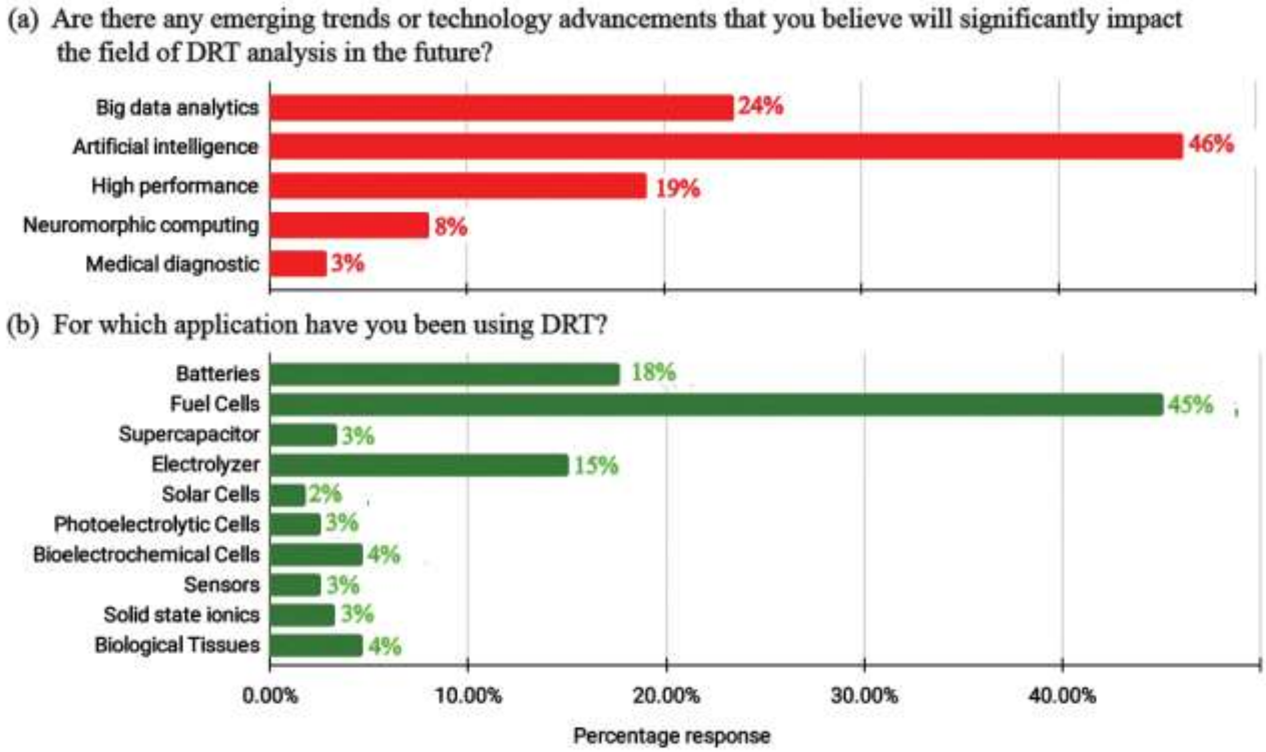

Figure S1: Bar chart showing the (a) responses regarding the emerging trend or technology advancements that may significantly impact the field of DRT in the future, and (b) the area of application of the DRT.

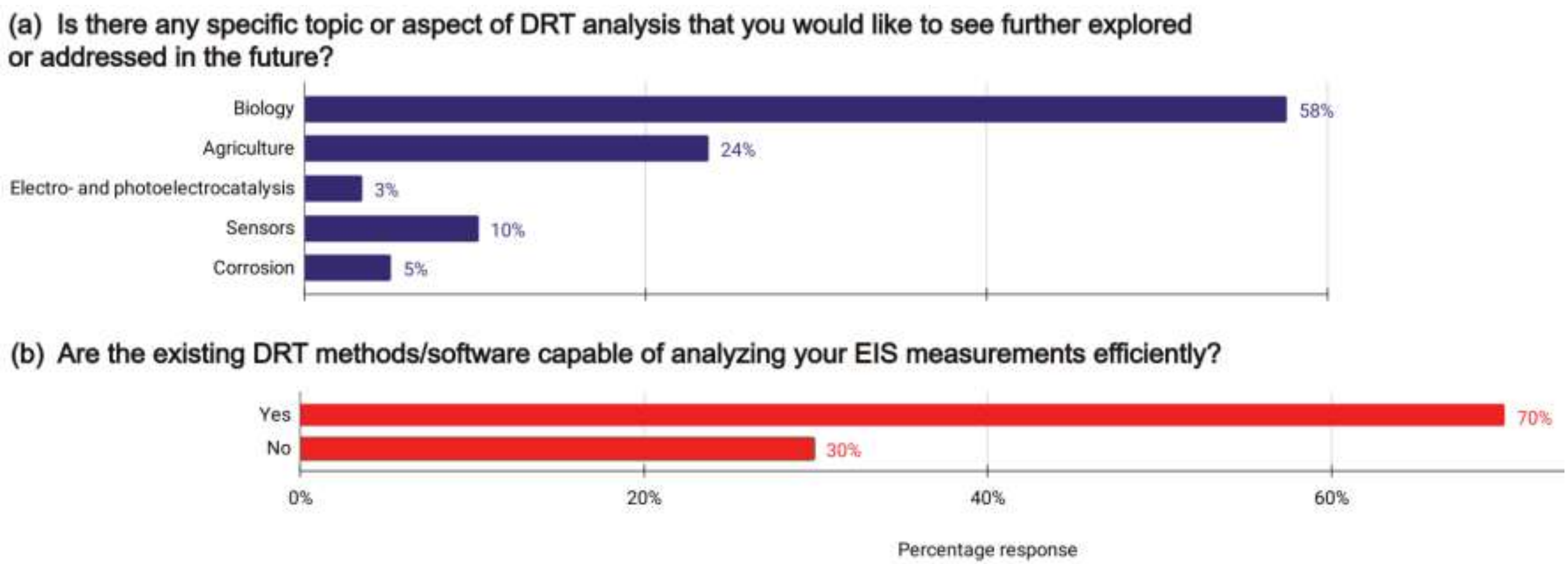

Figure S2: Bar chart showing the (a) responses regarding the specific research area or aspect of DRT analysis that should be further explored, and (b) the capability of the existing software methods/software in efficiently analyzing the EIS measurements.

3 DRT Software

This section showcases various DRT deconvolution software along with their salient features such as functionalities, operating systems, and source code. DRTtools [5], a MATLAB-based GUI, employs ridge regression and provides Bayesian and hierarchical Bayesian methods, including data quality assessment using Kramers-Kronig relations. The Python-based version, pyDRTtools retains all the features of DRTtools, and also offers optimal regularization parameter selection, robust peak deconvolution, and easy deployment as a Python package via “pip”. Additional Python-based open-access options include DRT-python-code [4], pyimpspec [10], and DearEIS [1], with ED-DRT being robust to data truncation and offering automatic regularization parameter selection [6]. Additional tools include LEVM/LEVMW (Fortran-based) [9], DRT-GELM, which discretization free inversion method but may produce spurious false peaks for timescale greater than 0.01 s [2,3], and impedance spectroscopy genetic programming (ISGP), which provides a user interface for DRT analysis and Kramers-Kronig compliance test [8] (see Table S10). Moreover, commercial options include Igor pro GUI for impedance analysis [7], RelaxIS3 for batch processing of multiple EIS spectra [12], and Ravdav DRT analysis and Kramers-Kronig relations tests [11].

3.1 Further challenges with EIS Analysis

For *in-situ* measurements conducted during cell operation, challenges arise as DRT plots struggle to consistently reflect trends with varying resistance (R_S) of solutions or Nafion membranes. The expected correlation between DRT results and changes in R_S remains elusive. Moreover, the identification of constant phase elements (CPE), particularly in cases reliant on double layers, poses challenges that can be addressed through DRT analysis.

In practical measurements, the Nyquist plots show the presence of inductance, especially at higher frequencies. The decision to either incorporate this inductance into the DRT as a negative peak or attribute it to cable so as to clearly distinguish the source of the inductance remains unaddressed. Exploring *in-situ* EIS for CO_2 reduction in a membrane electrode assembly reveals intricate reaction pathways and products. Consequently, the DRT results show overlapping peaks resulting from many electrochemical catalysis processes, thus complicating peak assignments. A DRT method that can directly associate peaks with different processes is needed to unravel these complexities.

While DRT timescale-based analysis effectively describes electrochemical processes, its applicability is hindered by its tendency to shift during electrochemical processes, causing issue in peak assignments. The discrepancy in timescale between experiments further complicates analysis. Reproducibility of EIS experiments, particularly in battery assembly and testing, presents challenges as variations in DRT plots are observed among assembled cells, despite similarities in Nyquist and Bode plots. The differences usually observed in the DRT plots are confusing and complicate the analysis, emphasizing the need for tailored DRT methods/software to address this issue.

4 Designed Survey Questionnaire

4.1 English Version

Dear Respondents,

We are currently conducting a survey aimed at gathering information on the utilization of DRT methods/software in analyzing EIS measurements. We kindly request your assistance in completing this questionnaire. The primary objective of this survey is to gain insights into the requirements and preferences of experimentalists, engineers, and professionals regarding DRT methods/software, as well as to understand the underlying reasons behind their choices. This survey forms a crucial part of our ongoing efforts to develop new, automated DRT methods/software, which can significantly contribute to expediting material discovery.

We greatly appreciate your participation in this survey, as your input is valuable to us. Your responses will be used exclusively for research purposes and will remain entirely anonymous. We are fully committed to ensuring the confidentiality of the information collected, which will solely serve the purpose of this research and will not be utilized for any other activities. The questionnaire is designed to be completed within approximately 5 minutes. Should you have any inquiries or concerns, please do not hesitate to contact me directly at amaradesa@connect.ust.hk .

The survey period is 2 weeks. Therefore, we expect all responses within the 2 weeks window. Thank you for your time and consideration.

1. How long have you been using DRT?

   Please specify the number of years ☐

2. For which applications have you been using DRT (Select all that apply)

   Batteries ☐ Fuel Cells ☐ Supercapacitors ☐

   Electrolyzers ☐ Solar Cells ☐ Others (please specify) ---------------

3. Are the existing DRT methods/software capable of analyzing your EIS measurements efficiently?

   Yes ☐ No ☐

4. If "No" in question 3 above, what limitations do you find in the existing DRT methods/software? (Select all that apply)

Mathematical complexity ☐ Physical interpretability ☐

Limited understanding of underlying mechanisms ☐

others (please specify) --

5. In your experience, how has the DRT analysis contributed to the understanding of materials and systems? (Select all that apply)

   Identifying multiple reaction mechanisms ☐

   characterizing electrode/electrolyte interfaces ☐

   Evaluating battery performance ☐

   Optimizing fuel cells efficiency ☐

   Others (please specify) --------------------------

6. DRT has been recently applied in Biology, Medicine, and Agriculture.

   Strongly agree ☐ Agree ☐ Undecided ☐

   Disagree ☐ Strongly disagree ☐

7. What functionalities do you expect new DRT software to possess? (Select all that apply)

   Automatic deconvolution ☐

   Associating peaks to electrochemical processes ☐

   Timescale identification ☐

   Others (please specify) ----------------------------------

8. Are there any emerging trends or technology advancements that you believe will significantly impact the field of DRT analysis in the future?

Please elaborate ---

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9. In your own opinion, what can still be improved regarding DRT methods/software?

Please elaborate ---

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10. Is there any specific topic or aspect of DRT analysis that you would like to see further explored or addressed in future research?

Biology ☐

Agriculture ☐

Others (please specify) ---

4.2 Chinese Version

尊敬的老师和同仁们，

近年来，学界对使用 DRT 方法/软件分析 EIS 测量信息的研究方法越来越感兴趣。德国拜罗伊特大学 Francesco Ciucci 教授（原港科大教授）的研究团队开发的 DRTtools 开源软件已经在学术界广泛应用了近十年。为了顺应 DRT 的发展趋势并提供更多功能和更易用的软件，Ciucci 教授计划对 DRTtools 软件进行版本更新。因此，他授权我们在内地和亚洲学术圈内进行一项中文调查，旨在了解实验人员、工程师和学术界专业人员对 DRT 方法/软件的需求和偏好。我们诚恳地邀请您抽出时间填写这份预计 5 分钟完成的问卷，以共同促进 DRT 方法/软件的发展。您的回答将仅用于研究

目的，并保持完全匿名。参与调查，请点击以下链接或直接扫描二维码。如果您有任何问题，请随时通过 yuhao.wang@connect.ust.hk 与我联系。

问卷链接：https://ust.az1.qualtrics.com/jfe/form/SV_6DpspmIO4rG9WTk

非常感谢您的参与和支持！祝您身体健康，工作顺利！

1. 您使用 DRT 多长时间了？

   请写下具体的年数 ☐

2. 您在哪些电化学器件中使用了 DRT？（可多选）

   电池 ☐ 燃料电池 ☐ 超级电容器 ☐

   电解池 ☐ 太阳能电池 ☐ 其他（请注明）---------------

3. 您认为现有的 DRT 方法/软件是否能够有效地分析您的 EIS 测量结果？

   是 ☐ 否 ☐

4. 如果您在第三道问题的答案为“否”，您认为现有 DRT 方法/软件有哪些局限性？（可多选）

   数学复杂性 ☐ 物理可解释性 ☐

   对潜在机理的了解有限 ☐

   其他（请注明）---

5. 根据您的经验，DRT 分析如何有助于理解材料和系统？（可多选）

   识别多种弛豫机制 ☐

   表征电极/电解质界面 ☐

评估电池性能

优化燃料电池效率

其他（请注明） ------------------------

6. DRT 最近已/可被应用于生物学、农业、医学和腐蚀

完全同意 比较同意 既不同意也不反对

比较反对 完全不同意

7. 您希望新的 DRT 软件具备哪些功能？（可多选）

自动解卷积

将 DRT 峰与电化学过程相关联

时标识别

其他（请注明） ---------------------------------

8. 您认为未来是否有任何新兴趋势或技术进步会对 DRT 分析领域产生重大影响？请详细说明。

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9. 在您看来，DRT 方法/软件还有哪些可以改进的地方？

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10. 在未来的研究中，您是否希望看到使用 DRT 分析哪些具体主题或方面？如果有的话，您最希望是哪些器件和具体问题？

生物学

农业 ☐

其他（请注明） ---

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