「蓄電固体デバイスの創成に向けた界面イオンダイナミクスの科学」

領域番号：6107

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研究成果報告書

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はしがき

本領域では、固体内で電子・ホール以外にイオンが電荷キャリアに含まれる材料（＝蓄電固体材料）のヘテロ・ホモ接合界面で発現する特異なイオンダイナミクスの機構を解明し、イオンを自在に超高速輸送・高濃度蓄積し得る界面構築のための指導原理を確立する研究を行った。目的達成に向けて、①モデル界面構築、②高度計測、③計算・データ科学、④機能開拓の4つの研究項目のもと、化学・物理・情報・材料の異分野にわたる研究を融合し、新たな固体界面科学の学理構築を目指した。この学理構築をもとに、全固体電池などの蓄電固体デバイスの飛躍的高性能化という社会的インパクトを与えるだけでなく、既存の概念に捉われない新世代イオンデバイスの創成と更なる創造的学問体系への発展に繋がる成果が得られた。

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３　交付決定額

年度	合計	直接経費	間接経費
令和元年度	285,090,000 円	219,300,000 円	65,790,000 円
令和 2 年度	297,050,000 円	228,500,000 円	68,550,000 円
令和 3 年度	297,180,000 円	228,600,000 円	68,580,000 円
令和 4 年度	297,050,000 円	228,500,000 円	68,550,000 円
令和 5 年度	289,770,000 円	222,900,000 円	66,870,000 円
合計	1,466,140,000 円	1,127,800,000 円	338,340,000 円

研究発表

雑誌論文（論文はすべて査読有り。*は責任著者。）

1.

	著者名	論文題目	*雑誌名,* **巻・頁・（発行年）**
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55	Yoshitaka Tateyama, “Theoretical Analysis of Microscopic Interface Ionics at Heterogeneous Solid-Solid Interfaces in Batteries” PRiME 2020 Online
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58	Yukio Takahashi, “XAFS-Ptychography at 4th generation storage rings”, PETRA IV Workshop - Materials and Processes for Energy and Transport Technology, Onlin
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71	Kan Hatakeyama-Sato, "Machine learning-assisted exploration of glassy organic electrolytes for solid-state lithium-ion batteries", 45th International Conference and Expo on Advanced Ceramics and Composites
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75	Yasutoshi Iriyama, Preparation of anti-perovskite structured Li2HOBr and its application to all-solid-state batteries” 45th International Conference and Expo on Advanced Ceramics and Composites Online
76	Takashi Tsuchiya, Tohru Tsuruoka, Kazuya Terabe "Ionic nanoarchitectonics for information storage, neuromorphic and decision making functionalities", MANA International Symposium 2021 Jointly with ICYS, Online
77	Yasutoshi Iriyama, "Challenges toward large-sized oxide-based all-solid-state batteries", Virtual International Workshop on Energy Storage Technology for E-mobility (IWESTE-2021), Online,
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79	本山 宗主, “酸化物系無機固体電解質を介した Li 析出溶解反応のその場 SEM 観察”, 日本顕微鏡学会 その場観察分科会 2020 年研究討論会～電場中のその場観測～, Online
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81	Akichika Kumatani, “Recent Advances in Scanning Electrochemical Microscopic Analysis and Visualization”, MSM-AIMR Joint Online Workshop 2020, Online
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84	藪内 直明, “多電子固相酸化還元反応を利用した Li イオン電池用高容量正極材料”, 第 112 回新電池構想部会, Online
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86	林 晃敏, “全固体電池実現にむけた材料開発と界面プロセス”, 日本ゾル-ゲル学会第 17 回セミナー, Online
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92	笠松 秀輔, "拡張アンサンブル法・第一原理計算結合フレームワークによる固体中の配置不規則性の大規模サンプリング", 物性研究所スパコン共同利用・CCMS 合同研究会「計算物質科学の新展開 2020」, Online
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102	Yukio Takahashi, "X-ray Spectro-Ptychography: Visualization of Chemical State at Mesoscale", MIRAI 2.0 R&I Week 2021, Online,
103	武藤俊介, "ポリマーブレンドのナノスケール化学イメージング", 第 70 回高分子討論会 S6. ソフトマテリアルイメージングの最前線, Online,
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111	Teruo Kanki, "Power-free synaptic transistor", MEMRISYS 2021, Online.
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117	林 晃敏, “Ductile Solid Electrolytes for All-Solid-State Batteries”, Solid-state Batteries 4.0 (SSB 4.0)- from Fundamentals to Application an online International Bunsen Discussion Meeting, Online
118	林 晃敏, “高ナトリウムイオン伝導性硫化物電解質の開発”, 電気化学会電池技術委員会第 406 回委員会, Online
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121	林 晃敏, “全固体電池の研究開発動向”, NEDIA 第 8 回 SSIS-NEDIA 関西シンポジウム, Online
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141	中山　将伸, "Li イオン導電性 NASICON 型材料の固固界面構造と材料計算", 第 1 回計算イオニクス研究会（第 78 回固体イオニクス研究会）, Online
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145	Kaoru Dokko, "Ion Hopping Conduction in Molten Solvates of Li Salts for Li-S Batteries", 46th International Conference and Expo on Advanced Ceramics and Composites (ICACC2022), Online
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148	Yoshitaka Tateyama, "Ion and Electron Transfer at Interfaces in Solid-state Batteries Via First-principles Calculations", International Battery Association 2021, International Battery Materials Association, Hybrid: Xiamen, China
149	館山 佳尚, "第一原理計算", 電気化学会関東支部第 49 回先端科学セミナー　電気化学のための計算化学入門 電気化学会関東支部, Online
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図書

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研究成果

本領域の研究項目は、(1)蓄電固体界面のモデル構築（A01）、(2)界面近傍の物性の高度計測（A02）、(3) 蓄電固体界面の計算・データ科学（A03）、(4) 界面機能開拓（A04）、の 4 項目であり、A01－A04 の各計画研究で実施した。この計画研究を補強・強化・発展するために前半では 21、後半では 22 の公募研究が参画した。総括班は計画研究・公募研究の連携推進、HP 等を活用した成果公開やシンポジウム等の企画、連携戦略の立案や領域内の問題解決などを領域評価委員・オブザーバーの先生方からの意見を参考に進めた。これら研究項目の有機的連携により学理構築を目指した（図 2）。

蓄電固体界面の学理構築が難航しているのは、実験系で組みあわされる材料とその構築プロセスが多岐にわたり、情報が発散していることも要因の一つである。本領域は A01 あるいは A04 が提供する標準材料を多角的に計測し、この結果を理論と照らし合わせることで、実験・計測・理論が整合する研究を進めた。本領域の計画研究では、室温近傍で作動し材料・計測の基礎構築が出来ている Li および Na に絞った。国際共同研究、NEDO の SOLiD-EV（後継の SOLiD-Next）及び JST の ALCA-SPRING（後継の GteX）との実用化に向けた連携研究、「富岳」との計算・データ科学における連携研究は、いずれも本領域の研究を強化・補強する役割であるとともに、構築されていく基礎学理をテストする場ともなった。

界面近傍の物性変化の探求にはモデル界面が必要であり、A01 が担当した。このモデル界面の物性変化を調べるためには、高度計測の統合による界面物性（イオン物性、電子物性、構造物性、力学物性）の計測が必要であり、A02 が担当した。得られた計測結果をもとに界面イオンダイナミクスの機構を解明するためには理論構築が必要であり、A03 が担当した。蓄電固体界面が備える特性を知り、それを材料とするには複合材料を開発する必要があり、A04 が担当した。こうした連携で蓄電固体界面が機能発現する特性決定因子が抽出され、物性分布と特性決定因子との相関を明らかにしながら学理構築を進めた。この際、鍵となる 8 つの界面基礎の横串連携を構築し、計画研究連携・横串連携の相乗効果を活用して蓄電固体界面科学の学理を深化させた。

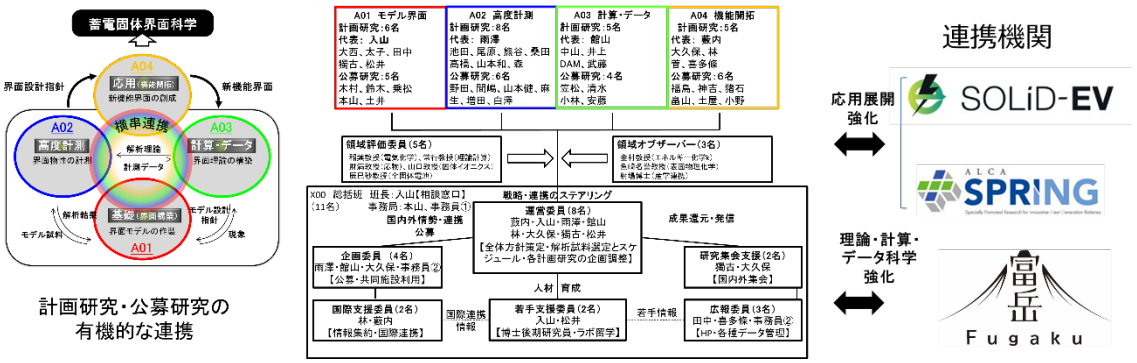

図 2　領域研究の連携・強化の模式図.

提案時の 4 つの柱（計画研究）の戦略的連携のもと、界面イオンダイナミクスに関わる因子の解明（基礎学理構築）を目標とした研究を行った。構築した学理をもとに、全固体電池などの産業競争力強化に資する次世代固体イオニクスデバイスの界面設計指針を明確化し、多くの成果が得られた。その中でも、特に顕著な成果群を下記に示す（図 3）。

① **空間電荷層の実証と新規蓄電固体デバイスの創成**：電極と固体電解質の界面では空間電荷層が数 nm の厚みで存在しうることが実験的に実証され、それを活用したイオントランジスタ等、新規蓄電固体デバイスが開発された。
*T. Tsuchiya *et al.*, ***Commun. Chem.*** (2021) (A04)
M. Takayanagi, *T. Tsuchiya *et al.*, ***Mat. Today Phys.*** (2023) (A04)

② **無歪高容量電極材料の開発**：電極反応における高い電荷蓄積を実現しつつ、その体積変化を“ゼロ”にできる材料が実現し、車載用全固体電池の長寿命化に極めて効果的な材料となることが見出された。
I. Konuma, K. Ohara, T. Miyuki, *N. Yabuuchi *et al.*, ***Nature Mater.*** (2022) (A04+A02+SOLiD-EV).

③ **結晶化ガラス界面での高速イオン伝導機構の解明**：結晶化ガラスでは可動イオンの配位環境が大きく 3 タイプ存在し、結晶／ガラス界面近傍で高イオン伝導配位構造の割合が増加することを見出した。また、こうした界面を高濃度で形成するプロセスも開発された。
H. Yamada, *K. Ohara, K. Ikeda, R. Kobayashi, K. Amezawa, Y. Tateyama, S. Mori, *A. Hayashi, *et al.*, ***Energy Environ. Mater.*** (2023) (A04+A03+A02).
[P5] T. Kimura, A. Hayashi *et al.*, ***J. Am. Chem. Soc.***(2023) (A04)

④ **界面の高抵抗化トリガー解明**：電極と固体電解質を接合する際に界面近傍では可動イオンの化学ポテンシャル差に起因する不定比性が生じ、これが高抵抗化のトリガーとなることを解明した。この成果は高性能被覆層の材料開発にもつながった。また、電極と有機電解液界面を高抵抗化させる溶媒物性が明確となった。
H.-K. Tian, S. Muto, Y. Iriyama, *Y. Tateyama *et al.*, ***ACS Appl. Mat. Int.*** (2020) (A03+A01+ ALCA-SPRING)
K. Onoue, *M. Matsui *et al.*, ***ACS Appl. Mater. Int.*** (2023) (A01)

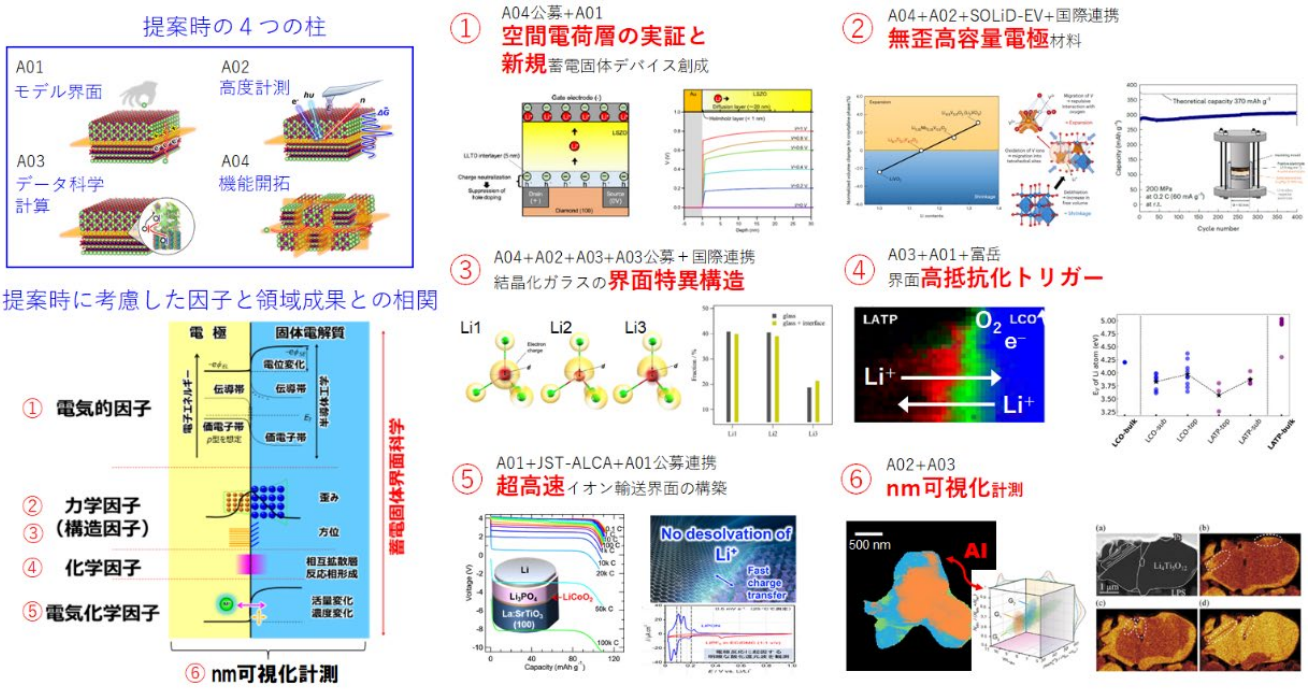

図 3　領域研究で得られた注目成果の一覧.

K. Yoshikawa, T. Ohnishi, K. Amezawa, *Y. Iriyama *et al.*, ***Adv. Sci.*** (2024) (A01+A02+SOLiD-Next)
Y. Ugata, *K. Dokko *et al*., ***J. Phys. Chem. C*** (2023) (A01).

⑤ **超低抵抗界面イオン輸送**：可動イオンの化学ポテンシャルを制御した界面構築手法を開発し、エピタキシャル電極と融合して、正極側・負極側ともに超高速界面イオン輸送界面を実現し、特異な電荷蓄積現象も見出された。

K. Kawashima, T. Ohnishi *et al.*, ***ACS Appl. Ene. Mat.*** (2020) (A01)
S. Yamamoto, *M. Motoyama, W. Norimatsu, A. Kumatani, Y. Iriyama *et al.* ***ACS Nano*** (2023) (A01+A02)

⑥ **nm 可視化計測**：平衡状態における蓄電固体材料の微構造分布や全固体電池の微視的反応分布を高度計測と AI を連携して可視化する計測手法を開発した。更に、定常状態の界面イオンダイナミクスを可視化する計測手法も確立した。

H. Uematsu, *N. Ishiguro, H. Dam, M. Okubo, Y. Takahashi *et al*., ***J. Phys. Chem. Lett.*** (2021) (A02+A03+A04)
*N. Ishiguro, K. Yamamoto, Y. Iriyama, Y. Takahashi et al., ***ACS Appl. Ene. Mat.*** (2023).(A02+A01)
Y. Nomura, *K. Yamamoto et al., ***J. Mater. Chem. A*** (2023) (A02)

以上のように、提案時に考慮した電気的因子、力学因子、構造因子、化学因子、電気化学因子に関わる卓越した成果が得られ、計測と計算の連携で nm 計測手法が開発された。これらにより、蓄電固体界面科学の学理構築とそれを用いた蓄電固体デバイスの高性能化・新規デバイス創成が実現した。

研究成果の社会への発信については、総括班を中心に下記の取り組みを行った（図 4）。

領域ホームページ（HP）　：　https://interface-ionics.jp/
※領域研究者の論文出版・講演、領域ニュース・イベント情報などをタイムラグなしに発信した。また、公開シンポジウムのオンライン受付にも使用した。

領域全体会議　　**：計 10 回実施**
※半年に 1 回実施した。これに加えて計画研究及び横串連携での会議も多数実施した。また、運営委員による会議を 43 回（毎回 2 時間）実施し、コロナ禍においても密な連携と迅速な軌道修正に務めた。

国際ニュースレター（NL）：計 8 回発行
※半年に 1 回発行した。領域成果を国際的アピールし、国際連携や若手研究者のキャリアパス支援等に活用した。

公開シンポジウム　　**：計 5 回実施**
※1 年に 1 回実施した。第 2-5 回はオンライン配信で実施し、毎回 300-400 名の方に参加登録頂き領域成果を効果的に配信した。第 2 回では吉野彰先生（2019 ノーベル化学賞受賞）にご講演頂いた。

国際会議・シンポジウム　：計 4 回実施
※第 1 回は静岡で開催し、11 カ国 157 名（44 名が海外）が参加した。これにより国際ネットワークが効果的に構築され、これをもとに第 2-3 回をオンラインで開催した。第 4 回はシンガポールで開催し、6 か国 49 名が参加した。この際には、M. S. Whittingham 先生（2019 ノーベル化学賞受賞）にご講演頂いた。

学会シンポジウム　　**：計 5 回実施**
※セラミックス協会、電池討論会、日本化学会、応用物理学会、電気化学会で各 1 回実施し、いずれも盛況であっ

図 4 : 総括班活動成果　(a) 領域 HP、(b) 国際ニュースレター、(c) 公開シンポジウム、(d) 学会シンポジウム、(e) 若手勉強会、(f)アウトリーチ、(g) 国際シンポジウム、(h)領域会議.

た。

若手勉強会　　：計 5 回実施

※蓄電固体材料・界面の基礎理解の定着と向上に加え、若手研究者の発表賞を設置して若手をエンカレッジすることを目的に取り組んだ。また、領域内での共通装置を紹介して国内ラボ留学の機会を創出し、多くの連携成果を生み出すきっかけとなった。

書籍化　　：計 2 冊取りまとめ中

※最先端成果を英文書籍 1 冊に取りまとめた（2024 年 10 月発行）。また、基礎成果を和文書籍 1 冊に取りまとめる予定で現在進めている。

以上のように、トップジャーナル誌への掲載や国内外での招待講演を多数実施し、HP を中心とした迅速な成果公開や多数のアウトリーチ活動、国際 NL の発行や国際会議・シンポジウムの実施による国際的な成果の公表、学会シンポジウムや公開シンポジウム主催など積極的に成果の公表・普及に努めた。