

FCC 鈴木章先生記念未来創造ラボ
研究成果報告書

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研究課題名	分野横断エレメント工学産学国際連携研究		
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研究組織

氏名（年齢）	所属	現在の専門	役割分担
伊藤 肇 (54)	応用化学部門	有機合成化学	研究統括・連携促進
長谷川靖哉 (54)	応用化学部門	錯体化学	発光材料・ナノ材料開発
大利 徹 (61)	応用化学部門	応用生物化学	生体関連物質研究計画立案
小笠原泰志 (45)	応用化学部門	応用生物化学	生体関連物質の合成と利用
島田敏宏 (57)	応用化学部門	固体反応化学	有機・無機ハイブリッド材料設計

活動計画の概要（研究の背景、意義・価値・構想理由・問題の所在等）

本研究計画は、鈴木章先生が長年研究を重ねられた「ホウ素化学」をベースにすべての元素の特長を活かし、あたらしいイノベーションをもたらす「エレメント工学」を研究することを目的としている。本研究計画の前段階に当たる平成 26 年度から令和元年度までの研究期間において、FCC 鈴木章先生記念未来創造ラボは研究参加者の共同研究の場として機能し、共同研究が成功裏に進められた。例を挙げると、伊藤と大利らによるペプチド合成関連の成果や伊藤と長谷川らによる新しい発光材料の研究、メカノケミストリーに関する研究が本ラボから発信されている。国際共同研究として、スイス連邦工科大学並びに米国 UCLA との国際共著論文も発表した。また、2016 年から 2021 年には、大利らを計画班とする新学術領域研究「生合成リデザイン」、2017 年から 2022 年では、伊藤（計画班）と長谷川らの共同研究から、新学術領域研究「ソフトクリスタル」、また 2018 年からは伊藤（拠点長）と長谷川が参画する北海道大学 WPI 反応創成研究拠点がスタートした。また 2019 年からは伊藤が CREST「革新的反応」に採択、また 2018 年には長谷川らが北海道大学ロバスト農林水産工学国際連携研究教育拠点到採択されている。これらのことは当ラボが共同研究のゆりかごとして機能していることを示している。

近年の科学研究における大きな問題は、分野の細分化と研究機器の高度化による専門化が進むことにより、分野間交流が妨げられ、イノベーションにつながる新発見が得られにくいという点が挙げられる。本研究では、分野の異なる 4 つの研究チームが、FCC 鈴木章先生記念未来創造ラボという自由度が高くかつ施設の充実したスペースを媒介に、分野を横断した連携研究と、将来的には産学並びに国際連携を視野に入れた研究行うものであった。3 年 9 ヶ月の研究の結果、別紙論文リストに示すような多大な研究成果に加えて、新しい共同研究に基づく外部資金の獲得を通じて、FCC と工学研究院だけでなく、北海道大学全体に多大な貢献をもたらした。

研究活動成果の概要（研究の進捗・研究目的の達成度、問題点等について）

FCC鈴木章記念未来創造ラボにおいて「分野横断エレメント工学産学国際連携研究」という研究課題名で2014年11月より研究をスタートし多くの成果をあげた。多くの研究成果を背景に、2019年から2022年まで上記ラボにおける研究を延長するに至った。我々は延長期間においても共同研究を活発に展開し、それぞれの専門領域の境界において研究論文を114報報告した（研究業績欄を参照）。

延長期間中に得た研究成果は非常に多彩であり、化学分野全体に影響を与える重要な結果を含む。代表的な成果として、延長前に成果を上げていた、伊藤・長谷川らが中心となった「メカノケミストリーを用いた固体反応」の開発 (K. Kubota, T. Seo, K. Koide, Y. Hasegawa, H. Ito, *Nature Commun.* **2019**, *10*, 111)。が大きな発展を遂げている。ボールミルを用いることで、これまで溶液中では不可能出会った多く反応を開発した。島田らのアドバイスのもと、無機化合物である圧電材料を共存させた状態で、電気化学的な有機合成反応を実施することに世界ではじめて成功した (Kubota, K.*; Pang, Y.; Miura, A.; Ito, H.* *Science* **2019**, *366*, 1500; Pang, Y.; Lee, J. W.; Kubota, K.*; Ito, H.* *Angew. Chem. Int. Ed.* **2020**, *59*, 22570)。さらに固体でのみはじめて発現するモノクロスカップリングに成功した (Seo, T.; Kubota, K.*; Ito, H.* *J. Am. Chem. Soc.* **2020**, *142*, 9884)。この固体クロスカップリングは溶媒が不溶であるため、その特徴を生かして、不溶性固体のクロスカップリング技術の確立に成功した。さらに有機合成化学分野で120年以上使われているGrignard試薬を世界で始めてボールミルで合成する事に成功した (Takahashi, R.; Hu, A.; Gao, P.; Gao, Y.; Pang, Y.; Seo, T.; Maeda, S.; Jiang, J.; Takaya, H.; Kubota, K.*; Ito, H.* *Nature Commun.* **2021**, *12*, 6691)。この論文はアクセス数2万3000回を超え、*Nature Commun* 誌における2021 Top 25 Chemistry and Materials Sciences Articles のトップにあげられた。大村らは、シクロプロパン環形成反応を触媒する新規金属依存酵素を見出した (Li, X.; Shimaya, R.; Dairi, T.; Chang, W.-C.; Ogasawara, Y. *Angew. Chem. Int. Ed. Engl.* **2022**, *61*, e202113189)。長谷川らは、プロペラ型レアアース分子からの強らせん発光を実現し、高集積型の円偏光発光体を新規開発した (Kitagawa, Y.; Wada, S.; Islam, M. D. J.; Saita, K.; Gon, M.; Fushimi, K.; Tanaka, K.; Maeda, S.; Hasegawa, Y. *Communications Chemistry* **2020**, *3*, 1)。島田らはナノレベルで厚さを制御した自立型COF膜の作製に成功した (Kato, M.; Ota, R.; Endo, T.; Yanase, T.; Nagahama, T.; Shimada, T. *ACS Appl. Nano Mater.* **2022**, *5*, 2367)。いくつかの研究論文が特に重要度の高い研究成果であると評価され、ジャーナルカバーアートに採択された(右下図)。また、大村らは、本ラボでの研究成果が契機となり、2022年より科学研究費補助金基盤研究(S)「天然ペプチド系化合物に構造・機能多様性をもたらす新規酵素・生合成機構の解明と応用」に採択された。また伊藤らは2022年より科学研究費補助金基盤研究(A)「官能化および不斉シリルボランを利用した非対称有機ケイ素化合物の精密合成」および挑戦的研究(開拓)「化学反応で駆動する結晶性錯体分子触媒エンジンの開発」に採択されている。長谷川らは2022年より新学術領域研究(研究領域提案型)「水圏環境で生体分子と会合体を形成する発光性希土類錯体の開発」に採択されており、外部資金の獲得が活発に行われている。

研究活動が当該研究センターに寄与・貢献した点など

延長期間においても、研究メンバーが本研究ラボを積極的に活用した効果は絶大で、それぞれの研究に関して飛躍的な進展が見られた。特に有機・材料・生物化学分野において、過去3年間の研究成果のインパクトは大きく、本センターの知名度を、引き続き全国的、国際的に高めることができたといえる。本ラボの存在は、外部資金の獲得にも大きく貢献している。また、本ラボでの活動を通じて、学士・修士課程のみならず、引き続き多くの博士課程学生を輩出できたことも、教育面における大きな成果であるといえる。

○本研究における研究業績

研究代表者・ 分担者氏名	論文、著書、工業所有権等、招待講演など
伊藤肇	<ol style="list-style-type: none"> 1. K. Kubota, K. Kondo, T. Seo, H. Ito, Insight into the Reactivity Profile of Solid-State Aryl Bromides in Suzuki-Miyaura Cross-Coupling Reactions Using Ball Milling. <i>Synlett</i>, 33, 898-902 (2022). 2. K. Kubota, T. Endo, M. Uesugi, Y. Hayashi, H. Ito, Solid-State C-N Cross-Coupling Reactions with Carbazoles as Nitrogen Nucleophiles Using Mechanochemistry. <i>ChemSusChem</i> 15, e202102132 (2022). 3. Y. Gao, C. Feng, T. Seo, K. Kubota, H. Ito, Efficient access to materials-oriented aromatic alkynes via the mechanochemical Sonogashira coupling of solid aryl halides with large polycyclic conjugated systems. <i>Chem. Sci.</i> 13, 430-438 (2022). 4. T. Yanase, H. Tanoguchi, N. Sakai, M. Jin, I. Yamane, M. Kato, H. Ito, T. Nagahama, T. Shimada, Single Crystal Growth of pi-Conjugated Large Molecules without Solubilizing Alkyl Chains via the Naphthalene Flux Method. <i>Crystal Growth & Design</i> 21, 4683-4689 (2021). 5. T. Takeuchi, R. Shishido, K. Kubota, H. Ito, Synthesis of hydrosilyboronates via the monoborylation of a dihydrosilane Si-H bond and their application for the generation of dialkylhydrosilyl anions. <i>Chem. Sci.</i> 12, 11799-11804 (2021). 6. R. Takahashi, T. Seo, K. Kubota, H. Ito, Palladium-Catalyzed Solid-State Polyfluoroarylation of Aryl Halides Using Mechanochemistry. <i>ACS Catal.</i> 11, 14803-14810 (2021). 7. R. Takahashi, A. Q. Hu, P. Gao, Y. P. Gao, Y. D. Pang, T. Seo, J. L. Jiang, S. Maeda, H. Takaya, K. Kubota, H. Ito, Mechanochemical synthesis of magnesium-based carbon nucleophiles in air and their use in organic synthesis. <i>Nat. Commun.</i> 12, 6691 (2021). 8. T. Seo, N. Toyoshima, K. Kubota, H. Ito, Tackling Solubility Issues in Organic Synthesis: Solid-State Cross-Coupling of Insoluble Aryl Halides. <i>J. Am. Chem. Soc.</i> 143, 6165-6175 (2021). 9. Y. Ozawa, K. Endo, H. Ito, Regio- and Stereoselective Synthesis of Multi-Alkylated Allylic Boronates through Three-Component Coupling Reactions between Allenes, Alkyl Halides, and a Diboron Reagent. <i>J. Am. Chem. Soc.</i> 143, 13865-13877 (2021). 10. K. Kubota, N. Toyoshima, D. Miura, J. L. Jiang, S. Maeda, M. Jin, H. Ito, Introduction of a Luminophore into Generic Polymers via Mechanoradical Coupling with a Prefluorescent Reagent. <i>Angew. Chem. Int. Ed.</i> 60, 16003-16008 (2021). 11. K. Kubota, D. Miura, T. Takeuchi, S. Osaki, H. Ito, Synthesis of Chiral alpha-Amino Tertiary Boronates via the Catalytic Enantioselective Nucleophilic Borylation of Dialkyl Ketimines. <i>ACS Catal.</i> 11, 6733-6740 (2021). 12. K. Kato, T. Seki, H. Ito, (9-Isocyanoanthracene)gold(I) Complexes Exhibiting Two Modes of Crystal Jumps by Different Structure Change Mechanisms. <i>Inorg. Chem.</i> 60, 10849-10856 (2021). 13. M. Jin, R. Ando, M. J. Jellen, M. A. Garcia-Garibay, H. Ito, Encapsulating N-Heterocyclic Carbene Binuclear Transition-Metal Complexes as a New Platform for Molecular Rotation in Crystalline Solid-State. <i>J. Am. Chem. Soc.</i> 143, 1144-1153 (2021). 14. M. Jin, R. Ando, H. Ito, Distinct Fold-Mode Formation of Crystalline Cu(I) Helical Coordination Polymers with Alternation of the Solid-State Emission Using Shape of the Counter Anions. <i>Inorg. Chem.</i>, (2021). 15. H. Iwamoto, Y. Ozawa, Y. Takenouchi, T. Imamoto, H. Ito, Backbone-Modified C-2-Symmetrical Chiral Bisphosphine TMSQuinoxP*: Asymmetric Borylation of Racemic Allyl Electrophiles. <i>J. Am. Chem. Soc.</i> 143, 6413-6422 (2021). 16. K. Hayama, R. Takahashi, K. Kubota, H. Ito, Copper(I)-Catalyzed Stereoselective Silylative Dearomatization of Indoles and Pyrroles using Silylboronates. <i>Chem. Lett.</i> 50, 289-292 (2021). 17. R. Ando, M. Jin, H. Ito, Charge-transfer crystal with segregated packing structure constructed with hexaarylbenzene and tetracyanoquinodimethane. <i>Crystengcomm</i> 23, 5564-5568 (2021). 18. S. Akiyama, N. Oyama, T. Endo, K. Kubota, H. Ito, A Copper(I)-Catalyzed Radical-Relay

- Reaction Enabling the Intermolecular 1,2-Alkylborylation of Unactivated Olefins. *J. Am. Chem. Soc.* **143**, 5260-5268 (2021).
19. R. Takahashi, K. Kubota, H. Ito, Air- and moisture-stable Xantphos-ligated palladium dialkyl complex as a precatalyst for cross-coupling reactions. *Chem. Commun.* **56**, 407-410 (2020).
 20. J. Taguchi, S. Matsuura, T. Seki, H. Ito, Synthesis and Tunable Optical Properties of C,N-Chelated Borate Luminophores Derived from Potassium Acyltrifluoroborates. *Chemistry-a European Journal* **26**, 2450-2455 (2020).
 21. R. Shishido, M. Uesugi, R. Takahashi, T. Mita, T. Ishiyama, K. Kubota, H. Ito, General Synthesis of Trialkyl- and Dialkylarylsilylboranes: Versatile Silicon Nucleophiles in Organic Synthesis. *J. Am. Chem. Soc.* **142**, 14125-14133 (2020).
 22. T. Seo, K. Kubota, H. Ito, Selective Mechanochemical Monoarylation of Unbiased Dibromoarenes by in Situ Crystallization. *J. Am. Chem. Soc.* **142**, 9884-9889 (2020).
 23. T. Seki, N. Toyoshima, H. Ito, Mixed crystal formation of two gold isocyanide complexes with various ratios for continuous tuning of photophysical properties. *Dalton Trans.* **49**, 2073-2076 (2020).
 24. T. Seki, K. Ida, H. Sato, S. Aono, S. Sakaki, H. Ito, Auophilicity-Mediated Construction of Emissive Porous Molecular Crystals as Versatile Hosts for Liquid and Solid Guests. *Chemistry-a European Journal* **26**, 735-744 (2020).
 25. T. Seki, C. Feng, K. Kashiya, S. Sakamoto, Y. Takasaki, T. Sasaki, S. Takamizawa, H. Ito, Photoluminescent Ferroelastic Molecular Crystals. *Angew. Chem. Int. Ed.* **59**, 8839-8843 (2020).
 26. Y. D. Pang, J. W. Lee, K. Kubota, H. Ito, Solid-State Radical C-H Trifluoromethylation Reactions Using Ball Milling and Piezoelectric Materials. *Angew. Chem. Int. Ed.* **59**, 22570-22576 (2020).
 27. K. Omoto, T. Nakae, M. Nishio, Y. Yamanoi, H. Kasai, E. Nishibori, T. Mashimo, T. Seki, H. Ito, K. Nakamura, N. Kobayashi, N. Nakayama, H. Goto, H. Nishihara, Thermosalience in Macrocyclic-Based Soft Crystals via Anisotropic Deformation of Disilanyl Architecture. *J. Am. Chem. Soc.* **142**, 12651-12657 (2020).
 28. K. Kubota, R. Takahashi, M. Uesugi, H. Ito, A Glove-Box- and Schlenk-Line-Free Protocol for Solid-State C-N Cross-Coupling Reactions Using Mechanochemistry. *ACS Sustainable Chemistry & Engineering* **8**, 16577-16582 (2020).
 29. K. Kubota, H. Ito, Mechanochemical Cross-Coupling Reactions. *Trends in Chemistry* **2**, 1066-1081 (2020).
 30. M. Jin, S. Yamamoto, T. Seki, H. Ito, M. A. Garcia-Garibay, Anisotropic Thermal Expansion as the Source of Macroscopic and Molecular Scale Motion in Phosphorescent Amphidynamic Crystals. *Angew. Chem. Int. Ed.*, (2020).
 31. H. Iwamoto, Y. Hayashi, Y. Ozawa, H. Ito, Silyl-Group-Directed Linear-Selective Allylation of Carbonyl Compounds with Trisubstituted Allylboronates Using a Copper(I) Catalyst. *ACS Catal.* **10**, 2471-2476 (2020).
 32. K. Hayama, R. Kojima, K. Kubota, H. Ito, Synthesis of Chiral N-Heterocyclic Allylboronates via the Enantioselective Borylative Dearomatization of Pyrroles. *Org. Lett.* **22**, 739-744 (2020).
 33. S. Akiyama, S. Nomura, K. Kubota, H. Ito, Copper(I)-Catalyzed Boryl Substitution of 1-Trifluoromethyl Allenes for the Synthesis of 3-Boryl-Substituted 1,1-gem-Difluorodienes. *J. Org. Chem.* **85**, 4172-4181 (2020).
 34. J. Taguchi, T. Takeuchi, R. Takahashi, F. Masero, H. Ito, Concise Synthesis of Potassium Acyltrifluoroborates from Aldehydes through Copper(I)-Catalyzed Borylation/Oxidation. *Angew. Chem. Int. Ed.* **58**, 7299-7303 (2019).
 35. R. Shishido, I. Sasaki, T. Seki, T. Ishiyama, H. Ito, Direct Dimesitylborylation of Benzofuran Derivatives by an Iridium-Catalyzed C-H Activation with Silyldimesitylborane. *Chemistry-a European Journal* **25**, 12924-12928 (2019).
 36. T. Seo, T. Ishiyama, K. Kubota, H. Ito, Solid-state Suzuki-Miyaura cross-coupling reactions: olefin-accelerated C-C coupling using mechanochemistry. *Chem. Sci.* **10**, 8202-8210 (2019).
 37. T. Seki, T. Mashimo, H. Ito, Anisotropic strain release in a thermosalient crystal: correlation

- between the microscopic orientation of molecular rearrangements and the macroscopic mechanical motion. *Chem. Sci.* **10**, 4185-4191 (2019).
38. Y. Pang, T. Ishiyama, K. Kubota, H. Ito, Iridium(I)-Catalyzed C-H Borylation in Air by Using Mechanochemistry. *Chem. Eur. J.* **25**, 4654-4659 (2019).
39. K. Kubota, M. Uesugi, S. Osaki, H. Ito, Synthesis of 2-alkyl-2-boryl-substituted-tetrahydrofurans via copper(i)-catalysed borylative cyclization of aliphatic ketones. *Org. Biomol. Chem.* **17**, 5680-5683 (2019).
40. K. Kubota, R. Takahashi, H. Ito, Mechanochemistry allows carrying out sensitive organometallic reactions in air: glove-box-and-Schlenk-line-free synthesis of oxidative addition complexes from aryl halides and palladium(0). *Chem. Sci.* **10**, 5837-5842 (2019).
41. K. Kubota, T. Seo, K. Koide, Y. Hasegawa, H. Ito, Olefin-accelerated solid-state C-N cross-coupling reactions using mechanochemistry. *Nat. Commun.* **10**, 111 (2019).
42. K. Kubota, Y. Pang, A. Miura, H. Ito, Redox reactions of small organic molecules using ball milling and piezoelectric materials. *Science* **366**, 1500-1504 (2019).
43. M. Kato, H. Ito, M. Hasegawa, K. Ishii, Soft Crystals: Flexible Response Systems with High Structural Order. *Chemistry-a European Journal* **25**, 5105-5112 (2019).
44. M. Jin, S. Yamamoto, T. Seki, H. Ito, M. A. Garcia-Garibay, Anisotropic Thermal Expansion as the Source of Macroscopic and Molecular Scale Motion in Phosphorescent Amphidynamic Crystals. *Angew. Chem. Int. Ed.* **58**, 18003-18010 (2019).
45. H. Iwamoto, K. Endo, Y. Ozawa, Y. Watanabe, K. Kubota, T. Imamoto, H. Ito, Copper(I)-Catalyzed Enantioconvergent Borylation of Racemic Benzyl Chlorides Enabled by Quadrant-by-Quadrant Structure Modification of Chiral Bisphosphine Ligands. *Angew. Chem. Int. Ed.* **58**, 11112-11117 (2019).
46. T. Isono, N. Kawakami, K. Watanabe, K. Yoshida, I. Otsuka, H. Mamiya, H. Ito, T. Yamamoto, K. Tajima, R. Borsali, T. Satoh, Microphase separation of carbohydrate-based star-block copolymers with sub-10 nm periodicity. *Polymer Chemistry* **10**, 1119-1129 (2019).
47. Y. Hasegawa, T. Matsui, Y. Kitagawa, T. Nakanishi, T. Seki, H. Ito, Y. Nakasaka, T. Masuda, K. Fushimi, Near-IR Luminescent Yb-III Coordination Polymers Composed of Pyrene Derivatives for Thermostable Oxygen Sensors. *Chemistry-a European Journal* **25**, 12308-12315 (2019).
48. S. Aono, T. Seki, H. Ito, S. Sakaki, Dependence of Absorption and Emission Spectra on Polymorphs of Gold(I) Isocyanide Complexes: Theoretical Study with QM/MM Approach. *J Phys Chem C* **123**, 4773-4794 (2019).
49. S. Akiyama, K. Kubota, M. S. Mikus, P. H. S. Paioti, F. Romiti, Q. H. Liu, Y. B. Zhou, A. H. Hoveyda, H. Ito, Catalytic Enantioselective Synthesis of Allylic Boronates Bearing a Trisubstituted Alkenyl Fluoride and Related Derivatives. *Angew. Chem. Int. Ed.* **58**, 11998-12003 (2019).
1. M. Tsurui, Y. Kitagawa, S. Shoji, H. Ohmagari, M. Hasegawa, M. Gon, K. Tanaka, M. Kobayashi, T. Taketsugu, K. Fushimi, Y. Hasegawa, Asymmetric Lumino-Transformer: Circularly Polarized Luminescence of Chiral Eu(III) Coordination Polymer with Phase-Transition Behavior, *J. Phys. Chem. B* **126**, 3799-3807 (2022).
2. J. Hayashi, S. Shoji, Y. Kitagawa, Y. Hasegawa, Amorphous lanthanide complexes for organic luminescent materials, *Coord. Chem. Rev.* **467**, 214607 (2022).
3. Y. Kitagawa, R. Moriake, T. Akama, K. Saito, K. Aikawa, S. Shoji, K. Fushimi, M. Kobayashi, T. Taketsugu, Y. Hasegawa, Effective Photosensitization in Excited-State Equilibrium: Brilliant Luminescence of Tb^{III} Coordination Polymers Through Ancillary Ligand Modifications, *ChemPlusChem*, e202200236 (2022).
4. P. P. Ferreira da Rosa, Y. Kitagawa, S. Shoji, H. Oyama, K. Imaeda, N. Nakayama, K. Fushimi, H. Uekusa, K. Ueno, H. Goto, Y. Hasegawa, Preparation of photonic molecular trains via soft-crystal polymerization of lanthanide complexes, *Nature Commun.*, **13**, 3660 (2022).
5. Y. Hasegawa, S. Shoji, Y. Kitagawa, Luminescent Eu(III)-based Coordination Polymers for Photonic Materials, *Chem. Lett.* **51**, 185-196 (2022).

6. Y. Hasegawa, Y. Kitagawa, *J. Photochem. Photobio. C, Photochem. Rev. Luminescent Lanthanide Coordination Polymers with Transformative Energy Transfer Processes for Physical and Chemical Sensing Applications*, 100485 (2022).
7. Y. Kitagawa, M. Tsurui, Y. Hasegawa, Bright red emission with high color purity from Eu(III) complexes with π -conjugated polycyclic aromatic ligands and their sensing applications, *RSC Adv.* **12**, 810-821 (2022).
8. Y. Kitagawa, A. Naito, K. Aikawa, K. Shima, S. Shoji, K. Fushimi, Y. Hasegawa, Tribo-Excited Chemical Reaction Using an Eu(III) Complex with a Stacked Anthracene Framework, *Chem. Eur. J.* e202104401 (2022).
9. J. Hayashi, S. Shoji, Y. Kitagawa, K. Fushimi, Y. Hasegawa, Amide-bridged Eu(III) coordination polymer for stable luminescent glass material, *Mater. Lett.* **297**, 130012 (2021).
10. Y. Kitagawa, K. Matsuda, P. P. Ferreira da Rosa, K. Fushimi, Y. Hasegawa, Long-lived emission beyond 1000 nm: control of excited-state dynamics in a dinuclear Tb(III)-Nd(III) complex, *Chem. Comm.* **57**, 8047-8050 (2021).
11. H. Shirakura, Y. Manabe, C. Kasai, Y. Inaba, M. Tsurui, Y. Kitagawa, Y. Hasegawa, T. Yoneda, Y. Ide, Y. Inokuma, Isopyrazole-Masked Tetraketone: Tautomerism and Functionalization for Fluorescent Metal Ligands, *Eur. J. Inorg. Chem.*, 4345-4349 (2021).
12. T. Zhang, Y. Kitagawa, R. Moriake, P. P. Ferreira da Rosa, Md J. Islam, T. Yoneda, Y. Inokuma, K. Fushimi, Y. Hasegawa, Hybrid Eu(III) Coordination luminophore Standing on Silica Nanoparticles By Two Legs for Enhanced Luminescence, *Chem. Eur. J.* **27**, 14438-14443 (2021).
13. Md Jahidul Islam, Y. Kitagawa, P.-P. Ferreira da Rosa, Y. Hasegawa, Strong circularly polarized luminescence of mixed lanthanide coordination polymers with control of 4f electronic structures, *Dalton Trans.* **50**, 5433-5436 (2021).
14. Y. Kitagawa, P.-P. Ferreira da Rosa, Y. Hasegawa, Charge-transfer excited states of π - and 4f-orbitals for development of luminescent Eu(III) complexes, *Dalton Trans.*, **50**, 14978-14984 (2021).
15. Y. Kitagawa, A. Naito, K. Fushimi, Y. Hasegawa, First Tribo-excited Chemical Reaction of a Stacked Lanthanide Coordination Polymer with in Situ Reaction Monitor, *Chem. Eur. J.* **27**, 264 - 269 (2021).
16. P. P. Ferreira da Rosa, S. Miyazaki, H. Sakamoto, Y. Kitagawa, K. Miyata, T. Akama, M. Kobayashi, K. Fushimi, K. Onda, T. Taketsugu, Y. Hasegawa, Coordination Geometrical Effect on Ligand-to-Metal Charge Transfer-Dependent Energy Transfer Processes of Luminescent Eu(III) Complexes, *J. Phys. Chem. A* **125**, 209-217 (2021).
17. Y. Kitagawa, A. Naito, K. Fushimi, Y. Hasegawa, Bright Sky-Blue Fluorescence with High Color Purity: Assembly of Luminescent Diphenyl-Anthracene Lutetium-based Coordination Polymer, *RSC Adv.* **11**, 6604-6606 (2021).
18. T. Inoue, M. Tsurui, H. Yamagishi, Y. Nakazawa, N. Hamaguchi, S. Watanabe, Y. Kitagawa, Y. Hasegawa, Y. Yamamoto, H. Tsuji, Long-wavelength visible to near infrared photoluminescence from carbon-bridged styrylstilbene and thiadiazole conjugates in organic and aqueous media, *RSC Adv.* **11**, 6008-6013 (2021).
19. Y. Hasegawa, N. Sato, J. Hayashi, Y. Kitagawa, K. Fushimi, Thermo-Sensitive Eu(III) Coordination Polymers with Amorphous Networks, *ChemistrySelect* **6**, 2812 -2816 (2021).
20. M. Tsurui, Y. Kitagawa, K. Fushimi, M. Gon, K. Tanaka, Y. Hasegawa, Electronic strain effect on Eu(III) complexes for enhanced circularly polarized luminescence, *Dalton Trans.* **49**, 5352-5361 (2020).
21. T. Yoneda, C. Kasai, Y. Manabe, M. Tsurui, Y. Kitagawa, Y. Hasegawa, P. Sarkar, Y. Inokuma, Luminescent coordination polymers constructed from flexible, tetradetate diisopyrazole ligand and copper(I) halides, *Chem. Asian J.* **15**, 601-605 (2020).
22. Y. Inaba, T. Yoneda, Y. Kitagawa, K. Miyata, Y. Hasegawa, Y. Inokuma, Splitting and Reorientation of π -Conjugation by an Unprecedented Photo-Rearrangement Reaction, *Chem. Commun.* **56**, 348-351 (2020).
23. M. Kasai, Y. Sugioka, M. Yamamoto, T. Nagata, T. Nonomura, K. Asai, Y. Hasegawa, Characteristic Evaluation of Chameleon Luminophore Dispersed in Polymer,

- Sensors* **20**, 2623 (2020).
24. A. Urushizaki, T. Yumura, Y. Kitagawa, Y. Hasegawa, H. Imoto, K. Naka, Dithieno[3,4-b:3',4'-d]arsole: A Novel Class of Hetero[5]radialenes, *Eur. J. Org. Chem.* 0000 (2020).
25. Y. Kitagawa, M. Kumagai, K. Fushimi, Y. Hasegawa, Aggregation-induced emission of a Eu(III) complex via ligand-to-metal charge transfer, *Chem. Phys. Lett.* **749** 137437 (2020).
26. Y. Kitagawa, S. Wada, MD J. Islam, K. Saita, M. Gon, K. Fushimi, K. Tanaka, S. Maeda, Y. Hasegawa, Chiral lanthanide lumino-glass for a circularly polarized light security device, *Comms. Chem.* **3**, 119 (2020).
27. Y. Kitagawa, M. Kumagai, P-P. Ferreira da Rosa, K. Fushimi, Y. Hasegawa, Long-range LMCT coupling in Eu(III) coordination polymers for an effective molecular luminescent thermometer, *Chem. Eur. J* **27**, 264 – 269 (2020).
28. S. Miyazaki, K. Miyata, H. Sakamoto, F. Suzue, Y. Kitagawa, Y. Hasegawa, K. Onda, Dual Energy Transfer Pathways from an Antenna Ligand to Lanthanide Ion in Trivalent Europium Complexes through Phosphine-Oxide Bridges, *J. Phys. Chem. A* **124**, 33, 6601-6606 (2020).
29. Y. Hasegawa, K. Koide, M. Tsurui, Y. Kitagawa, T. Nakanishi, Y. Doi, Y. Hinatsu and K. Fushimi, Circularly polarized absorption and luminescence of semiconductor Eu-OCN nanocrystals in blue light region, *ChemPhysChem.* **21**, 2019-2024 (2020).
30. Y. Hasegawa, T. Sawanobori, Y. Kitagawa, S. Shoji, K. Fushimi, Y. Nakadaka, T. Masuda, I. Hisaki, An Europium (III) Luminophore with Pressure-Sensing Units: Effective Back Energy Transfer in Coordination Polymers with Hexadentate Porous Stable Networks, *ChemPlusChem.* **85**, 1989-1993 (2020).
31. S. Aoki, Y. Kitagawa, Y. Hasegawa, S. Sonoike, Y. Saga, M. Hatanaka, Evaluation of Zn²⁺ Coordination Structures in Chiral Zn²⁺ Complexes Based on Shape Measurement Factors: Relationships between Catalytic Activity and the Coordination Structure, *Eur. J. Inorg. Chem.* 4740-4751 (2019).
32. P-P. Ferreira da Rosa, Y. Kitagawa, Y. Hasegawa, Luminescent lanthanide complex with seven-coordination geometry, *Coord. Chem. Rev.* 213153 (2020).
33. Y. Kitagawa, F. Suzue, T. Nakanishi, K. Fushimi, T. Seki, H. Ito, Y. Hasegawa, Stacked nanocarbon photosensitizer for efficient blue light excited Eu(III) emission, *Communication Chemistry*, **3**, 3 (2020).
34. Y. Kitagawa, M. Tsurui, Y. Hasegawa, Steric and Electronic Control of Chiral Eu(III) Complexes for Effective Circularly Polarized Luminescence, *ACS Omega* **5**, 3786-3791 (2020).
35. Y. Kitagawa, M. Kumagai, T. Nakanishi, K. Fushimi and Y. Hasegawa, First aggregation-induced emission of a Tb(III) luminophore based on modulation of ligand– ligand charge transfer bands, *Dalton Trans.* **49**, 2431–2436 (2020).
36. Y. Kitagawa, M. Kumagai, P. P. Ferreira da Rosa, K. Fushimi and Y. Hasegawa, First demonstration of the π -f orbital interaction depending on the coordination geometry in Eu(III) luminophores, *Dalton Trans.* **49**, 3098-3101(2020).
37. Y. Kitagawa, M. Kumagai, T. Nakanishi, K. Fushimi and Y. Hasegawa, The Role of π -f Orbital Interactions in Eu(III) Complexes for an Effective Molecular Luminescent Thermometer, *Inorg. Chem.* **59**, 5865-5871 (2020).
38. Y. Shichibu, M. Zhang, T. Iwasa, Y. Ono, T. Taketsugu, S. Omagari, T. Nakanishi, Y. Hasegawa, K. Konishi, Photoluminescence Properties of [Core+exo]-Type Au₆ Clusters: Insights into the Effect of Ligand Environments on the Excitation Dynamics, *J. Phys. Chem. C* **123**, 6934-6939 (2019).
39. L. A. Galán, S. Wada, L. Cameron, A. N. Sobolev, Y. Hasegawa, E. Zysman-Colman, M. I. Ogden M. Massi, Photophysical investigation of near infrared emitting lanthanoid complexes incorporating tris(2-naphthoyl)methane as a new antenna ligand, *Dalton Trans.* **48**, 3768-3776 (2019).
40. Y. Hasegawa, Y. Kitagawa, Luminescent Lanthanide Complexes, Clusters, Coordination Polymers and Metal-organic Frameworks with Temperature-sensing properties, *J. Mater. Chem. C.* **7**, 7494-7511(2019).

41. M. Morisue, I. Ueno, K. Muraoka, S. Omagari, T. Nakanishi, Y. Hasegawa, T. Hikima, S. Sasaki, Perfluorophenyl-Directed Giant Porphyrin J-Aggregates, *Chem. Eur. J.* **25**, 7322–7329 (2019).
42. Y. Hasegawa, T. Matsui, Y. Kitagawa, T. Nakanishi, T. Seki, H. Ito, Y. Nakasaka, T. Masuda, K. Fushimi, Near-IR luminescent Yb(III) coordination polymers composed of pyrene derivatives for thermo-stable oxygen sensors, *Chem. Eur. J.* **25**, 12308–12315 (2019).
43. Y. Hirai, A. Kotani, H. Sakaue, Y. Kitagawa, Y. Hasegawa, Lifetimes of Lanthanide(III) Triboluminescence Excited by Aerodynamic Shock Waves, *J. Phys. Chem. C* **123**, 27251–27256 (2019).
44. Y. Hirai, P. P. Ferreira da Rosa, Y. Kitagawa, Y. Hasegawa, Thermal and crystallographic investigation of luminescent Eu(III) coordination polymers with dithiane and dioxane hexyl rings, *Chem. Lett.* **48**, 1544–1546 (2019).

大利 徹

1. W. Xiao, Y. Satoh, Y. Ogasawara, T. Dairi, Biosynthetic Gene Cluster of linaridin Peptides Contains Epimerase Gene. *ChemBioChem* **23**, e202100705 (2022).
2. X. Li, R. Shimaya, T. Dairi, W.-c. Chang, Y. Ogasawara, Identification of Cyclopropane Formation in the Biosyntheses of Hormaomycins and Belactosins: Sequential Nitration and Cyclopropanation by Metalloenzymes. *Angew. Chem. Int. Ed.* **61**, e202113189 (2022).
3. Y. Ogasawara, S. Umetsu, Y. Inahashi, K. Nonaka, T. Dairi, Identification of pulvomycin as an inhibitor of the futilosine pathway. *J. Antibiot.* **74**, 825 (2021).
4. Z. Feng, Y. Ogasawara, T. Dairi, Identification of the Peptide Epimerase MslH Responsible for D-amino Acid Introduction at the C-terminus of Ribosomal Peptides. *Chem. Sci.* **12**, 2567–2574 (2021).
5. T. Kamide, S. Takusagawa, N. Tanaka, Y. Ogasawara, Y. Kawano, I. Ohtsu, Y. Satoh, T. Dairi, High production of ergothioneine in *Escherichia coli* using the sulfoxide synthase from *Methylobacterium* strains. *J. Agric. Food Chem.* **68**, 6390–6394 (2020).
6. S. Hayashi, Y. Ogasawara, Y. Satoh, C. Maruyama, Y. Hamano, T. Dairi, Off-loading Mechanism of Products in Polyunsaturated Fatty Acid Synthases. *ACS Chem. Biol.* **15**, 651–656 (2020).
7. Y. Ogasawara, Y. Shimizu, Y. Sato, T. Yoneda, Y. Inokuma, T. Dairi. Identification of actinomycin D as a specific inhibitor of the alternative pathway of peptidoglycan biosynthesis. *J. Antibiot.* **73**, 125–127 (2020).
8. M. Naka, K. Ikeuchi, S. Hayashi, Y. Satoh, Y. Ogasawara, T. Dairi. Subtle control of carbon chain length in polyunsaturated fatty acid synthases. *ACS Chem. Biol.* **14**, 2553–2556 (2019).
9. Y. Ogasawara, Y. Nakagawa, C. Maruyama, Y. Hamano, T. Dairi, *In vitro* characterization of MitE and MitB: formation of *N*-acetylglucosaminyl-3-amino-5-hydroxybenzoyl-MmcB as a key intermediate in the biosynthesis of antitumor antibiotic mitomycins. *Bioorg. Med. Chem. Lett.* **29**, 2076–2078 (2019).
10. Y. Ogasawara, M. Shigematsu, S. Sato, H. Kato, T. Dairi, Involvement of Peptide Epimerization in Poly- γ -glutamic Acid Biosynthesis. *Org. Lett.* **21**, 3972–3975 (2019).
11. R. Feng, Y. Satoh, H. Morita, Y. Ogasawara, T. Dairi, Amino Acid Residues Recognizing Isomeric Glutamate Substrates in UDP-*N*-acetylmuramic acid-L-alanine-glutamate Synthetases. *ACS Chem. Biol.* **14**, 975–978 (2019).
12. S. Hayashi, M. Naka, K. Ikeuchi, M. Otsuka, K. Kobayashi, Y. Satoh, Y. Ogasawara, C. Maruyama, Y. Hamano, T. Ujihara, T. Dairi, Control mechanism for carbon chain length in polyunsaturated fatty acid synthases. *Angew. Chem. Int. Ed.* **58**, 6605–6610 (2019).

島田 敏宏

1. M. Kato, R. Ota, T. Endo, T. Yanase, T. Nagahama, T. Shimada, Free-standing Nanometer-Thick Covalent Organic Framework Films for Separating CO₂ and N₂

ACS Appl. Nanomaterials, 5, 2367–2374 (2022)

2. T. Yanase, H. Tanoguchi, N. Sakai, M. Jin, I. Yamane, M. Kato, H. Ito, T. Nagahama, T. Shimada,
Single Crystal Growth of π -Conjugated Large Molecules without Solubilizing Alkyl Chains via Naphthalene Flux Method
Crystal Growth and Design, 21, 4683-4689(2021).
3. H. Takehana, I. Yamane, T. Yanase, T. Nagahama, T. Shimada
Interaction between alkali metals and diamond: etching and charge states of NV centers
Carbon, 182 585-592 (2021).
4. W. Liu, C. Song, M. Kou, Y. Wang, Y. Deng, T. Shimada, L. Ye,
Fabrication of ultra-thin g-C₃N₄ nanoplates for efficient visible-light photocatalytic H₂O₂ production via two-electron oxygen reduction
Chem. Eng. J. 425, 130615 (2021).
5. I. Yamane, K.Sato, R. Ohtomo, T. Yanase, A. Miura, T. Nagahama, Y. Kamiya, T. Shimada,
Ultrahigh-Pressure Preparation and Catalytic Activity of MOF-Derived Cu Nanoparticles
Nanomaterials 11,1040 (2021).
6. Healing Sulfur Vacancies in Monolayer MoS₂ by High-Pressure Sulfur and Selenium Annealing: Implication for High-Performance Transistors
Yanase, Takashi; Uehara, Fumiya; Naito, Itsuki; Nagahama, Taro; Shimada, Toshihiro
ACS Applied Nanomaterial 3(10),10462-10469 (2020).
7. W. Liu, T. Yanase, N. Iwasa, S. Mukai, S. Iwamura, T. Nagahama, T. Shimada
Post-annealed graphitic carbon nitride nanoplates obtained by sugar-assisted exfoliation with improved visible-light photocatalytic performance
Journal of Colloid & Interface Science 567,369-378(2020)
8. W. Liu, T. Yanase, N. Iwasa, H. Koizumi, S. Mukai, S. Iwamura, T. Nagahama, T. Shimada
Sugar-assisted Mechanochemical Exfoliation of Graphitic Carbon Nitride for Enhanced Visible-light Photocatalytic Performance
International J. of Hydrogen Energy 44,8444-8455(2020)
9. W. Liu, N. Iwasa, S. Fujita, H. Koizumi, M. Yamaguchi, T. Shimada
Porous Graphitic Carbon Nitride Nanoplates Obtained by A Combined Exfoliation Strategy for Enhanced Visible Light Photocatalytic Activity
Applied Surface Science 499,143901 (2020).