アカデミックプログラム［B講演］ | 02. 理論化学・情報化学・計算化学：口頭B講演

2025年3月27日(木) 14:00 ~ 15:40 [C]C302(第2学舎 2号館 [3階] C302)

[[C]C302-2pm2] 02. 理論化学・情報化学・計算化学

座長：小林 正人、松林 伸幸

英語
14:00 ~ 14:20
[[C]C302-2pm2-01]
エレクトロクロミックポリマーの開発におけるデータサイエンスの導入

○趙 愛瑋[1,2]、Dines Chandra Santra[1]、永田 賢二[1]、櫻井 惇也[1]、出村 雅彦[1]、樋口 昌芳[1,2] (1. 国立研究開発法人物質・材料研究機構、2. 大阪大学大学院情報科学研究科)

英語
14:20 ~ 14:40
[[C]C302-2pm2-02]
Ab initio量子化学計算による三重項オリゴアセンの電子スピン緩和の解析

○御代川 克輝[1]、倉重 佑輝[1,2,3] (1. 京都大学、2. CREST, JST、3. FOREST, JST)

英語
14:40 ~ 15:00
[[C]C302-2pm2-03]
改良型３次元離散コサイン変換を組み込んだ最大エントロピー法によるMg_3BN_3高圧相と低圧相の新規の電子密度分布解析

○平口 英夫[1] (1. （公）日本技術士会)

日本語
15:00 ~ 15:20
[[C]C302-2pm2-04]
遺伝的アルゴリズムを活用した新物質探索とその合成法予測

○菊池 夏希[1]、中野 高毅[1]、山﨑 久嗣[1]、齋藤 信[1] (1. トヨタ自動車株式会社)

日本語
15:20 ~ 15:40
[[C]C302-2pm2-05]
量子化学計算データを活用した深層学習モデルによる質量スペクトル予測

○黒田 峻裕[1]、中谷 直輝[1] (1. 東京都立大学)

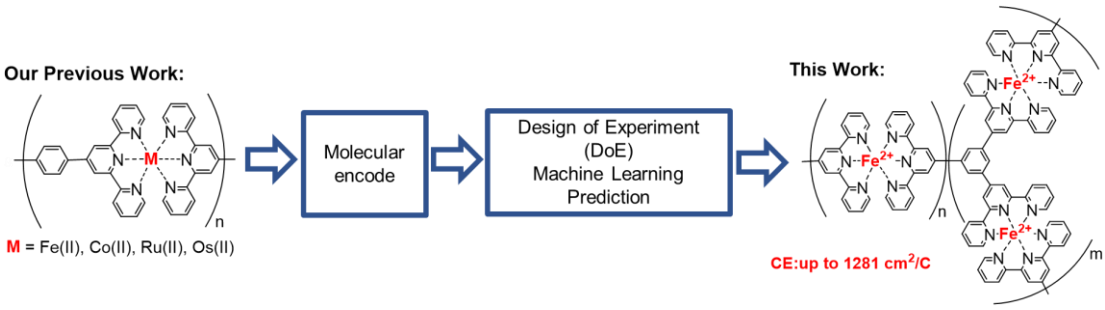
Introduction of Data Science in the Development of Electrochromic Polymers

(1 *National Institute for Materials Science*, 2*Graduate School of Information Science and Technology, Osaka University*) ○Aiwei Zhao[1,2], Dines Chandra Santra[1], Kenji Nagata[1], Junya Sakurai[1], Masahiko Demura[1], Masayoshi Higuchi[1,2]

Keywords: *electrochromic, coloration efficiency, data-science*

Materials informatics has emerged as a powerful tool for developing functional materials, offering the potential to streamline polymer synthesis with tailored properties, enhancing efficiency and specificity in material engineering. Machine learning, a subset of AI, uses algorithms to learn from data for predictions or decisions.[1] Bayesian optimization, widely used in materials science, optimizes objective functions and is particularly useful in hyperparameter tuning for machine learning models.[2]

In this presentation, we report our approach to discovering electrochromic (EC) metallo-supramolecular polymers (MSPs) using materials informatics. Four MSP components were evaluated, and selected combinations synthesized via an orthogonal table. Four machine learning model categories, including tree-based, kernel-based, linear, and neural network models, were trained to predict MSP properties effectively. This study achieved a high coloration efficiency of 1281 cm^2/C, which surpassed the values reported in our previous study.[3] This statistics-based approach demonstrates its effectiveness in the rapid identification of polymers with enhanced EC properties.



Acknowledgments：This research was supported by the Mirai project (JPMJMI21I4) from the Japan Science and Technology Agency (JST) and the Environment Research and Technology Development Fund (JPMEERF20221M02) from Environmental Restoration and Conservation Agency (ERCA).

1) Jordan, M. I.; Mitchell, T. M. Science, 2015, 349, 255. 2) Snoek, J.; Larochelle, H.; Adams, R. P. Adv. Neural Inf. Process. Syst., 2012, 25, 2951. 3) Hu, C.; Sato, T.; Zhang, J.; Moriyama, S.; Higuchi, M. *ACS Appl. Mater. Interfaces*, **2014**, *6*, 9118.

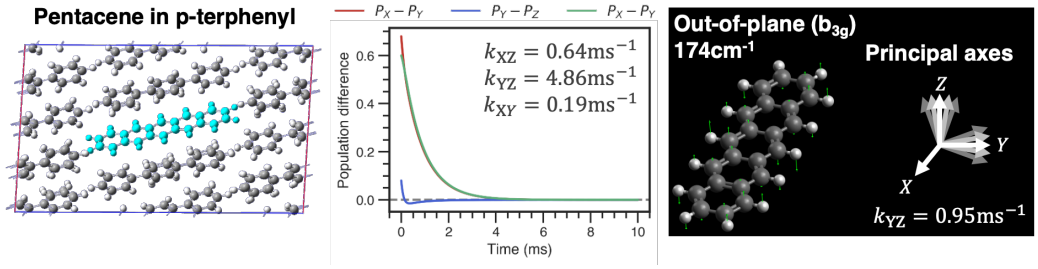
Ab Initio Study of Electron Spin-Lattice Relaxation of Triplet Oligoacenes

([1]*Graduate School of Science, Kyoto University*, [2]*CREST, JST,* [3]*FOREST, JST*) ○Katsuki Miyokawa,[1] Yuki Kurashige,[1,2,3]

Keywords: Electron Spin-Lattice Relaxation; Triplet State; Quantum Chemical Calculation; Redfield Theory; Oligoacene

Organic chromophores are converted into triplet species through photoexcitation followed by intersystem crossing (ISC). Focusing on the spin sublevel degrees of freedom, the anisotropy of ISC rate constants for each sublevel can generate a highly spin-polarized state, which can be achieved even under room temperature and low magnetic fields. Pentacene doped in p-terphenyl host crystal is often used as a polarizing source and applied to recently emerging quantum technologies such as (1) dynamic nuclear polarization (DNP)[1], which improve the sensitivity of NMR and MRI, (2) MASER[2], and (3) high sensitivity quantum sensing[3,4]. For the molecular design of the polarizing agents, it is important to understand the mechanism of spin polarization dynamics.

Electron spin-lattice relaxation (SLR) is the process of equilibration of the population between spin sublevels and determines the lifetime of spin polarization. Despite the importance of the SLR for polarizing agents, the anisotropy of the kinetic rates of the SLR depends on the surrounding complex vibrational environment, making its mechanism elusive. Recently, spin dynamics combined with ab initio quantum chemical calculations has been applied to the molecular qubit systems and successfully revealed the microscopic mechanism of the SLR[5]. Therefore, in this study, we conducted ab initio simulations of the SLR of triplet oligoacenes using the Redfield relaxation theory where the phonon frequencies and spin-phonon couplings were determined through quantum chemical calculations. By examining the contributions of each phonon mode to the rate constants, it was suggested that the experimentally observed characteristic fast SLR between T_Y and T_Z states in pentacene originates from the out-of-plane low-frequency vibrations.



[1] A. Henstra, P. Dirksen and W. Wenckebach, *Phys. Lett. A* **1988**, *134*, 134. [2] M. Oxborrow, J. D. Breeze and N. M. Alford, *Nature*, **2012**, *488*, 353. [3] A. Mena, S. L. Bayliss *et al.*, *Phys. Rev. Lett.* **2024**, *133*, 120801. [4] H. Singh, A. Ajoy *et al.*, arXiv:2402.13898. [5] A. Lunghi, S. Sanvito *et al.*, *Nat. Commun.* **2017**, *8*, 14620.

New Electron Density Distribution analysis of Mg_3BN_3 High Pressure Phase and Low Pressure Phase via Modified Three-Dimensional Discrete Cosine Transform and Maximum Entropy Method

([1]*The Institution of Professional Engineers, Japan*) ○Hideo Hiraguchi [1]

Keywords: Modified 3-dimensional Discrete Cosine Transform; Maximum Entropy method, Magnesium Boron Nitride

The previous researches[1),8)] have shown that the precise electron density distribution of Mg_3BN_3 high pressure phase (Mg_3BN_3(H), Orthorhombic Pmmm) which has both a center of symmetry and planes of symmetry can be calculated via the Maximum Entropy method[6),7)] incorporating the normal 3-dimensional Discrete Cosine Transform[5)] (3D-DCT) (Fig. 1). However, because Mg_3BN_3 low pressure phase (Mg3BN3(L)[2)-4)]) has a center of symmetry and a glide plane, the modified 3D-DCT[5)] is needed to calculate the electron density distribution. Therefore, in this research, the electron density distribution of Mg_3BN_3(L) (Fig. 3) has been calculated via the Maximum Entropy method incorporating the modified 3D-DCT by using a starting normal 3D-DCT map whose negative electron densities were replaced by 0.001 $Å^{-3}$ (Fig. 2).

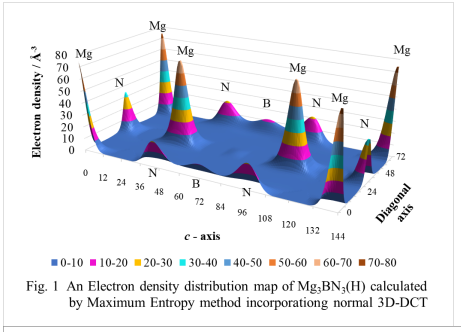


Fig. 1 An Electron density distribution map of Mg_3BN_3(H) calculated by Maximum Entropy method incorporationg normal 3D-DCT

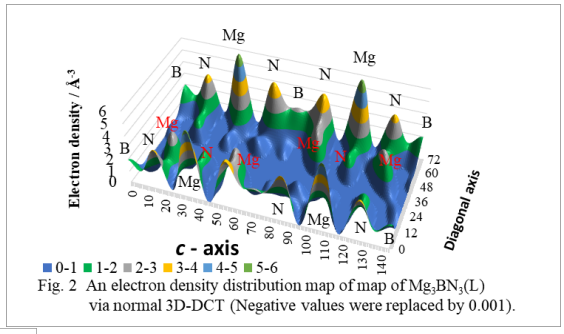


Fig. 2 An electron density distribution map of map of Mg_3BN_3(L) via normal 3D-DCT (Negative values were replaced by 0.001).

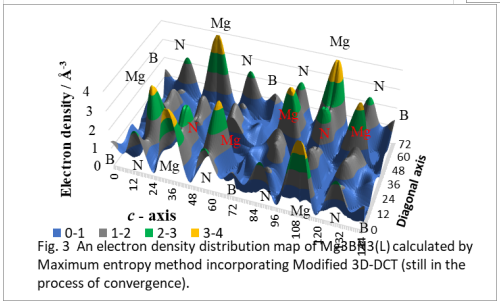


Fig. 3 An electron density distribution map of Mg_3BN_3(L) calculated by Maximum entropy method incorporating Modified 3D-DCT (still in the process of convergence).

1) H.Hiraguchi, The 104th CSJ Annual Meeting, H937-2pm-07, 2023. 2) H.HIRAGUCHI, O.SAKATA, H.HASHIZUME, A.TAKENAKA, O.FUKUNAGA.(1990). *J. Cryst. Soc.Jp.* OB-11, 32. 3) H. HIRAGUCHI, H. HASHIZUME, O. FUKUNAGA, A. TAKENAKA, M. SAKATA.(1991). *J. Appl. Cryst.* 24. 4) H. HIRAGUCHI, H. HASHIZUME, S. SASAKI, S. NAKANO, O. FUKUNAGA. (1993). *Acta Cryst.* B49. 5)H. HIRAGUCHI, (2021).*J.Appl.Cryst.* 6) M. Sakata & M. Sato, *Acta Cryst*. A46, 263-270, 1990. 7) M. Sakata, R. Mori, S. Kumazawa, M. Takata & H. Toraya, *J. Appl. Crvst.* 23, 526-534, 1990. 8) H.Hiraguchi, IUCr2023 in Melbourne.

遺伝的アルゴリズムを活用した新物質探索とその合成法予測

（トヨタ自動車株式会社[1]）○菊池 夏希[1]・中野 高毅[1]・山﨑 久嗣[1]・齋藤 信[1]
Novel Material Discovery and Synthesis Prediction using Genetic Algorithm ([1]*Toyota Motor Corporation*) ○Natsuki Kikuchi,[1] Koki Nakano,[1] Hisatsugu Yamasaki,[1] Makoto Saito[1]

With the advancement of Materials Informatics, the search for novel materials not listed in existing databases is accelerating. This study aims to establish simulation-based techniques for material discovery without synthesis attempts and to predict synthesis methods for new candidate materials. We developed a material exploration workflow for ionic conductive materials, combining genetic algorithms (GA) to generate novel crystal structures with the M3GNET machine learning force field for property prediction. Using this approach, we discovered metastable materials with high ionic conductivity (Fig. 1, 2). To validate these results, we attempted to synthesize the discovered materials by utilizing PIRO[1] for precursor search and heat treatment temperature prediction, but a more stable alternative material was produced, preventing successful experimental validation. Metastable candidate materials are often obtained in new material discovery, highlighting the importance of synthesis prediction techniques. This presentation will discuss the theory behind synthesis prediction tools, intermediate data, and directions for improvement.
Keywords : Materials Informatics, Evolutionary Algorithm, Novel Material Discovery, Computational Materials Science, Synthesis Route Prediction

マテリアルズ・インフォマティクスの進化と共に、材料データベースに掲載の無い新奇材料の探索が加速している。本研究は、シミュレーションによる試作レス材料探索技術と、新奇有望材料の合成法予測技術の確立を目標とする。

イオン伝導材料探索を題材に、遺伝的アルゴリズム(GA)による新しい結晶構造生成と機械学習力場 M3GNET による物性予測を組み合わせた材料探索フローを開発した。これを用いることで高いイオン伝導度を示す準安定材料を発見した(Fig. 1, 2)。本結果を検証すべく、PIRO[1]による試薬探索や熱処理温度予測を活用しながら、発見材料の合成を試みた。しかし、最安定な別材料が生成し合成実証には至らなかった。

新材料探索においては準安定な有望材料が得られる場合が多くあり、その合成法予測技術は重要と考えている。本発表では合成法予測ツールの理論や中間データを踏まえ、改善の方向性について議論する。

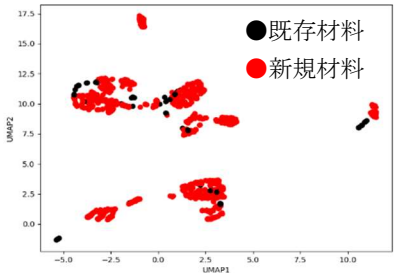


Fig. 1 GA/M3GNET で新材料を生成した結果 (UMAP)

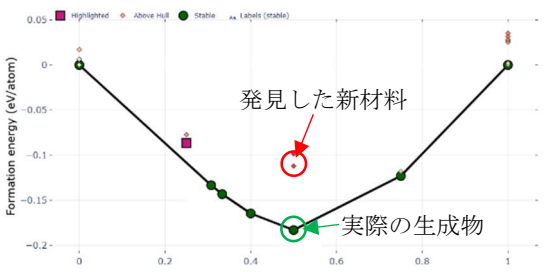


Fig. 2 凸包計算による安定性評価

[1] J. Am. Chem. Soc. 2021, 143, 9244−9259

量子化学計算データを活用した深層学習モデルによる質量スペクトル予測

（東京都立大学）◯黒田 峻裕・中谷 直輝
Mass Spectrum Prediction by Deep Learning Model Using Quantum Chemical Computational Data (*Tokyo Metropolitan University*) ◯Takahiro Kuroda, Naoki Nakatani

Accurate prediction of mass spectra (MS) used in the structural elucidation of organic molecules is a critical challenge, particularly from the perspectives of library augmentation and automated analysis of unknown samples. Machine learning-based predictive models have been developed to address this issue. In this study, we investigated whether the incorporation of quantitative information—such as bond energies and atomic charges, which were not considered in conventional models—calculated through quantum chemical methods could enhance prediction accuracy. Additionally, we computed the contributions of each descriptor.

For training, we utilized 5,043 EI-MS datasets from MassBank[1] along with their corresponding molecular structures. We examined an MPNN[3] model that included existing explanatory variables, such as atom types and connectivity, in addition to previously reported bond energy prediction models[2] and atomic charges in neutral and cationic states derived from quantum chemical calculations.

Although the model developed in this study did not improve prediction accuracy, we found that the combination of natural atomic charges in neutral molecules and the differences in natural atomic charges between neutral and cationic molecules exhibited high levels of contribution compared to existing explanatory variables. This suggests that incorporating information related to the ionization process into the explanatory variables is beneficial. Furthermore, analysis using molecular fingerprints revealed a very weak correlation between the number of similar molecules in the training data and prediction accuracy, indicating that prediction accuracy does not depend on specific molecular structures or substituents.
Keywords : Mass Spectra, Quantum Chemical Calculation, Deep Learning, Organic Molecules

有機分子の構造同定に利用される質量スペクトル(MS)の精密な予測は、スペクトルライブラリの補完や未知試料の自動解析の観点から重要な課題であり、機械学習による予測モデルが開発されている。本研究では、従来モデルでは考慮されていなかった結合エネルギーや原子電荷などの定量的な情報を量子化学計算によって算出し利用することで、予測精度の向上が可能かどうかの検証と各記述子の貢献度の計算を行った。

学習には、MassBank[1] の 5043 件の EI-MS データと、対応する分子構造を用い、説明変数に原子種・隣接関係などの既存のものに加えて、既報の結合エネルギー予測モデル[2]や量子化学計算から算出した中性・カチオン状態の原子電荷を考慮した MPNN[3] モデルについて検討を行った。

本研究のモデルでは、予測精度の改善には至らなかったが、既存の説明変数に比べて中性分子の自然電荷とカチオン分子との自然電荷差を組み合わせて使用することで、それぞれ高い貢献度を示すことが分かった。このことから、説明変数にイオン化の過程に関する情報を導入することが有用と考えられる。また、分子指紋を用いた解析の結果、学習データ中の類似分子数と予測精度との相関はきわめて弱く、特定の分子構造や置換基に予測精度が依存しないことも示された。

[1] *MassBank Open Access records on GitHub*. https://github.com/MassBank/MassBank-data (accessed 2024-05-14). [2] Shree S. S. V. *et al., Digital Discovery* **2023**, *2*, 1900. [3] Gilmer J. *et al., Proceedings of the 34th International Conference on Machine Learning* **2017**, *70*, 1263.