Battery Materials Group: Activity Report 2019

Battery Materials Group

[GL] M. Nakayama

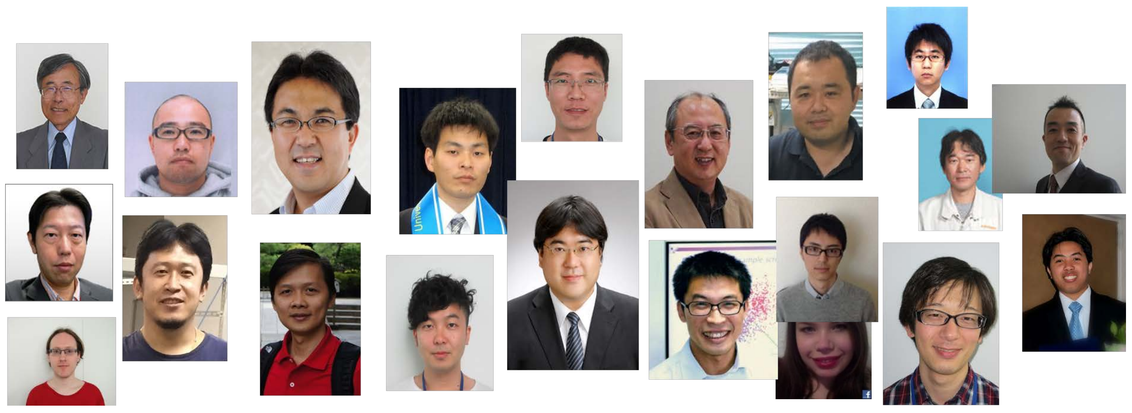
[SGL]Y. Tateyama, K. Sodeyama

(MN) masanobu@nitech.ac.jp

(YT) TATEYAMA.Yoshitaka@nitech.ac.jp

(KS) SODEYAMA.Keitaro@nims.go.jp

Group members and our aim



Group members (Including former members)

Development of large-scale rechargeable batteries are gaining attentions to realize electric vehicles with safety and sufficient cruising range (Fig.). Ideas of all ceramics batteries and multivalent batteries may solve technical issues above. Therefore, discovery of materials for these new-concept-battery (post LIB) are urgently requested today.

Efficient optimization of materials property is the aim of our studies, using high-throughput and informatics computational approaches. In particular, we focused on ion migration properties in solid and electrode|electrolyte interfacial reaction, since there are few studies, thus far, in terms of materials discovery. Total ~ 15 members are jointly studied currently.

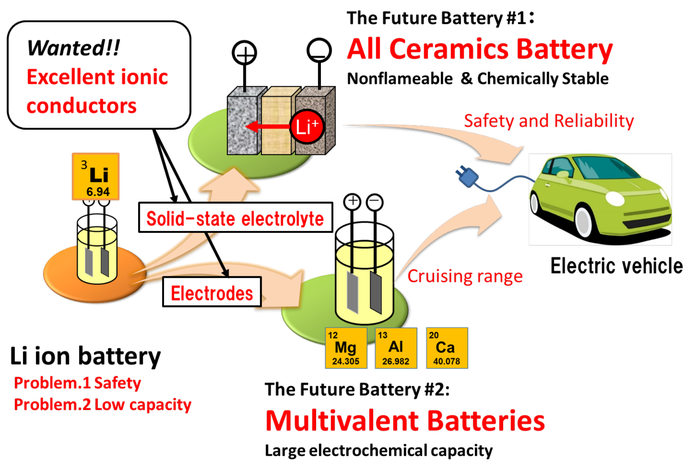
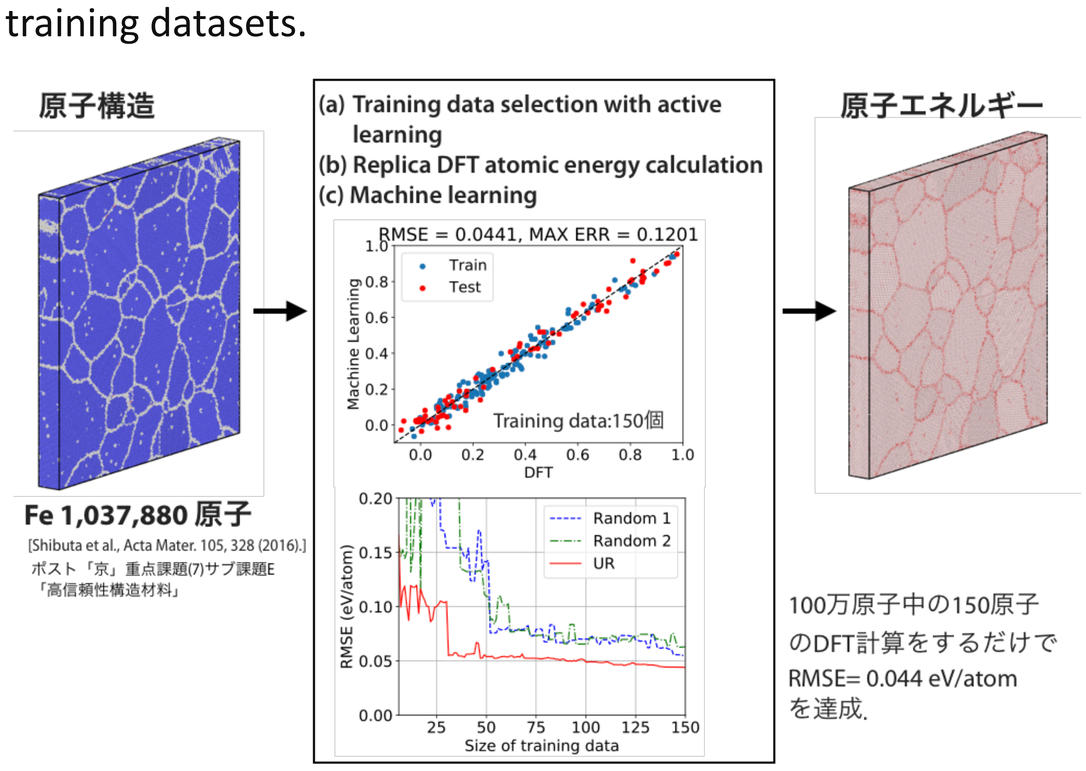


Fig. Post LIB

Acrivity report FY2019 -1

■Visualization of Interface Property by MI

Using MI and ab inito DFT, interfacial properties, such as energies, are visualized even for > 1M atom system. Active learning techniques are used for reasonable selection of training datasets.

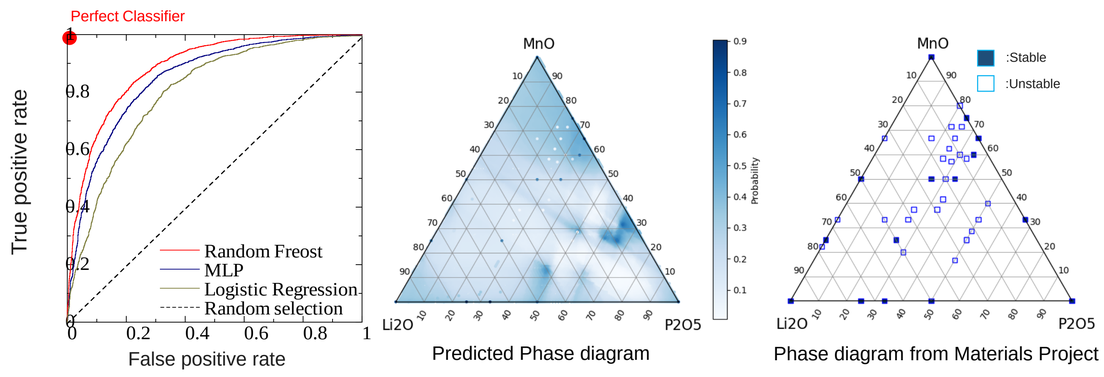


Acrivity report FY2019 -2

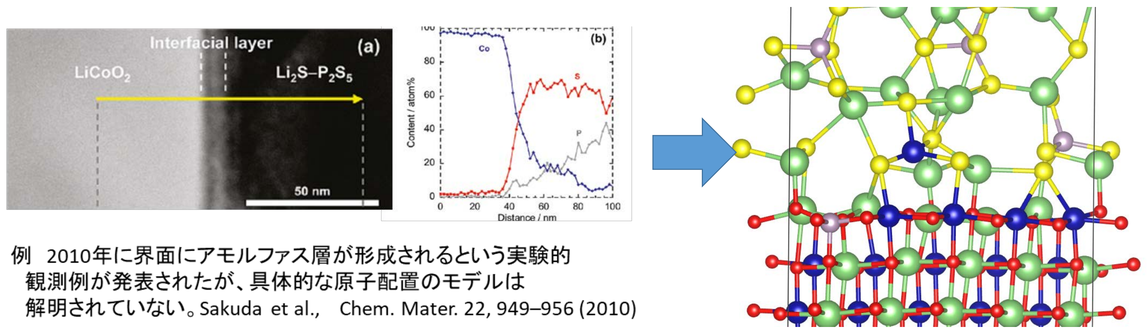
■ Prediction of Unknown Structure

New materials which is not registered in DB or interfacial structures are predicted by machine learning or meta-heuristic methods.

e.g.1） Materials property prediction only by composition



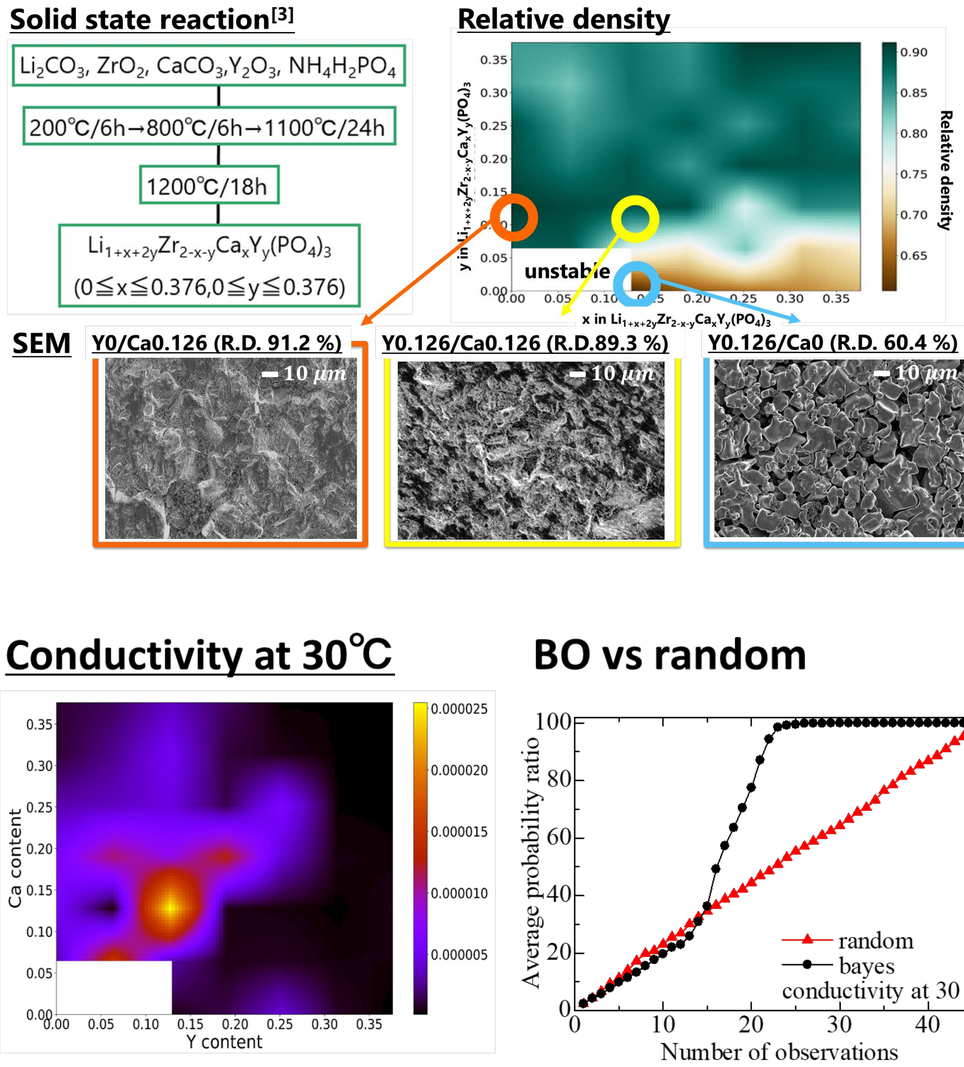
e.g.2) Ab initio prediction of interfacial structure by PSO



例 2010年に界面にアモルファス層が形成されるという実験的観測例が発表されたが、具体的な原子配置のモデルは解明されていない。Sakuda et al., Chem. Mater. 22, 949–956 (2010)

■Link to experiments with MI

Fast composition optimization is achieved by combined experimental and Bayes optimization techniques.

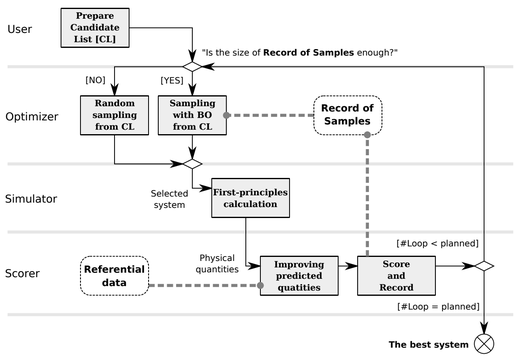


Magnet Materials Group

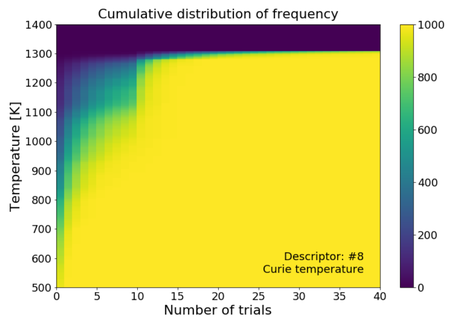
Magnet Materials Group
Takashi Miyake (GL) and Hiori Kino (SGL)
MIYAKE.Takashi@nims.go.jp

Optimizing Chemical Composition

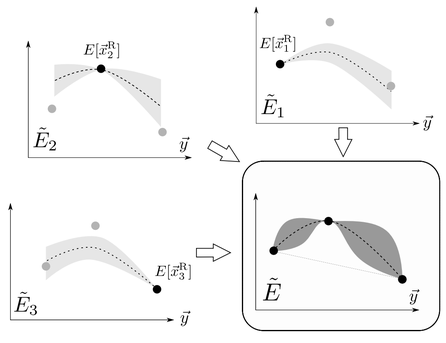
Fukazawa et al., Phys. Rev.. Mater **3**, 053807 (2019)

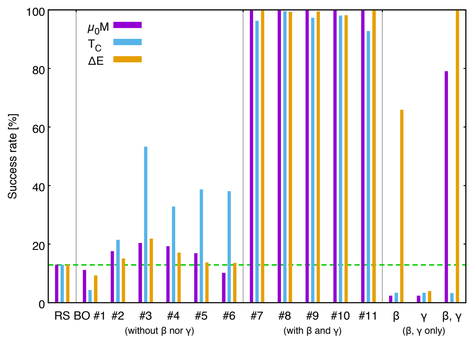


Bayesian Optimization



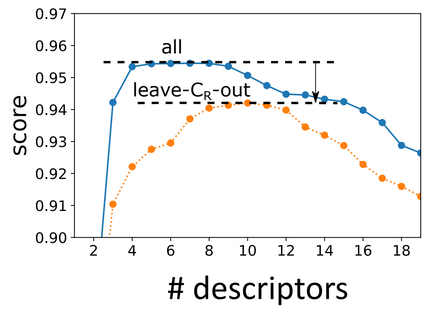
Data Assimilation





Analyzing Curie Temperature

① Exhaustive search

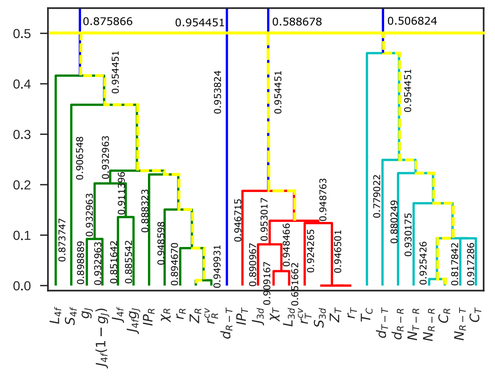


- 27 descriptors
- Kernel ridge regression

$$f(\vec{x}) = \sum_{i=1}^{N} \alpha_i k(\vec{x}^{(i)}, \vec{x})$$

n	score	descriptor(s)
2	0.870153	C_R,Z_T
3	0.942222	C_R,Z_R,Z_T
4	0.953386	J_{3d},C_R,Z_R,Z_T
5	0.954294	L_{3d},J_{3d},C_R,Z_R,Z_T
6	0.954391	L_{3d},J_{3d},χ_T,C_R,Z_R,Z_T
7	0.954452	L_{3d},J_{3d},χ_T,C_R,Z_R,Z_T,r_T^{cv}
8	0.954448	L_{3d},J_{3d},χ_T,IP_T,C_R,Z_R,Z_T,r_T^{cv}

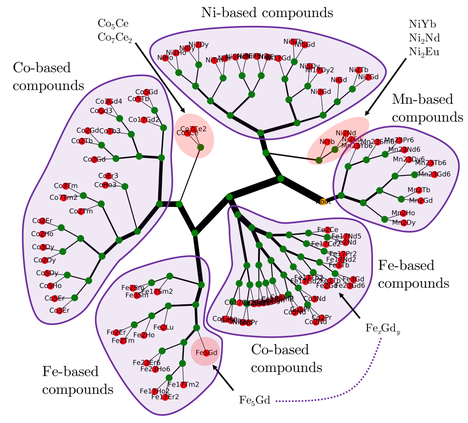
② Subgroup relevance analysis



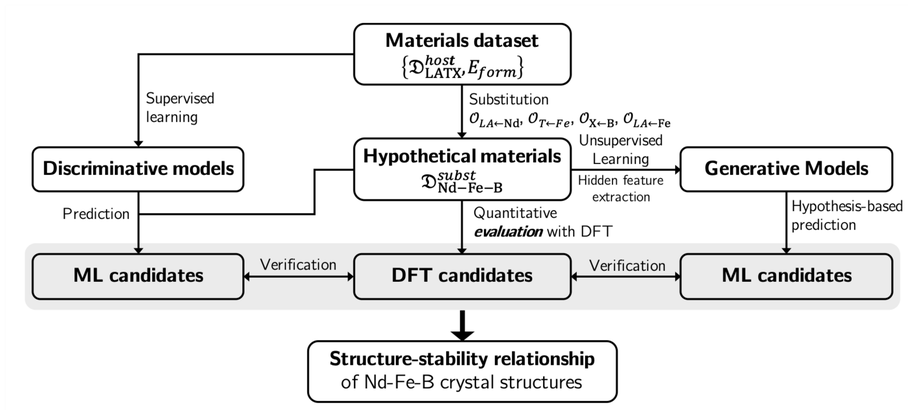
Dam et al., J. Phys. Soc. Jpn. **87**, 113801 (2018)

③ Dissimilarity voting machine

Nguyen et al., J. Phys. Mater **2**, 034009 (2019)

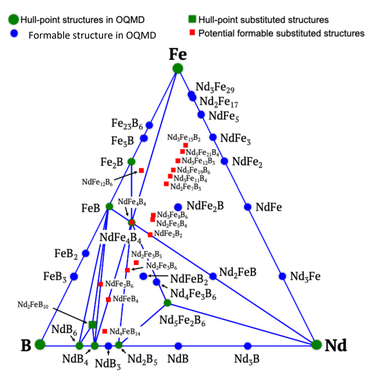


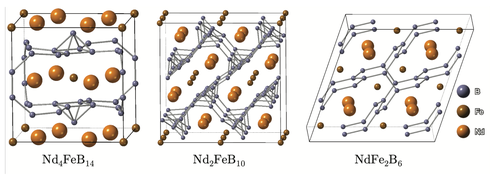
Search for Sub Phases



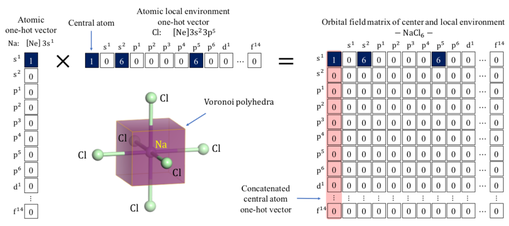
Computational Screening

Formula	E_f^{DFT} eV/atom	ΔE^{DFT} eV/atom	M μ_B (μ_B/Å^2)	$\Delta\tau$ Å
$Nd_4Fe_2B_{20}$	-0.522	-0.011	13.11 (0.050)	0.038
$NdFe_2B_6$	-0.473	0.008	3.30 (0.040)	0.150
$Nd_8Fe_2B_{28}$	-0.506	0.030	26.30 (0.063)	0.069
$NdFe_2B_2$	-0.343	0.046	4.41 (0.067)	0.085
$Nd_4Fe_4B_{16}$	-0.462	0.052	17.42 (0.073)	0.041
$Nd_4Fe_4B_{16}$	-0.455	0.060	18.73 (0.072)	0.050
$Nd_2Fe_3B_5$	-0.374	0.066	6.85 (0.055)	0.143
$Nd_2Fe_5B_4$	-0.284	0.069	10.31 (0.077)	0.206
$Nd_2Fe_8B_2$	-0.092	0.070	21.64 (0.134)	1.769
$Nd_2Fe_{24}B_{12}$	-0.231	0.072	45.56 (0.117)	1.012
$Nd_5Fe_{21}B_4$	-0.052	0.077	57.73 (0.140)	2.342
$Nd_5Fe_{19}B_6$	-0.115	0.080	50.02 (0.128)	1.820
$Nd_6Fe_{24}B_6$	-0.081	0.081	65.19 (0.135)	0.241
$Nd_3Fe_{13}B_2$	-0.027	0.081	36.12 (0.144)	2.961
$Nd_3Fe_{11}B_4$	-0.131	0.085	28.22 (0.122)	0.150
$Nd_4Fe_6B_{12}$	-0.375	0.088	16.02 (0.066)	0.132
$Nd_4Fe_{16}B_{16}$	-0.342	0.090	17.30 (0.048)	0.140
$Nd_2Fe_4B_4$	-0.297	0.092	7.25 (0.057)	0.142
$Nd_3Fe_8B_6$	-0.249	0.094	16.06 (0.079)	0.543
$Nd_4Fe_{14}B_6$	-0.147	0.096	35.04 (0.116)	0.209





Descriptor



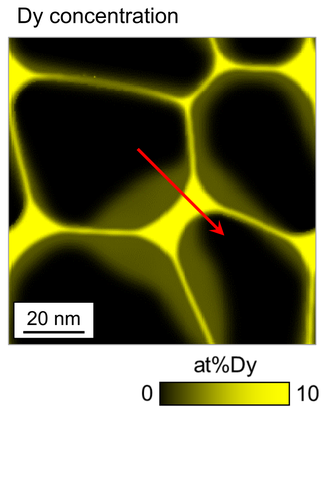
Orbital Field Matrix
Pham et al., STAM **18**, 756 (2017);
J. Chem. Phys. **148**, 204106 (2018)

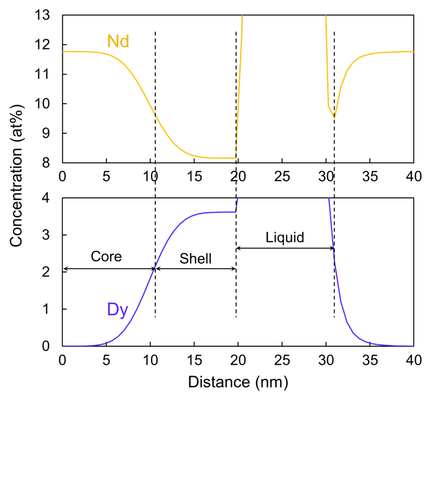
Virtual Screening

Model	Precision	Recall	f_1
KRR-model	0.533	0.534	0.376
LG-model	0.629	0.687	0.599
DT-model	0.704	0.676	0.687
GMM-model	**0.729**	**0.821**	**0.735**

Thermodynamic Parameters

Kim et al., Scripta Mater. **178**, 433 (2020)





Determine the Gibbs energy from the concentration profile

Progress in Thermal Management Materials Accelerated by MI

Thermal Management Materials Group

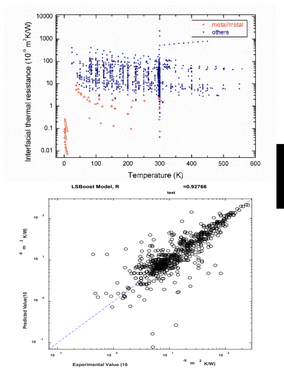
Y. Xu, Y. Shinohara, J. Shiomi, J. Morikawa, M. Goto, T. Baba

XU.Yibin@nims.go.jp SHINOHARA.Yoshikazu@nims.go.jp shiomi@photon.t.u-tokyo.ac.jp

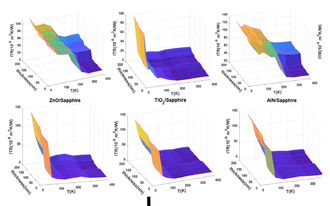
morikawa.j.aa@m.titech.ac.jp GOTO.Masahiro@nims.go.jp BABA.Tetsuya@nims.go.jp

Inorganic Composite with Ultra-Low Thermal Conductivity

We developed a machine learning model to predict interfacial thermal resistance (ITR) and designed the best combination of materials with high ITR. Inorganic composites with ultra-low thermal conductivity of 0.16 W/mK has been synthesized.

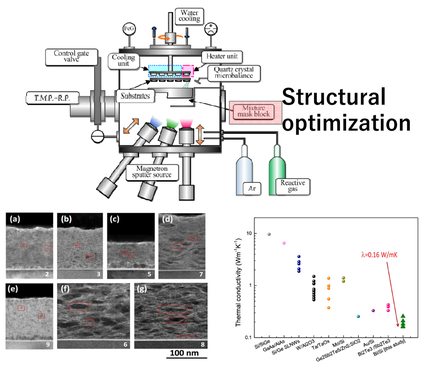


Machine learning model established to predict ITR



Best materials combination selected from 80,000 candidates

Y. Wu, et al., ACS Appl. Nano Mater. 2018,1,7, 3355

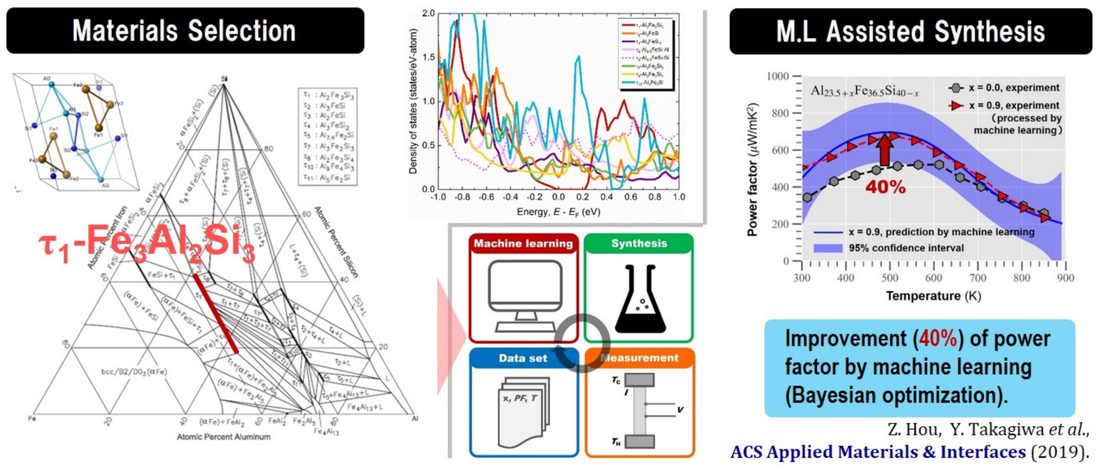


synthesis and evaluation of Bi/Si composites

Y. Xu ,et al., Patent application 2018-1587

New Thermoelectric Material

Materials selection was performed using theoretical calculations, and a novel Fe-Al-Si-based thermoelectric material was found. In addition, we succeeded in greatly improving the power factor using machine learning (Bayesian optimization).

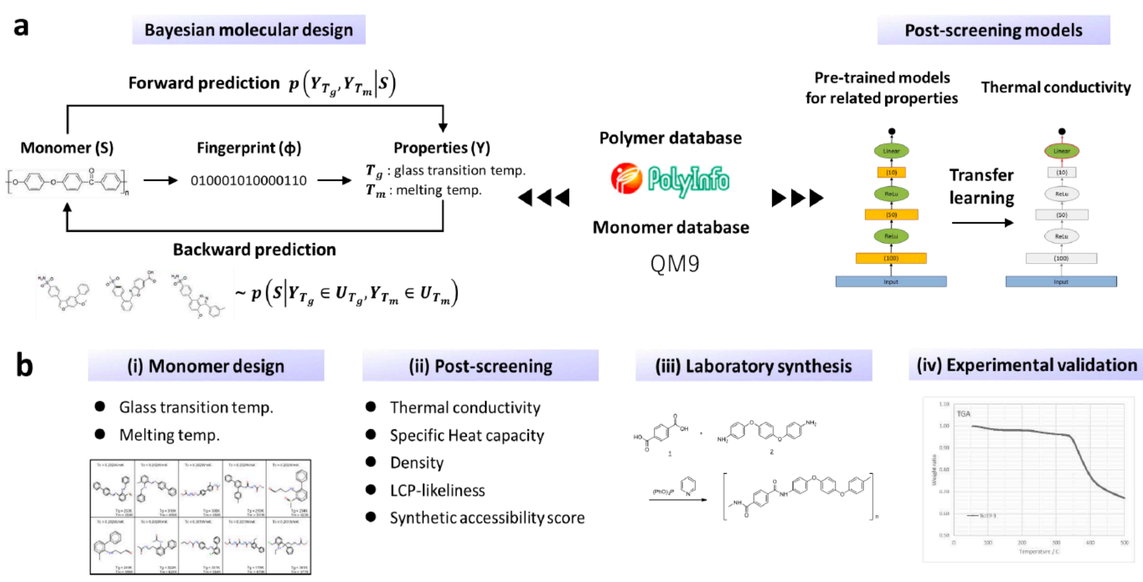


Improvement (40%) of power factor by machine learning (Bayesian optimization).

Z. Hou, Y. Takagiwa *et al.*, **ACS Applied Materials & Interfaces** (2019).

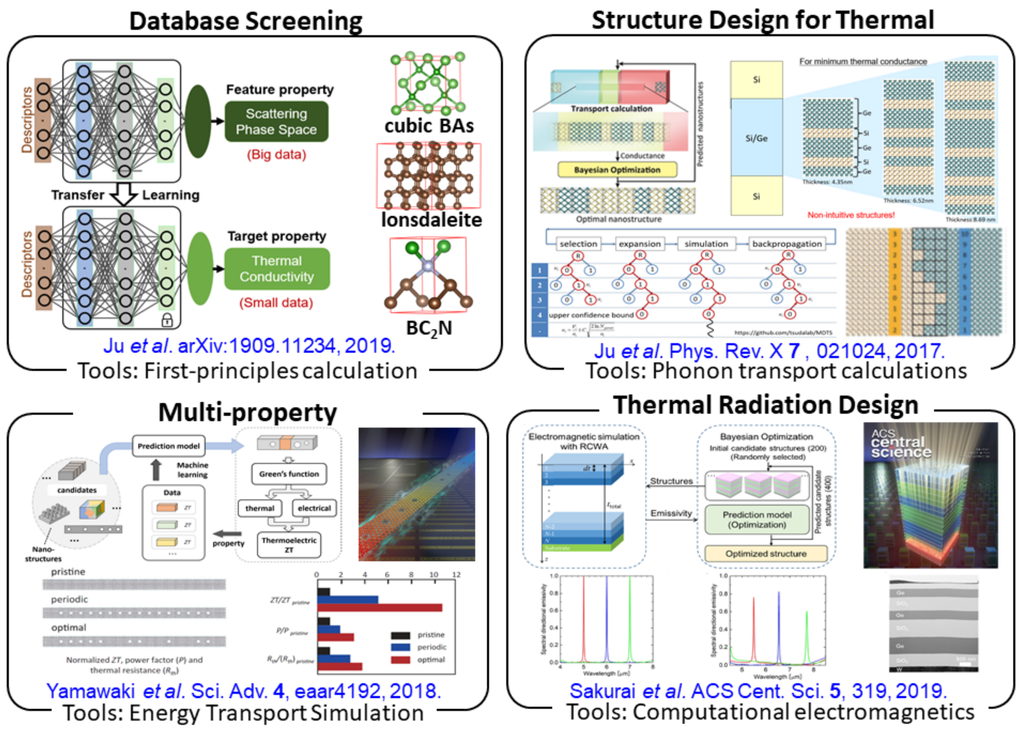
Polymers with High Thermal Conductivity

Python 3 , involving "transfer learning", accelerates efficient polymer design with desired properties even from a relatively small data set.

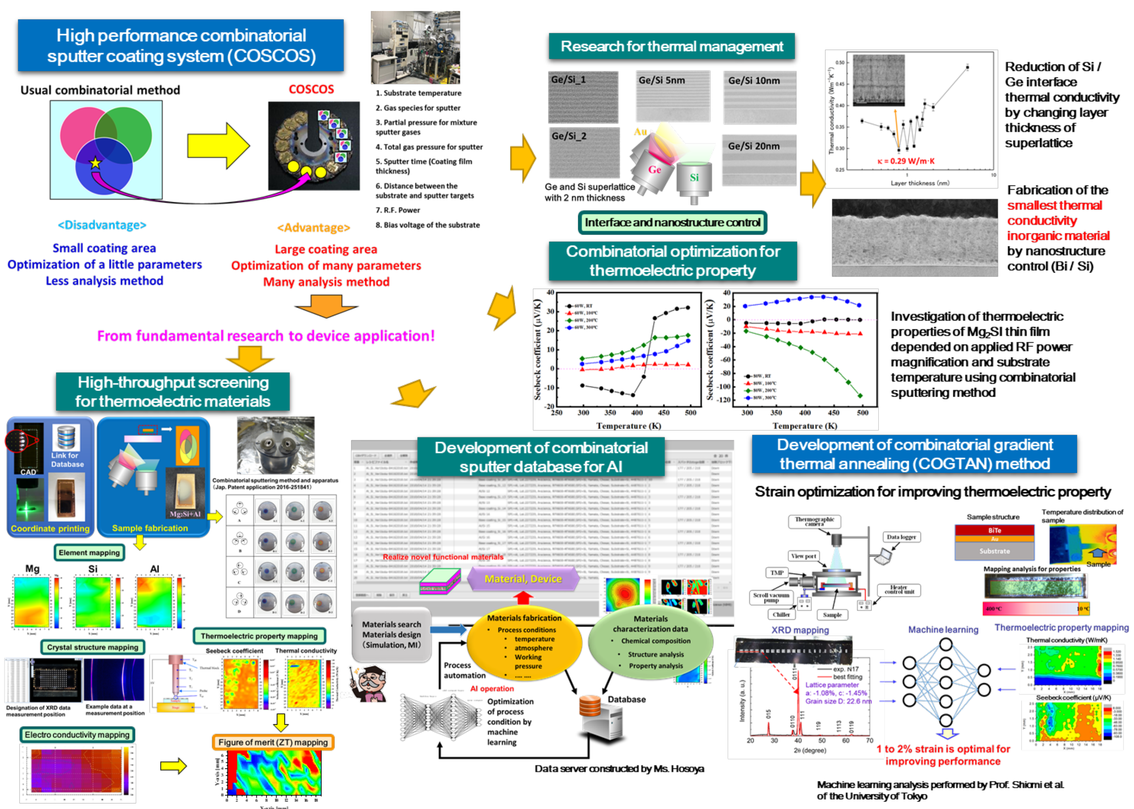


Considering the processability, new design of blends and block-copolymers of aromatic polyimides reached anisotropic in-plane thermal conductivity up to 1.0 W/mK without fillers.

New Substance exploration and Materials Design



Development of high-throughput material synthesis and evaluation technique



Thermophysical property database

A thermophysical property database was developed, and data of thermal conductivity, electrical conductivity, specific heat capacity, and thermal diffusivity were stored as a function of temperature (Figure 1). Analysis of specific heat capacity data visualized universality of Dulong-Petit law for single element solids and Neumann-Kopp rule for multi-element solids such as oxides and carbide nitrides and demonstrated importance of an experimental dataset for materials informatics (Figure 2).

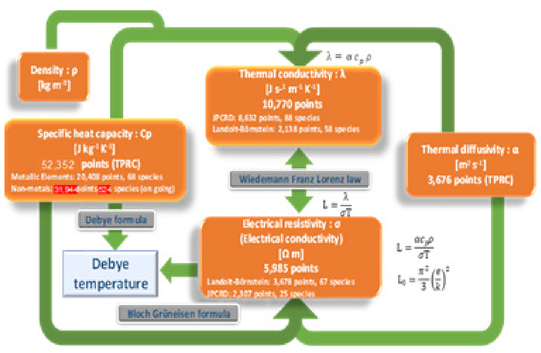


Fig.1 A thermophysical property database developed for materials informatics, in which thermal conductivity, electrical conductivity, specific heat capacity, and thermal diffusivity of solids are stored.

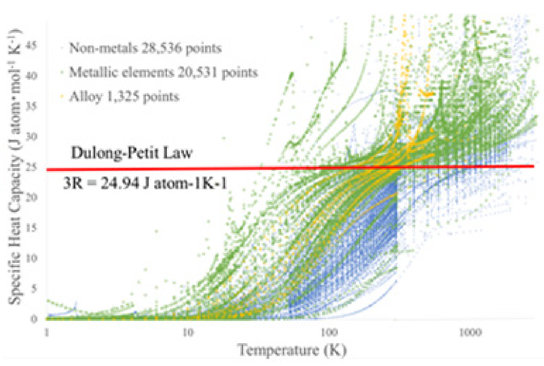


Fig.2 Per "atom・mole" unit: Temperature dependence of specific heat capacity of metals, alloys and non-metallic solids (Logarithmic scale for temperature)

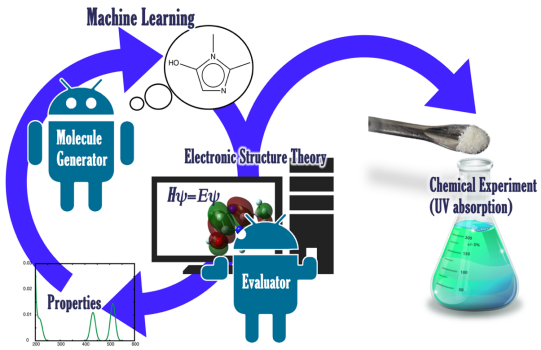
Overview of Data Science Group

Data Science Group

Koji Tsuda, Ryo Tamura tsuda.koji@nims.go.jp, tamura.ryo@nims.go.jp

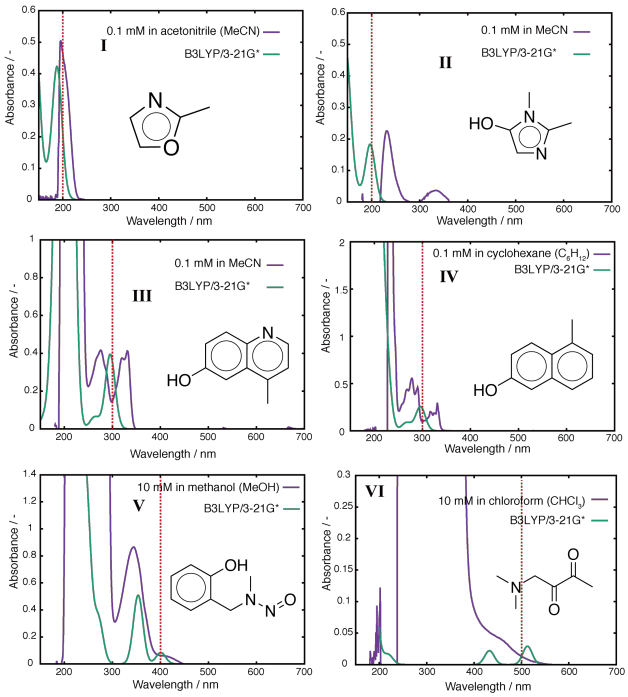
De novo molecule generation

Combining deep learning and quantum simulation for discovering new molecules



ChemTS+Gaussian
(https://github.com/tsudalab/ChemTS)

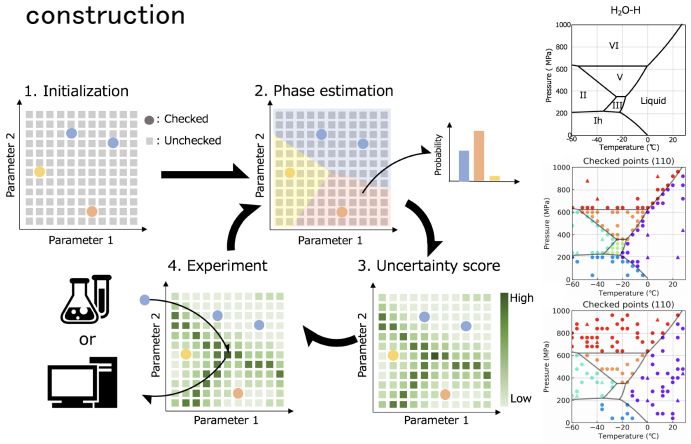
Target wavelength	200 nm	300 nm	400 nm	500 nm	600 nm
Generated	646	757	629	607	638
Simulator-Qualified	34	26	13	12	1
Synthesized	2	2	1	1	0
Functional	1	2	1	1	0



M. Sumita, R. Tamura, and K. Tsuda, et al.
ACS Cent. Sci. 4, 1126 (2018).

Phase diagram construction

New machine learning algorithm for phase diagram construction

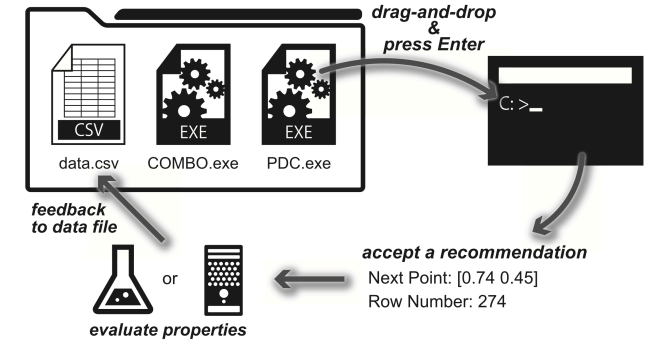


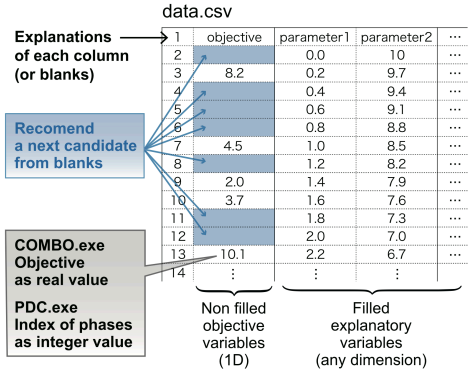
K. Terayama, R. Tamura, and K. Tsuda, et al.
Phys. Rev. Materials 3, 033802 (2019).

https://github.com/tsudalab/PDC

MI research by Windows computers

Executable files of COMBO and PDC on Windows without any installation





K. Terayama,
K. Tsuda,
and R. Tamura,
Jpn. J. Appl. Phys.
58, 098001 (2019).

https://tsudalab.org
/ja/projects/mitools

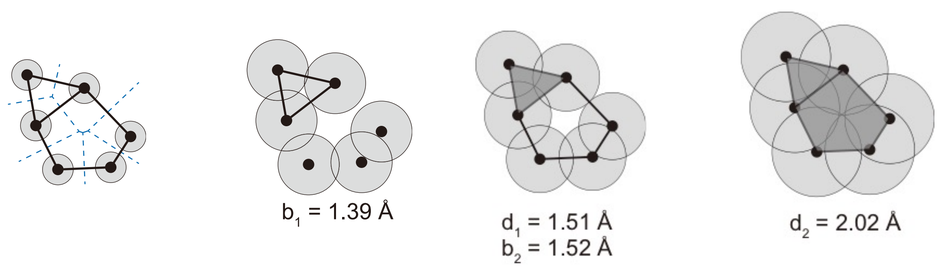
Overview of Topological Analysis Team

Topological Analysis Group
Kazuto Akagi, Shinji Kohara and Yasuaki Hiraoka akagi@wpi-aimr.tohoku.ac.jp

Outline of our Approach

- A new mathematical scheme handling the "shape of data".
- "Shape" : n-dimensional hole (n=0 connectivity, n=1 ring, n=2 cavity)
- Treating a set of discrete points as input data: atomic configurations, digital images, ...
- Translating the complex input data into two-dimensional histogram quantitatively.
- Detecting a hidden order and elucidating its origin (with machine-learning techniques, if necessary).
- Proposing new materials based on the obtained PSPP relation.

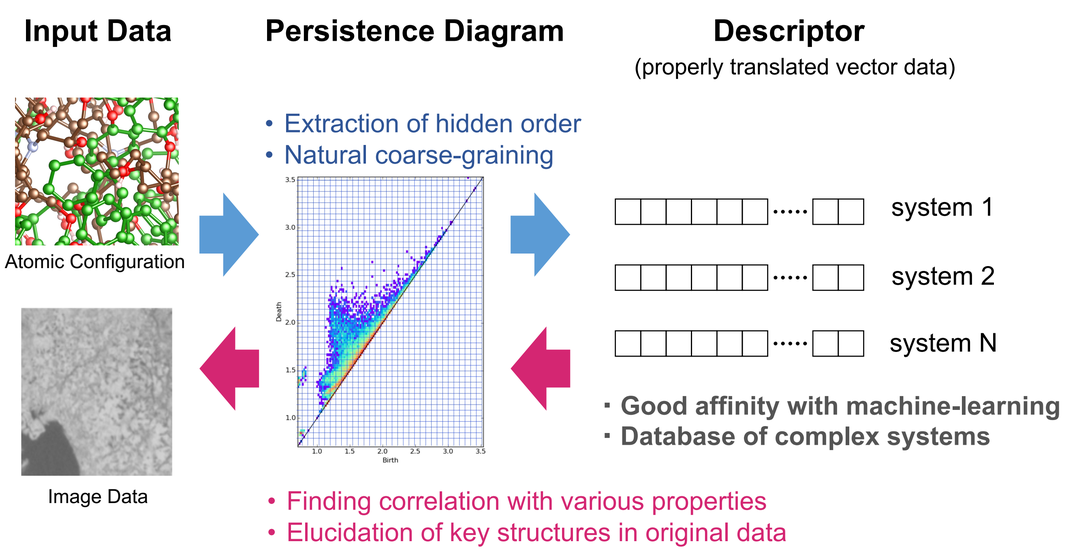
Persistence Diagram (PD)



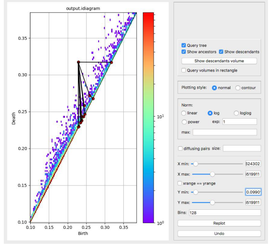
- One of the expressions of “Persistent Homology”
- Filtration based on "Alpha Shape"
- Born at r=b, dead at r=d
- Each (b,d) corresponds to each "Hole"
- Robust "Hole" : apart from diagonal line
- Noisy "Hole" : near by diagonal line

Hole: connectivity, ring, cavity

Forward and Inverse Analysis via PD



Development of “HomCloud” Package



- Input data: atomic configuration, image files
- Calculation of persistence diagram (forward)
- Mapping (b,d) onto input data (inverse)
- Written as python modules
- Open source under GPL v3
- Developed by I. Obayashi and Y. Hiraoka

Search now “HomCloud AIMR” !

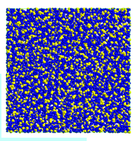
Members

Y. Hiraoka: Development and implemetation of mathematical framework
F. Ogushi: Analysis of grain boundary and dynamic phenomena
S. Kohara, Y. Onodera, S. Tahara, A. Masuno:
Synthesis and analysis of functional glass materials
M. Kotsugi: Analysis of magnetic domains toward reduction of “core loss”
K. Akagi: Analysis and quantification of microscopic observed images

Examples of Application

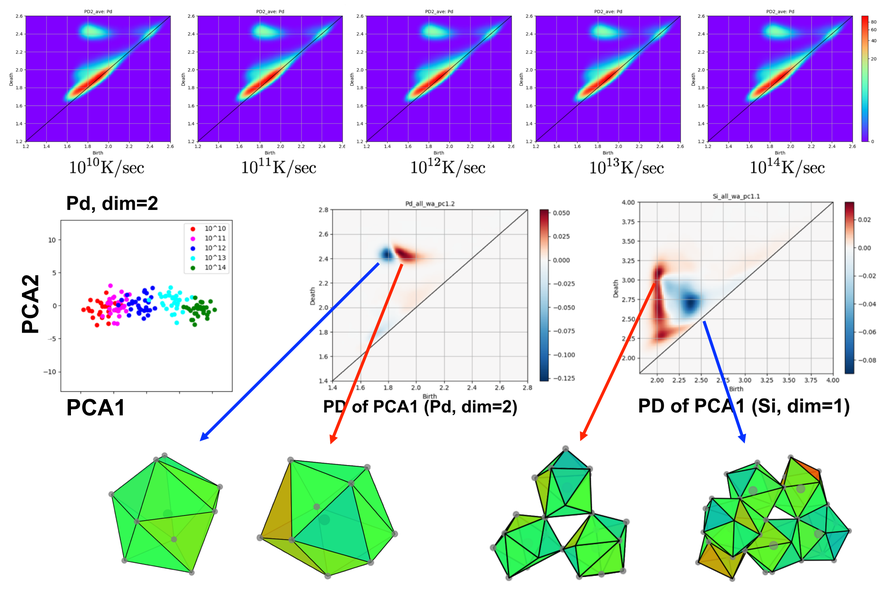
- Metallic Glass: cooling rate effect on glass structures

(Hiraoka *et al.*)



- Apply PDs to MDs for describing structures
- Combine ML and inverse analysis for explicitly identify those changes

Persistence diagrams for different cooling rates in MD simulations

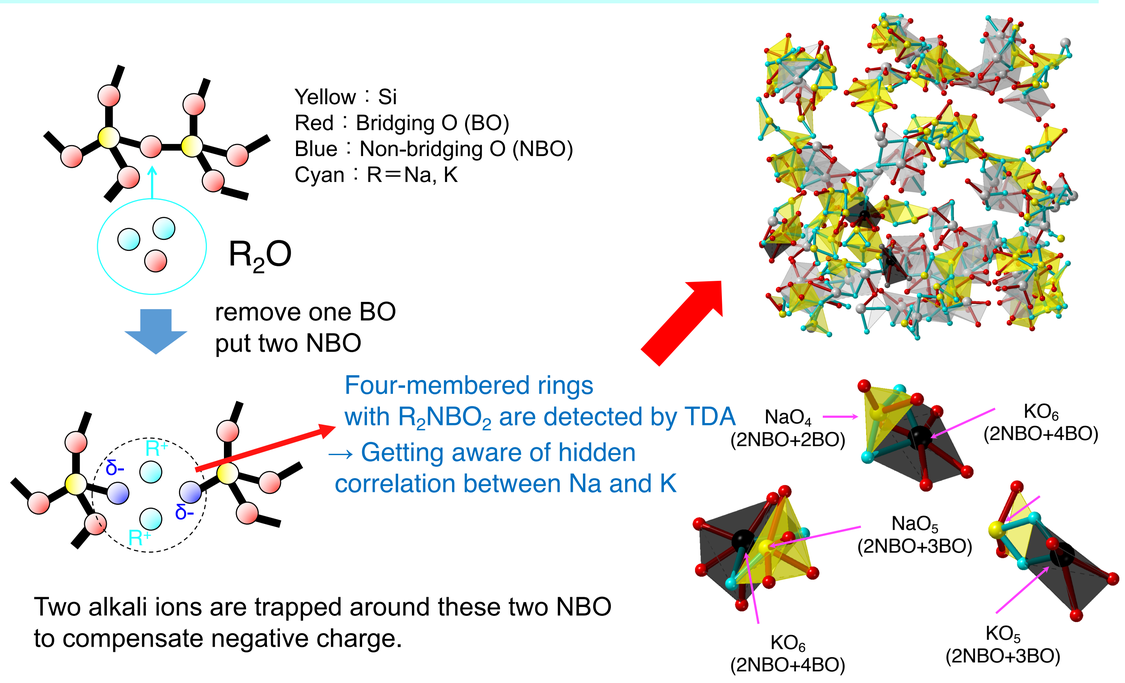


Inverse of PCA explicitly identifies geometry affected by cooling rates!

- Structural origin of mixed alkali effect in alkali silicate glass

(Kohara *et al.*)

- Experimental measurement of functional glass
- Topological data analysis (TDA) based on the MD simulation reproducing the experimental features

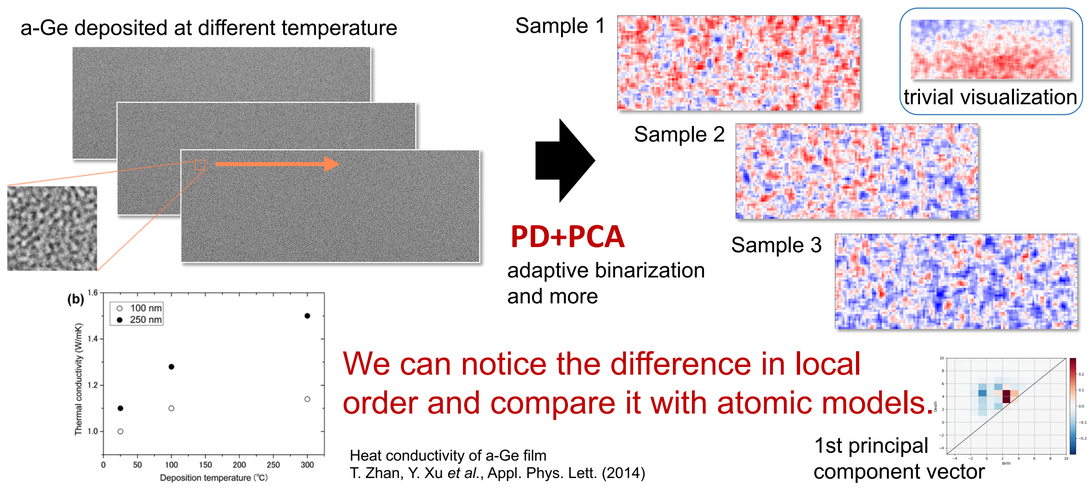


Information beyond “pair-correlation” plays an important role!

- Characterization of TEM images of amprphous materials

(Akagi and Xu)

- Non-trivial visualization by scanning PD and machine-learning
- Design of pre-process to avoid misleading characterization



Research Activity in Materials Exploration Group

Materials Exploration Group

Tamio Oguchi
Kohji Nakamura

oguchi@sanken.osaka-u.ac.jp
nakamura.kohji@mie-u.ac.jp

Background and Goals of Group

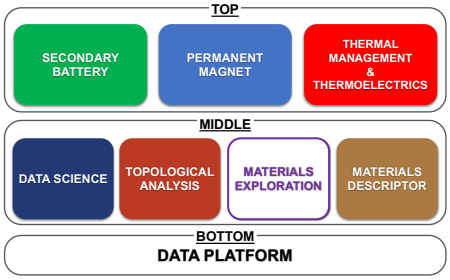
Goals of Materials Science

- To discover novel materials with desired property/function applicable to future devices
- To optimize the property/function of existing materials for realizing specified performance
- To disclose the underlying physics of materials property/function

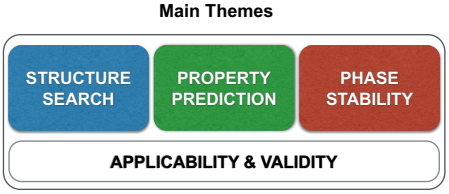
➡ Our target materials have become much more complex and demanding to meet the requirements for future applications.

➡ To accelerate R&D, much attention has been paid to Materials Informatics (MI).

Center for Materials research by Information Integration MaDIS-NIMS



NIMS-MaDIS-CMI² Materials Exploration Group



Selected Research Activity

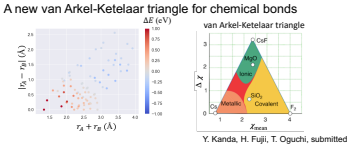
Sparse Modeling of Chemical Bonds
T Oguchi (Osaka University)

- Interpretable sparse model for quantifying the energy difference between rock salt (RS) and zinc blende (ZB) structures in octet binary compounds

$$E(RS) - E(ZB) \cong \frac{1}{(r_A + r_B)^3}\{-5.02|r_A - r_B| + 6.87\} - 0.18$$

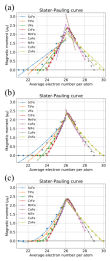
Atomic radius

- A new van Arkel-Ketelaar triangle for chemical bonds



Y. Kanda, H. Fujii, T. Oguchi, submitted

Development of linearly independent descriptor generation method
H. Fujii (NIMS)



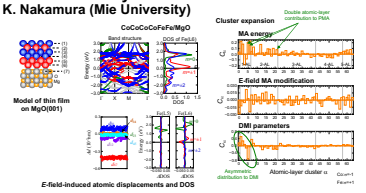
Interpretable modeling by LIDG method:
Descriptor generation + Linear independence + Sparse modeling

Ex. Magnetic moment of 3*d* binary alloys

$$M(A,B,x) \sim (1-x)M_P(A) + xM_P(B) + x(1-x)[-4.37 + 0.32Z(A) + 0.54M_1(B,A) + 0.42|M_1(A,B)|].$$

https://github.com/Hitoshi-FUJII/LIDG

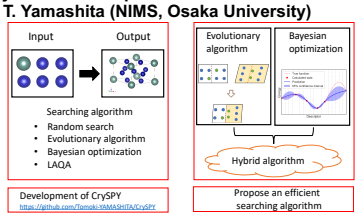
Layer-stacking dependence of magnetism in Fe-Co multilayer thin films
K. Nakamura (Mie University)



A double atomic-layer stacking of Fe on the MgO enhances a PMA but there is no role of the short-range sequence in the *E*-field-induced MA modification, whereas an asymmetry in the whole film along the stacking direction plays a role in determining the DMI.

Submitted (2019), MMM (2019).

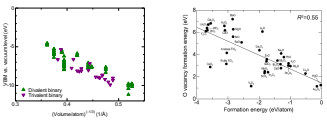
Development of crystal structure prediction method
T. Yamashita (NIMS, Osaka University)



Phys. Rev. Materials 2, 013803 (2018).
npj Computational Materials 4, 32 (2018).

Finding descriptors of surface properties
Y. Hinuma (Chiba University)

- **Correlations between surface and bulk properties**

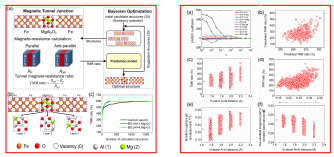


Correlation between ionization potential and volume per atom in binary oxides

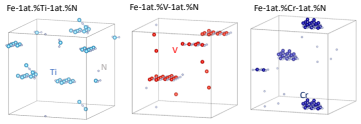
Correlation between surface O vacancy formation energy and bulk formation energy in binary oxides

Phys. Rev. Materials 2, 124603 (2018).
Phys. Rev. Materials 3, 84605 (2018).
J. Phys. Chem. C 122, 29435 (2018).

Machine learning analysis on tunnel magnetoresistance of Fe/MgAl$_2$O$_4$/Fe(001)
Y. Miura (NIMS)



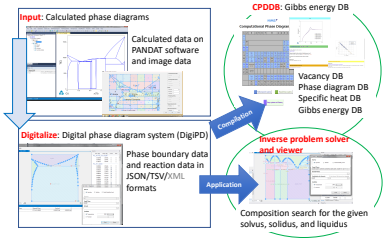
Atomic scale microstructure simulation using interatomic interactions obtained from DFT calculations
H. Ohtani, M. Enoki (Tohoku University)



☐ Multibody interactions to six-body cluster in the Fe-M-N (M=Ti, V, Cr) ternary systems were evaluated from the cluster expansion method, and the formation mechanism of the nanocluster was examined by introducing the interactions directly into the Monte Carlo simulation.
☐ According to the microstructure simulation based on the interaction energies, characteristic mono-layer clusters appear in the bcc Fe-Ti-N and Fe-V-N alloys, while spherical cluster forms in the Fe-Cr-N system.

M. Enoki, H. Ohtani, *Tetsu-to-Hagané*, 105 (2019) 334.

Digital phase diagram database for Open Science
T. Abe (NIMS)



A first-principles phase field method for quantitatively predicting multi-composition phase separation without thermodynamic empirical parameter
R. Sahara (NIMS)

Combining density functional theory, cluster expansion theory and potential renormalization theory, the free energy were derived as a function of compositions and construct a parameter-free PFM, which can predict microstructures in high-temperature regions of alloy phase diagrams.

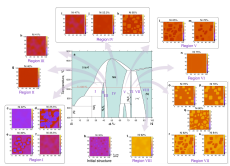
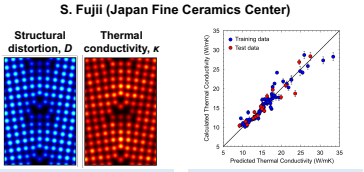


Figure: Microstructures of Ni-Al alloys at various alloy compositions at 1300K.

S. Bhattacharyya, R. Sahara, and K. Ohno, Nature Comm. 10 (2019) 3451.

Predicting Thermal Conductivity from Grain Boundary Atomic Structures
S. Fujii (Japan Fine Ceramics Center)



Significant correlation between *D* and *κ* near grain boundaries (GBs)

Predicting GB thermal conductivity using machine learning

S. Fujii, et al., Acta Mater. 171, 154-162 (2019).
S. Fujii, et al., in preparation.

Interface perpendicular magnetic anisotropy in various heterostructures
S. Mitani (NIMS)

New sputter-deposition process with oxidation and reduction reactions for large perpendicular magnetic anisotropy in Fe/MgO heterostructures

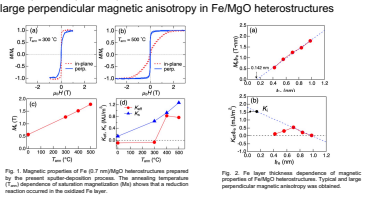


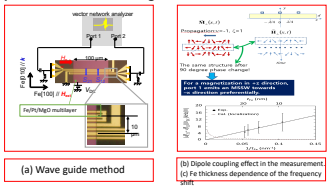
Fig. 1. Magnetic properties of Fe (0.7 nm)/MgO heterostructures prepared by the present sputter-deposition process. The annealing temperature (T_{ann}) dependence of saturation magnetization (Ms) shows that a reduction reaction occurred in the oxidized Fe layer.

Fig. 2. Fe layer thickness dependence of magnetic properties of Fe/MgO heterostructures. Typical and large perpendicular magnetic anisotropy was obtained.

Y. Iida et al., in preparation

Electric field effect in Magnetic metals
Y. Suzuki (Osaka University)

- **Dipole effect in the wave guide method**



(a) Wave guide method

(b) Dipole coupling effect in the measurement. (c) Fe thickness dependence of the frequency shift

Nawaoka et al., in preparation

XenonPy: a Python open-source project for Materials Informatics

Materials Descriptor Platform group

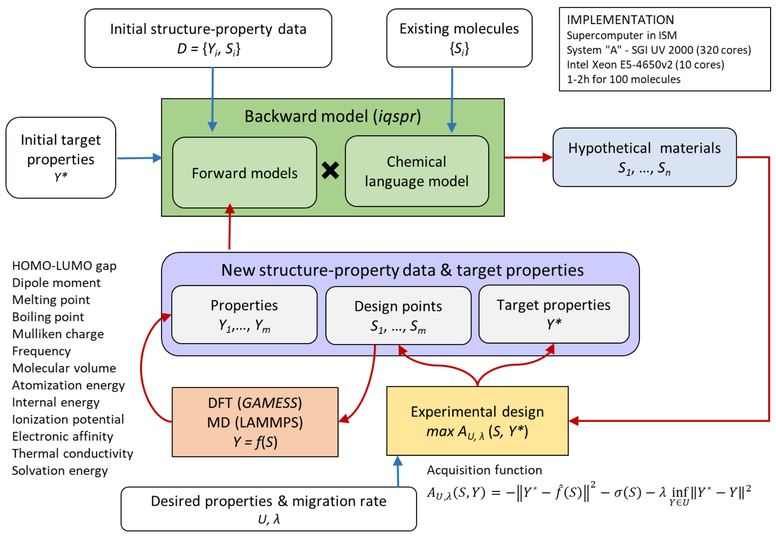
Ryo Yoshida, Chang Liu, Yukinori Koyama yoshidar@ism.ac.jp

XenonPy

https://github.com/yoshida-lab/XenonPy

- 組成・構造・分子記述子ライブラリ
- 約140,000個の訓練済みモデル (XenonPy.MDL)
- 転移学習モジュール
- 分子設計の機械学習アルゴリズム (iQSPR-X)

SPACIER Go Beyond Interpolative Prediction



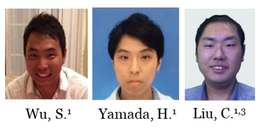
Pre-trained Model Library 'XenonPy.MDL'

Yamada & Liu et al. Predicting materials properties with little data using shotgun transfer learning. *ACS Cent Sci* (2019)

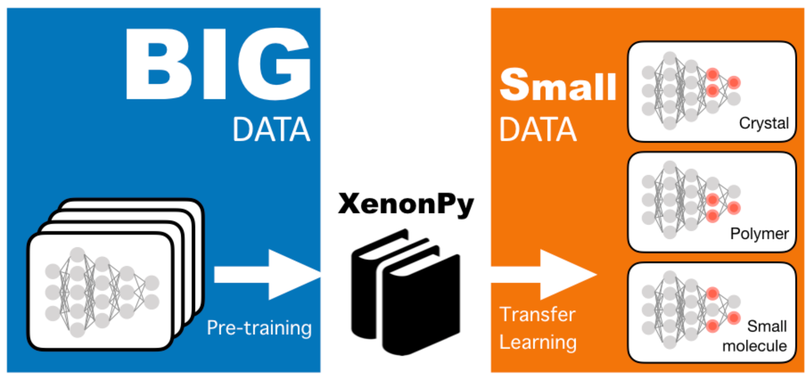
Database: ~140,000 pre-trained models on 45 tasks

ONLINE TUTORIAL

https://xenonpy.readthedocs.io/en/latest/tutorials/6-transfer_learning.html

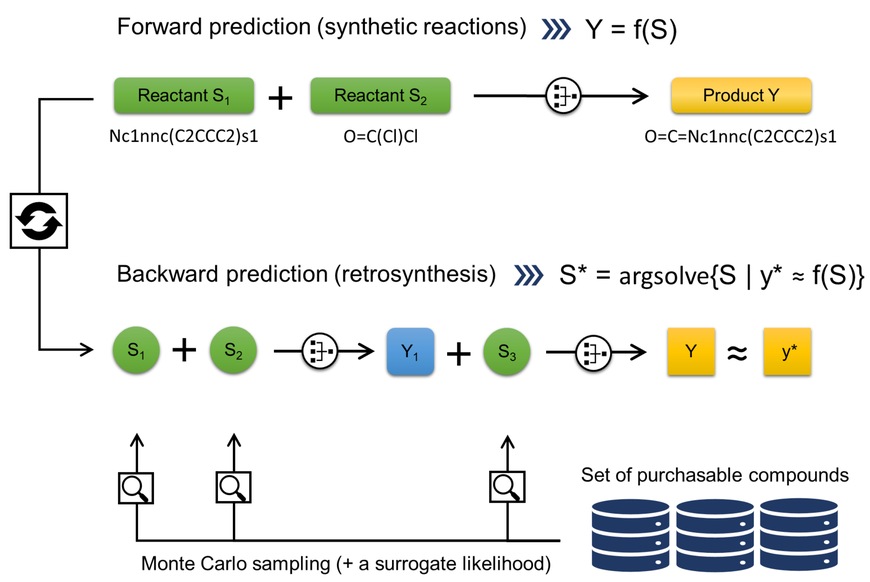


Wu, S.[1] Yamada, H.[1] Liu, C.[1,3]



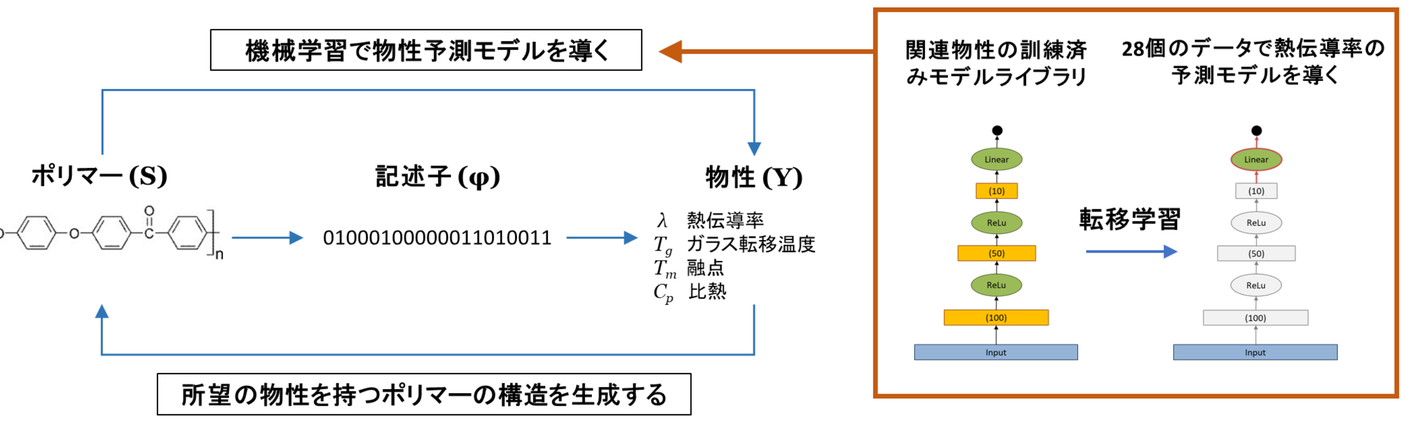
Bayesian Retrosynthesis

Guo et al. A Bayesian Algorithm for Retrosynthesis (in preparation)

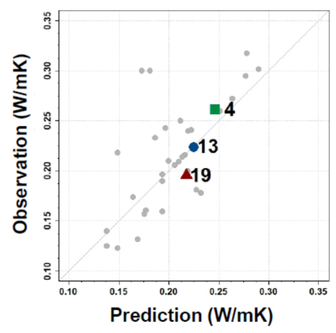


機械学習の「記憶」を活用し、高分子の熱伝導性の大幅な向上に成功
～少ないデータでも高精度な予測が可能に　高分子での材料インフォマティクス加速に期待～

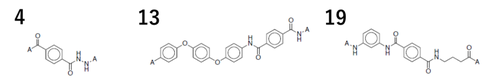
Wu et al. Machine-learning-assisted discovery of polymers with high thermal conductivity using a molecular design algorithm. *npj Comput Mater* 5:66 (2019)



転移学習による高分子熱伝導率の予測

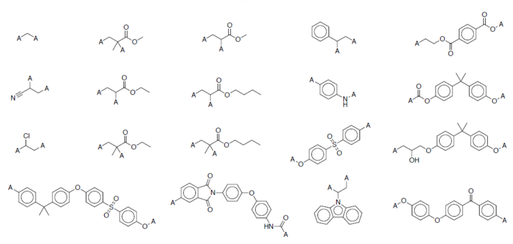


予測対象（メソゲン基を骨格とする新規合成高分子）



外挿領域に存在

モデルの訓練に使用した19個の高分子



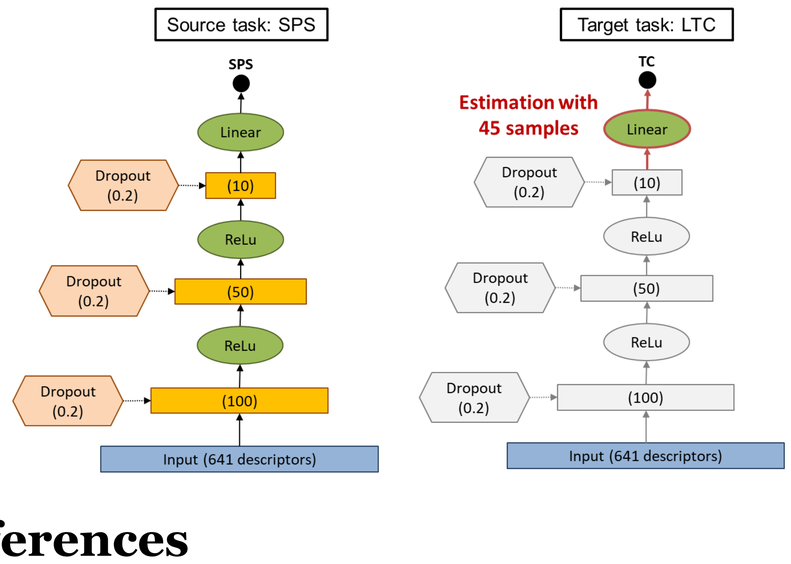
XenonPyを活用した超高熱伝導性無機化合物の探索
訓練済みモデルライブラリXenonPy.MDLと転移学習を活用し，超高熱伝導性無機化合物を発掘

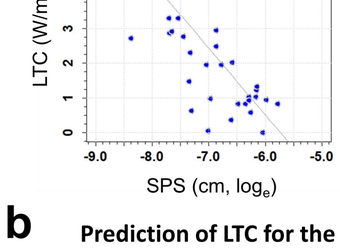
Ju et al. Exploring ultrahigh lattice thermal conductivity crystals via feature-based transfer learning. *ChemRxiv* (2019)

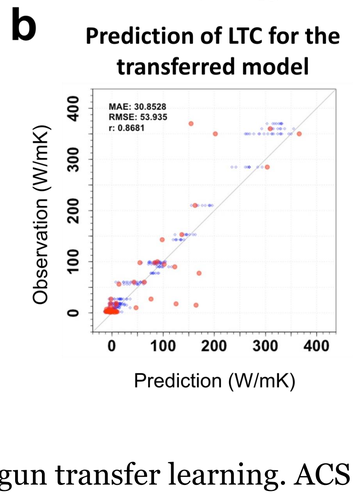
Yamada & Liu et al. Predicting materials properties with little data using shotgun transfer learning. ACS Cent Sci. 5(10):1717-1730 (2019)

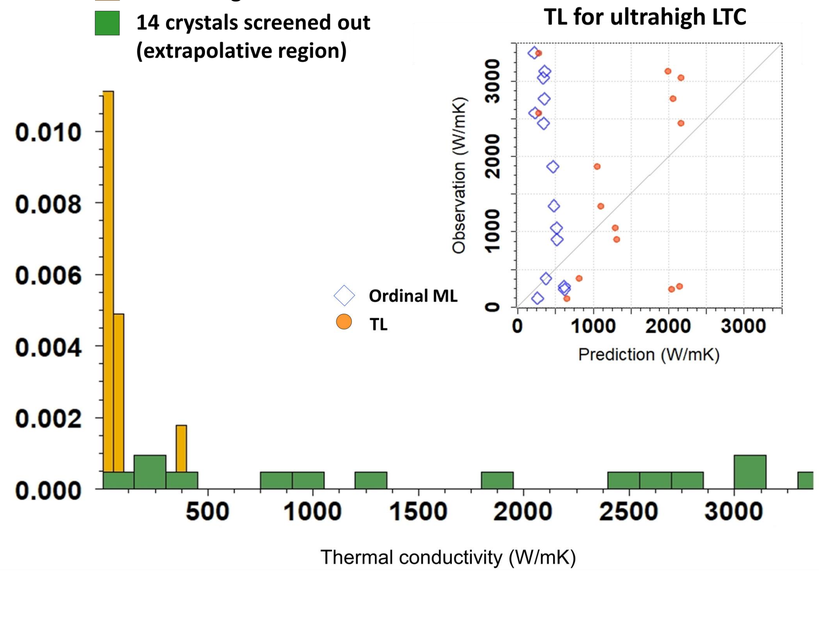
Number of samples on SPS and thermal conductivity

Property	Number of samples
Lattice Thermal conductivity (target)	45 materials
SPS (intermediate)	320 materials









References

1. Yamada & Liu et al. Predicting materials properties with little data using shotgun transfer learning. ACS Cent Sci. 5(10):1717-1730 (2019)
2. Wu et al. Machine-learning-assisted discovery of polymers with high thermal conductivity using a molecular design algorithm. npj Comput Mater 5:66 (2019)
3. Wu et al. iQSPR in XenonPy: a Bayesian inverse molecular design algorithm. Mol Inform (2019)
4. Ju et al. Exploring ultrahigh lattice thermal conductivity crystals via feature-based transfer learning. ChemRxiv (2019)
5. Ikebata et al. Bayesian molecular design with a chemical language model. J Compt Aided Mol Des. 31, 379-391 (2017)

MI²I Data Infrastructure

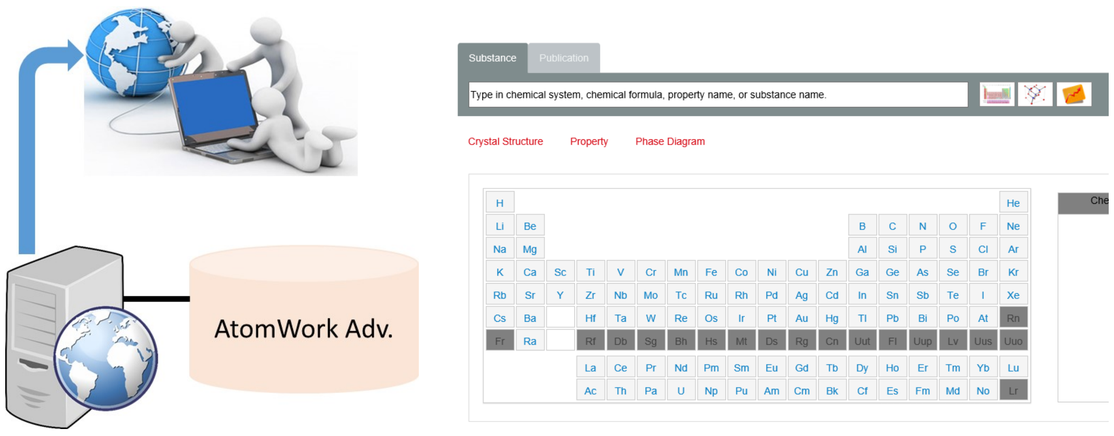
Data Platform Group

Yibin Xu, Junko Hosoya, Yuta Sakairi, Hiroyuki Yamasato

XU.Yibin@nims.go.jp

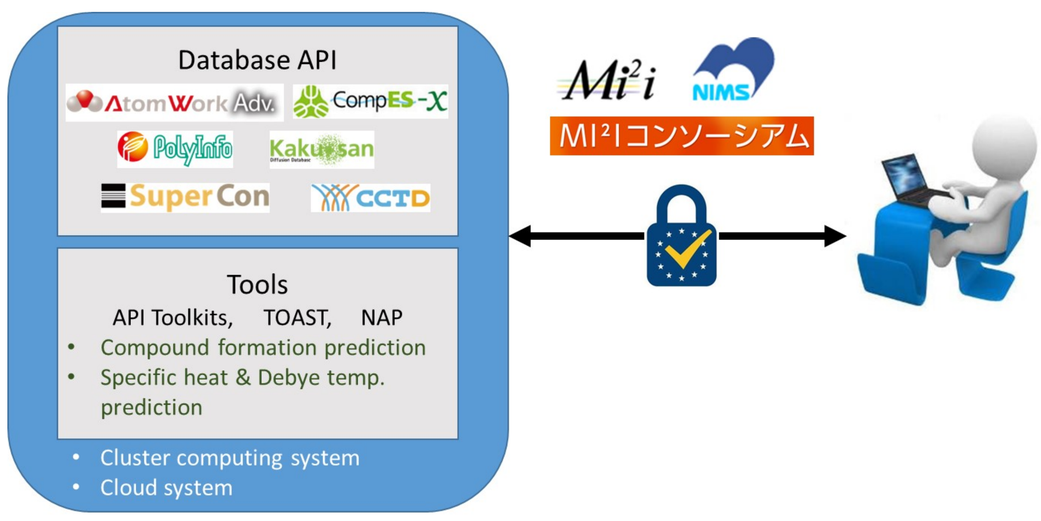
Inorganic Materials Database AtomWork-adv.

The world largest scale database for inorganic materials. At present, 318,837 crystals, 43,497 phase diagrams and 390,263 properties are available.

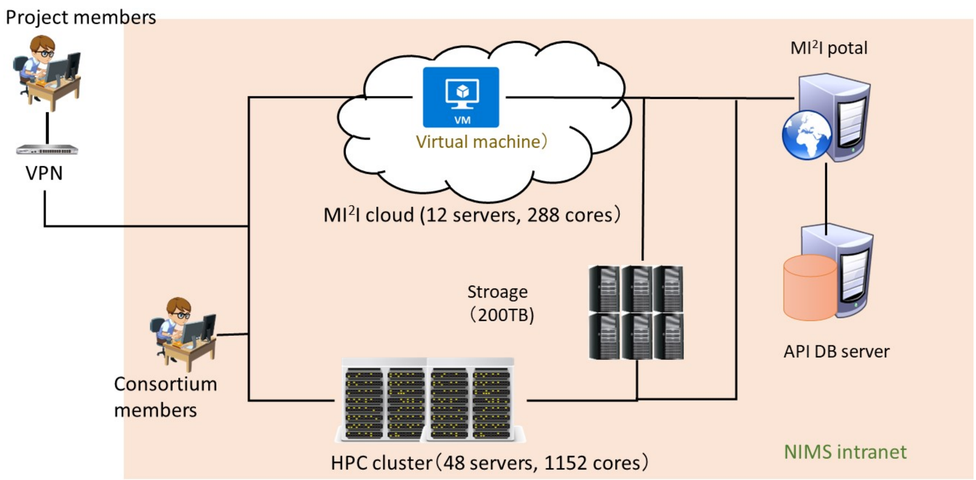


MI²I Data Platform

API service of MatNavi and AtomWork-Adv. data

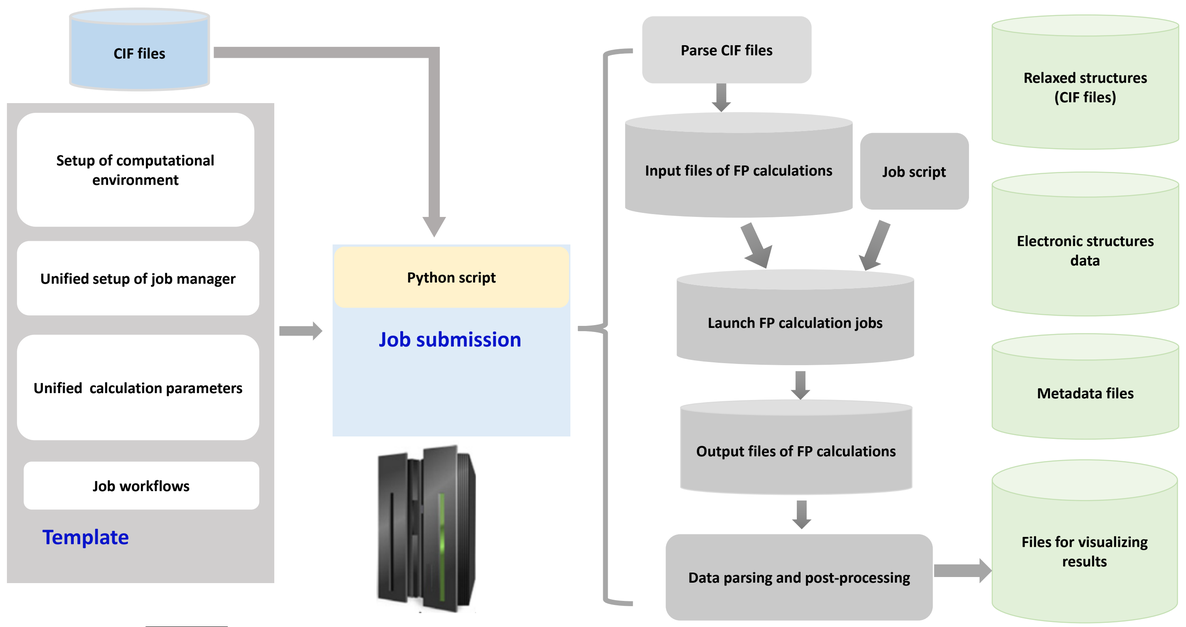


Computational platform of HPC cluster and cloud

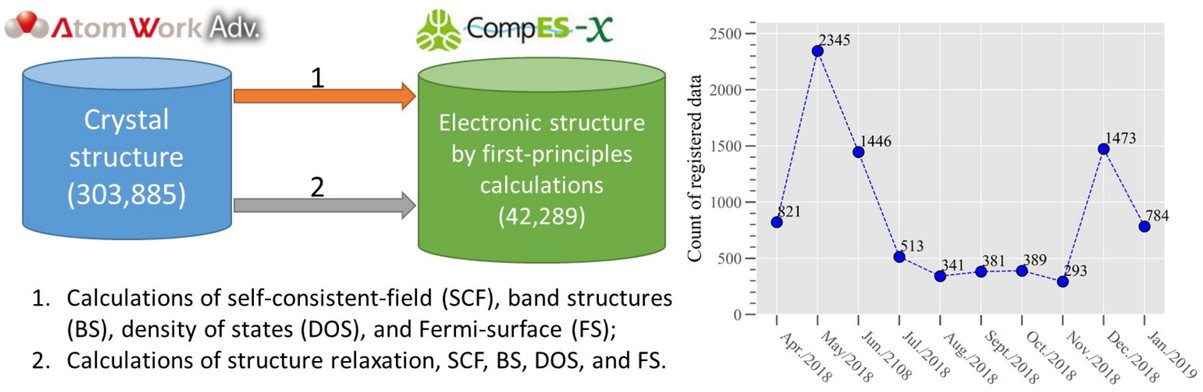


Data Applications

Framework of Template Oriented Atomic Simulation Toolkit (TOAST)



Electronic Structure Data Generation



1. Calculations of self-consistent-field (SCF), band structures (BS), density of states (DOS), and Fermi-surface (FS);
2. Calculations of structure relaxation, SCF, BS, DOS, and FS.

MI²I Portal Site

https://mi2i.nims.go.jp/

Structure of portal site :

Page for the public

- Outline of services
- Download of tool
- Usage guide

Page for specified users (Only accessible from NIMS LAN and MI²I-DPF)

- Details of services
- Download of usage manual
- Download of API manual & sample codes

Page for DPF users (User authentication needed)

- Changing password
- Display of API token
- Display of API call count

Specific Heat Prediction Tool. Predict specific heat and Debye temperature by Neumman-Kopp's law and machine learning based on experts evaluated data.