NATIONAL INSTITUTE FOR MATERIALS SCIENCE

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Center for Basic Research on Materials

Probing the cutting edge where informatics and measurement technologies meet

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State-of-the-art analytical and measurement technologies go beyond observing materials' nanostructures—they track atomic and electronic behavior in real time. Data-driven approaches streamline the search for materials with desirable properties. These tools are indispensable in modern materials research and development.

Coordinated use of measurement technologies and data-driven techniques is crucial in expediting materials research. Data science techniques extract vast amounts of useful information from measurement data, creating a virtuous cycle that significantly enhances the efficiency of materials development and leads to groundbreaking discoveries.

The NIMS Center for Basic Research on Materials (CBRM)—whose members consist of a team of measurement and data science experts—was launched to implement a coordinated approach to materials development. By combining cutting-edge techniques from both fields, the CBRM is building a versatile foundation to support diverse materials development efforts.

NIMS is strategically integrating these approaches to produce synergistic effects in its materials development.

Data : **Center for Basic Research on Materials**

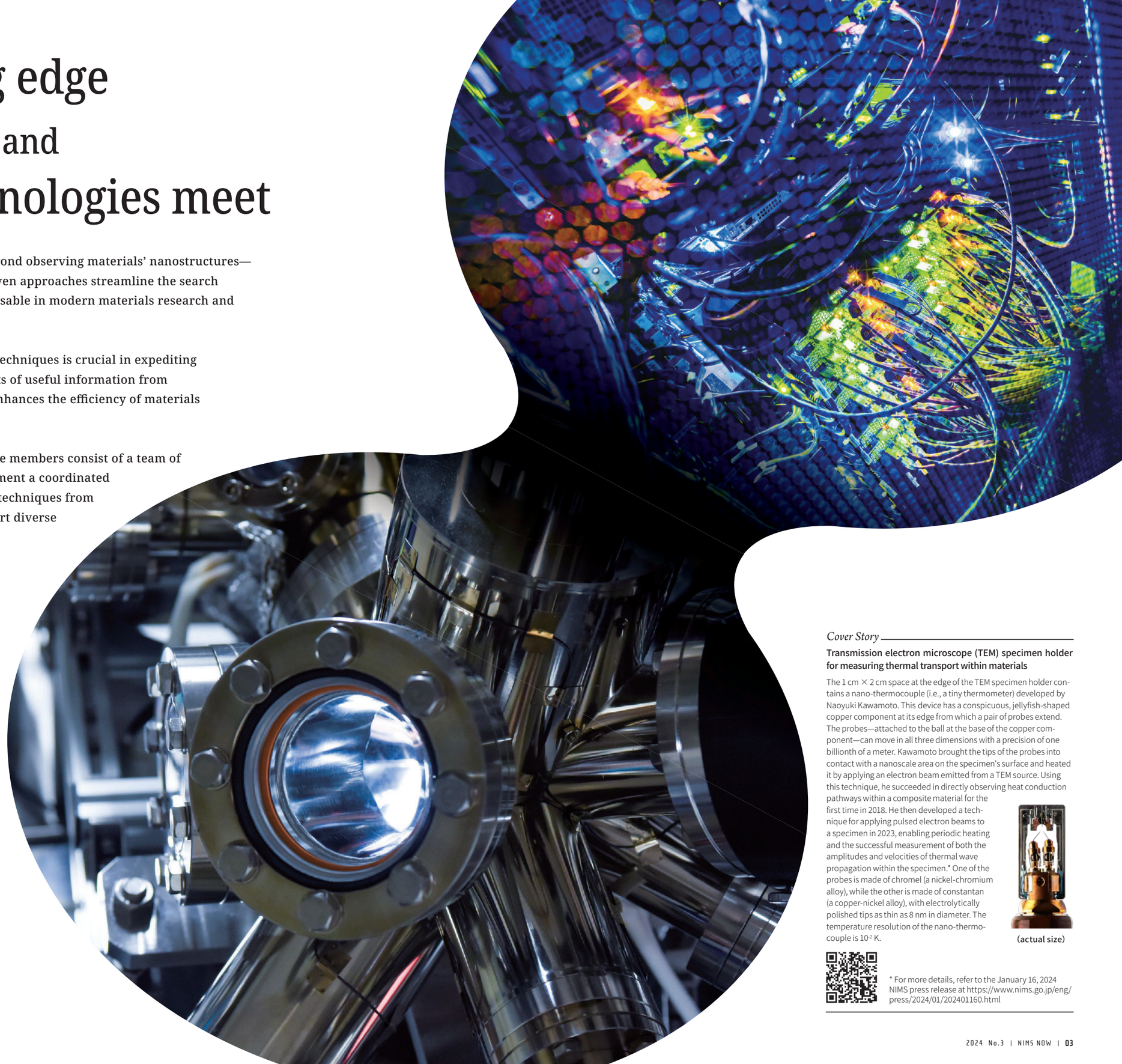
Director : Koji Kimoto  Number of permanent researchers: 47

Advanced Materials Characterization Field

- Electron Microscopy Group
- In-situ Electron Microscopy Technique Group
- Nanoprobe Group
- Solid-State NMR Group
- Photoemission Spectroscopy Group
- High Magnetic Field Physics Group
- Quantum Beam Diffraction Group
- Synchrotron Radiation Imaging Team

Data-driven Materials Research Field

- Data-driven Inorganic Materials Group
- Data-driven Materials Design Group
- Materials Modeling Group
- Computational Materials Science Group
- Data-driven Algorithm Team



Cover Story

Transmission electron microscope (TEM) specimen holder for measuring thermal transport within materials

The 1 cm × 2 cm space at the edge of the TEM specimen holder contains a nano-thermocouple (i.e., a tiny thermometer) developed by Naoyuki Kawamoto. This device has a conspicuous, jellyfish-shaped copper component at its edge from which a pair of probes extend. The probes—attached to the ball at the base of the copper component—can move in all three dimensions with a precision of one billionth of a meter. Kawamoto brought the tips of the probes into contact with a nanoscale area on the specimen's surface and heated it by applying an electron beam emitted from a TEM source. Using this technique, he succeeded in directly observing heat conduction pathways within a composite material for the first time in 2018. He then developed a technique for applying pulsed electron beams to a specimen in 2023, enabling periodic heating and the successful measurement of both the amplitudes and velocities of thermal wave propagation within the specimen.* One of the probes is made of chromel (a nickel-chromium alloy), while the other is made of constantan (a copper-nickel alloy), with electrolytically polished tips as thin as 8 nm in diameter. The temperature resolution of the nano-thermocouple is 10^{-2} K.

(actual size)

* For more details, refer to the January 16, 2024 NIMS press release at https://www.nims.go.jp/eng/press/2024/01/202401160.html

Key Projects

Overview of research projects carried out
at the Center for Basic Research on Materials (CBRM)

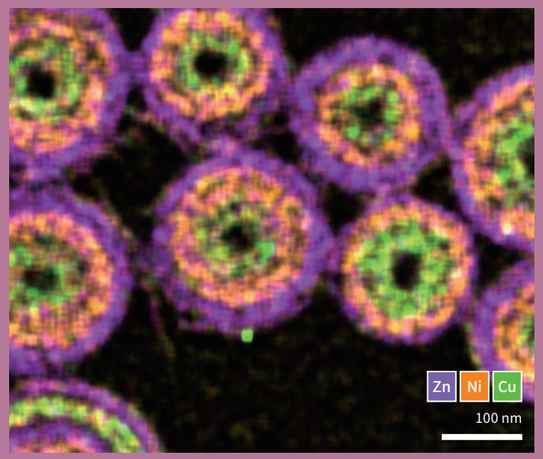
#1 Research on Cutting-Edge Analytical and Measurement Techniques Driving Innovation in Materials Science

Project leader
Kazutaka Mitsuishi

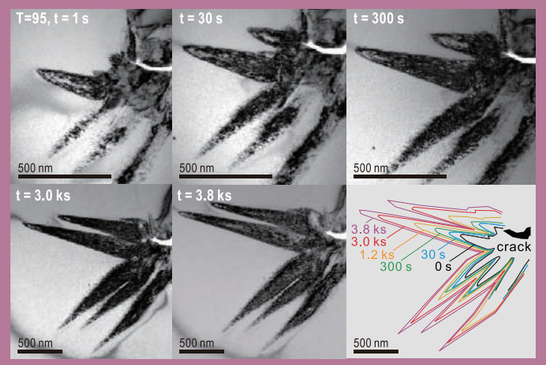
Atomic-Level Structural Observation and Analysis at Unprecedented Resolution

Developing advanced materials with desirable functions requires optimizing key parameters, such as surface and interfacial nanostructures, crystalline orientations, and electronic and spin states. To capture images of these material properties with unprecedented clarity and resolution, NIMS has significantly modified measurement instruments, analytical tools, and techniques. As a result, NIMS has developed world-class analytical technologies that offer exceptional spatial and temporal resolution, as well as advanced capabilities for energy, wavelength, and other measurements. Advances in magnetic and quantum materials have created a demand for new analytical techniques. This project was launched to meet current demand by actively integrating existing techniques, including high-throughput structural analysis enabled by the integration of multiscale analytical methods and data science.

See Research 1 on **p. 6**
See Research 2 on **p. 8**



Elemental mapping low-density metal atoms (～ 0.2 nm^{-3}) in supramolecule.
Photo provided by Jun Kikkawa



TEM images showing martensitic transformation dynamics taking place in a specimen while it is being stretched.
Photo provided by Kodai Niitsu

Operando Analysis and Measurement Informatics: Tools for Measuring the Real-World Behavior of Materials

Demand is growing for operando analysis—techniques that enable observation of materials under actual working conditions. These techniques have the potential to help address urgent social challenges, including the pursuit of carbon neutrality. This type of analysis adopts a "seeing is believing" approach, enhancing material properties by observing their performance in real-world settings.

The goal of this project is to develop new techniques to expand the range of materials and experimental conditions suitable for operando analysis beyond those already achievable in gaseous atmospheres, liquids, and when subjected to heat or electric voltage. For example, we aim to develop a soft X-ray microscope with spatial resolution higher than 100 nanometers and temporal resolution higher than 100 picoseconds, as well as a multimodal operando microspectroscope at the "NanoTerasu" , high-brilliance synchrotron radiation facility.

Simultaneously, our team of measurement and data science experts will collaborate to improve the accuracy of measurement informatics approaches, extracting useful information from the vast amounts of data generated by our instruments.

#2 Laying the Groundwork for Data-Driven Materials Research

Project leader
Keitaro Sodeyama

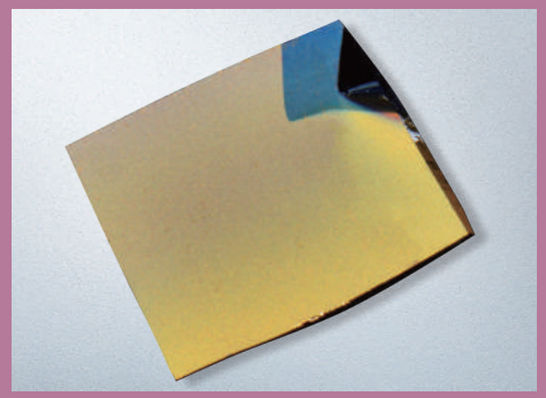
Building a Foundation to Improve Data Quality and Materials Development Efficiency

The effectiveness of data-driven materials development depends on data quality and quantity. The CBRM has been upgrading data collection techniques—mainly those used to extract relevant information from scientific papers and collect data generated by autonomous, automated experimentation and large-scale calculations—to improve both the quality and quantity of data in NIMS' materials databases. This project aims to intensify these efforts by developing techniques to extract physical property features from data generated by instruments. The CBRM will also develop schemes to automate all the steps involved in large-scale first-principles calculations, from executing the computations to data aggregation.

Additionally, the CBRM is adding value to existing data by compiling a dictionary to enable AI to understand the data's semantics, by coordinating materials databases with other databases across different disciplines, and through other means. Through these efforts, NIMS is producing ever more reliable data with the goal of vigorously promoting data-driven materials development.

See Research 4 on **p. 12**

The TurboRVB software can be used to accurately calculate the force acting on an atom and lattice vibrations in solids (see p. 14).



This thermal insulation film (a composite of amorphous silicon and bismuth) with one of the world's highest insulation performances was designed using a machine learning model developed by NIMS.

Offering User-Friendly Data-Driven Techniques

Data-driven approaches to identifying materials with desirable properties are becoming more widely adopted by materials developers. To further broaden the range of materials these methods can be applied to, thereby promoting the discovery of practical materials, it is necessary to develop methodologies tailored to specific materials and desired functions. This project aims to develop tools to address the practical issues faced by materials scientists. These include a machine learning model to optimize inorganic materials design, a system capable of controlling autonomous, automated experimentation, and techniques to create a model capable of predicting the four key elements of materials engineering: material fabrication processes, structures, properties, and performance.

The goal of these efforts is to integrate all the types of materials data stored across Japan, making them available for data science-based analysis. NIMS serves as Japan's core organization responsible for managing and consolidating materials data and facilitating its use.
The CBRM will work diligently to develop the fundamental technologies needed to fulfill these responsibilities.

See Research 3 on **p. 10**

RESEARCH 1

Cutting-Edge Single Molecular Chemistry: Synthesizing Molecules with a Local Probe

Shigeki Kawai

Group Leader,
Nanoprobe Group

Kawai is currently developing a low-temperature magnetic field AFM system for spin detection of carbon nanostructures. The photo shows him adjusting the microscope head.

A scanning probe microscope (SPM) is indispensable for observing and evaluating the structures of materials at the atomic and molecular level. SPMs have also become important tools for single molecular chemistry—manipulation of individual atoms and molecules to synthesize novel materials. We asked Shigeki Kawai, an active researcher in this field, about his recent research achievements.

What is single molecular chemistry?

A scanning probe microscope (SPM) uses an ultrasharp probe to scan the surface of various materials and generate images that enable observation of its surface topography and electronic properties at atomic-level resolution. The two main types of SPMs are atomic force microscopes (AFMs), which detect atomic forces between the probe and the sample, and scanning tunneling microscopes (STMs), which detect the tunneling current between the probe and the sample.

Kawai noted that SPMs are no longer mere observational tools.

"A research paper published about 15 years ago by an overseas research institute found that the spatial resolution of SPMs can be greatly improved by the probe apex terminated with a single carbon monoxide molecule," Kawai said. "This improvement made it possible to observe molecules in greater detail—even their backbones were resolved for the first time. In addition, the performance of SPMs was improved, significantly enhancing probe controllability with a digital SPM controller. This has enabled the probes to be positioned at any site on the sample surface with picometer precision using three-dimensional coordinates, allowing the breaking of specific bonds in a molecule by flowing the tunneling current to a target site and the synthesis of new molecules. As a result, the popularity of single molecule chemistry grew rapidly."

Organic molecules are usually synthesized through chemical reactions in solutions. However, this method cannot manipulate individual molecular structures. By contrast, scanning probe microscopy enables the manipulation of individual atoms and molecules—the bottom-up synthesis of organic molecules. This approach may be used to create innovative devices that take advantage of atomic and electronic interactions.

Kawai is one of the scientists leading efforts to advance the field of single molecule chemistry using SPMs. He began to develop a new instrument—a combination of an AFM and STM (hereinafter referred to as an "AFM/STM")—while working at the University of Basel in Switzerland. He has since made a series of modifications to the AFM/STM and has been using it to carry out bottom-up synthesis of nanostructures.

The potential of carbon nanostructures for use in next-generation electronics

Kawai is currently focused on synthesizing carbon nanostructures, such as graphene, which has long been known for both its excellent electrical conductivity and great mechanical strength. Among the different forms of graphene known, graphene nanoribbons (GNRs), where graphene forms ribbon-like structures with given widths of a few nanometers, are particularly noteworthy. GNRs are semiconductors with a bandgap, and their properties change depending on their width and edge shape, making them highly promising as next-generation electronic materials.

Kawai says, "There is a growing demand to endow GNRs with various functions by incorporating atoms or molecules other than carbon to create new nanodevices. However, this requires the use of a probe to detach existing atoms and then attach new ones. The portions where atoms are removed become highly reactive radicals and can easily bond with atoms or molecules on the substrate surface, making control difficult. In response to this challenge, a joint research team led by NIMS and Osaka University has been working on the synthesis of three-dimensional GNRs (3D GNRs)."

First-ever success in adding a preselected molecule to a GNR using an SPM

Osaka University Professor Takashi Kubo succeeded in synthesizing a propeller-like molecule with three "blades" by substituting terminal hydrogen atoms at the tips of these blades with bromine atoms. This achievement inspired the launch of the joint research mentioned above. When this propeller-like molecule was placed on the substrate surface, two of its blades came into contact with the surface while the other blade projected upward from the surface (the first molecule from the left in figure 1). Kawai found this molecular orientation in relation to the substrate ideal for the purpose of substituting the terminal bromine atoms at the tip of the up-pointing blade with an external molecule.

"We decided to use the propeller-like molecule as a precursor to synthesize 3D GNRs," Kawai said. "We first deposited the precursor molecules on the surface of a silver substrate placed under the ultrahigh vacuum and heated them, causing them to polymerize into three-dimensional organometallic compounds. We further heated these compounds until they transformed into 3D GNRs with bromine atom-terminated up-pointing blades." The next step was to attach preselected molecules to the 3D GNRs.

Kawai chose a fullerene—C60, another carbon nanostructure—as the molecule to attach to the 3D GNRs. He first removed two terminal bromine atoms from a 3D GNR blade tip by flowing tunneling current to them at a given bias voltage, generating a radical site. He then attached a fullerene to the apex of the probe by picking up it from the substrate, moved the probe tip to a position 100 picometers above the radical site, and subsequently set the probe to the radical site to connect them. This resulted in the world's first successful attachment of an external molecule to a specific 3D GNR site using an AFM/STM probe. Kawai achieved this in 2020.

Creating radical molecules that could be used as quantum materials

Furthermore, in 2023, Kawai succeeded in more precisely controlling the structure of molecules that became radical states after the removal of two bromine atoms.

Using the same procedure he previously used to connect a fullerene to a 3D GNR in 2020, Kawai synthesized a 3D GNR using the AFM/STM and removed two bromine atoms from its up-pointing blade. Kawai then thoroughly analyzed the 3D GNR's molecular orbitals to understand the electronic and structural properties within it.

As a result, Kawai found that the debrominated blade composed of carbon atoms transforms into dehydroazulene—a molecule composed of five- and seven-membered carbon rings fused together (figure 2). Kawai then positioned the probe above the seven-membered ring and applied a greater bias voltage to it, thereby converting the seven-membered ring into a five-membered ring and the adjacent five-membered ring into a seven-membered ring. In other words, this conversion produced a chiral isomer of azulene, which was highly reproducible as encoding the ASCII characters of NIMS in the molecular array (Figure 3).

Kawai also found that an intermediate, diradical molecule composed of two six-membered rings fused together momentarily comes into existence during the transformation of a debrominated blade into azulene. This diradical molecule—with two unpaired electrons—is even more reactive and ephemeral than single-radical molecules. "Exchange interactions—quantum mechanical effects that play a crucial role in determining magnetic properties and electron spin behavior—occur between the unpaired electrons in diradicals," Kawai said. "I was able to extend the lifetime of the diradical by moving the probe as far away from the 3D GNR as possible without causing the unwilled structural change to the azulene, and then measured the exchange interaction in the diradical. This result demonstrated the possibility of manipulating quantum materials using an AFM/STM probe."

Kawai's success in extending the longevity of ephemeral diradical molecules and manipulating them was attributed to the improved cryogenic and vacuum environment of the AFM/STM and his exceptional SPM manipulation skills. There is great anticipation for the possibilities that could be unlocked by the new science using probes.

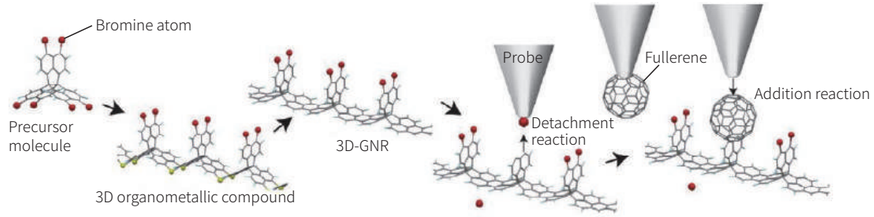


Figure 1. A research group led by Osaka University Professor Takashi Kubo developed a propeller-like molecule with three "blade tips" terminating in six bromine atoms (red). A joint research team led by NIMS and Osaka University then used this molecule as a precursor to synthesize 3D GNRs. The team first deposited the precursor molecules on the silver surface placed under ultrahigh vacuum conditions and heated them, causing the terminal bromine atoms in contact with the substrate to detach from the molecules, while allowing the terminal bromine atoms away from the substrate to remain attached to the molecules. This resulted in the formation of 3D GNRs. Kawai succeeded in substituting a 3D GNR's bromine atoms with preselected molecules (e.g., fullerenes) using the AFM/STM probe.

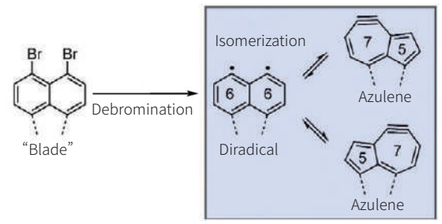
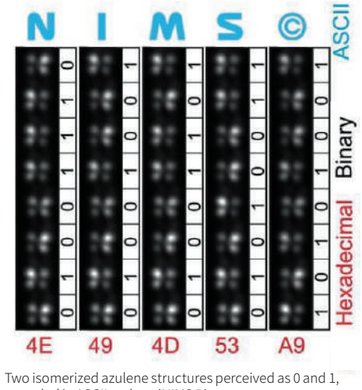


Figure 2. Detaching bromine atoms from the up-pointing "blade" of a 3D GNR causes the blade to transform into dehydroazulene—a molecule composed of five- and seven-membered carbon rings fused together. Isomerization of azulene can be achieved by applying the bias voltage to it using the probe. Kawai found that an intermediate, diradical molecule composed of two six-membered rings fused together momentarily comes into existence during this azulene isomerization. He succeeded in extending the life of this diradical molecule for a sub-second.



Two isomerized azulene structures perceived as 0 and 1, recorded in ASCII code as 'NIMS©'.

A spin-resolved photoemission spectroscopy system with imaging capability being developed by Yaji's group (see figure 2 for details).

RESEARCH 2

Quantum Beams: A Tool for Revealing the Fundamental Properties of Materials

Shinji Kohara
Group Leader,
Quantum Beam
Diffraction Group

Koichiro Yaji
Group Leader,
Photoemission
Spectroscopy Group

The number of large-scale facilities capable of generating "quantum beams" —a term popularized in Japan for particle and photon beams— is increasing globally, offering a broader range of beams. These advances allow scientists to explore the atomic and electronic states of materials in greater depth. Shinji Kohara uses quantum beams to unravel the unsolved secrets of glasses, aiming to develop higher-performance variants. Concurrently, Koichiro Yaji is working on photoemission spectroscopic measurement techniques and lab-scale systems that rival large-scale facilities. Kohara and Yaji shared their latest research activities.

Increasing and upgrading quantum beam facilities

Quantum beams are highly focused, directional beams of particles or light with quantum properties. By irradiating sample materials of interest with these beams and measuring diffraction and scattering patterns, the atomic and molecular arrangements can be analyzed in detail. Many countries have built advanced large-scale quantum beam facilities, providing significantly higher real-space resolution than lab-scale equipment. These facilities offer powerful tools for materials science R&D.

Japan also has several quantum beam facilities capable of generating different beam types. By choosing the most appropriate beams, multifaceted analysis can be performed for specific materials and research objectives.

For example, hard X-ray beams at SPring-8 and Photon Factory can sensitively measure heavier elements in interiors of sample materials, making them suitable for determining the samples' atomic arrangements and crystalline structures. In contrast, the neutron beams at the J-PARC facility are more effective for measuring lighter elements like oxygen and hydrogen.

The NanoTerasu facility, operational since April 2024, generates soft X-ray beams with higher brightness than other facilities. These beams enable world highest-resolution analysis of electronic and chemical bonding states on sample surfaces. The wider range of quantum beams provides scientists new methods for observing and measuring material properties.

Unraveling mysteries surrounding glass

Kohara—an expert in the structural analysis of glasses and liquids—has played a major role in developing high-performance glasses by analyzing data collected at SPring-8 and J-PARC. He explained the current status of glass research and the contributions being made to it by quantum beam-based measurements.

"Demands for glass have been diversifying in recent years," Kohara said. "For example, smartphone cover glass needs to be lightweight and crack-resistant, while glass used in optical fiber cable must minimize transmission loss. Developing high-performance glass requires in-depth analysis of its compositions and structures to optimize production conditions and processes. However, due to its amorphous nature and complex composition, glass analysis is challenging, and its formation mechanisms have long been a mystery. Only in the last decade or so has this mystery begun to be unfolded using quantum beam-based analyses."

Kohara recently researched crystallized glass with a private company. Although glass is inherently amorphous, heat treatment can induce a microcrystalline structure within its matrix. Crystallized glass typically exhibits improved mechanical properties, thermal stability, and chemical resistance.

Kohara's group conducted multiscale, element-specific analyses using various techniques, including anomalous X-ray scattering (AXS), X-ray absorption fine structure (XAFS) spectroscopy and small angle X-ray scattering (SAXS), mostly at SPring-8. As a result, the group revealed the mechanisms of improvements in the mechanical strength and heat resistance of the crystallized glass by adding zirconium oxide (ZrO_2) to raw glass materials (figure 1).

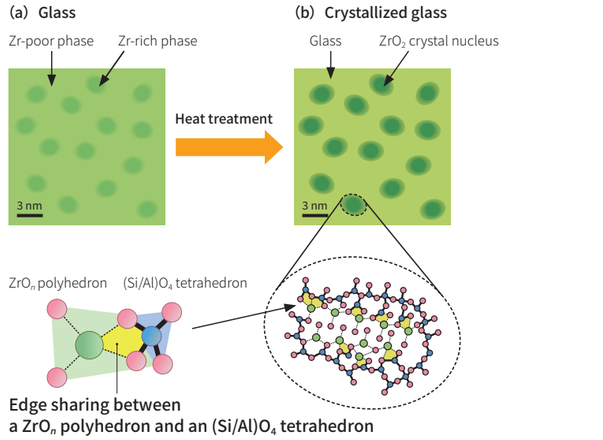


Figure 1. Nanoscale structure of crystallized glass depicted based on the analysis conducted at SPring-8.
(Left) ZrO_2-doped lithium aluminosilicate glass. (Right) Initial nucleation stage after heat treatment. The Zr concentration distribution is shown by green tones. Heat treatment increases Zr aggregation, causing separation into Zr-poor (glass) and Zr-rich (ZrO_2 crystal nucleus) phases.
Kohara's group analyzed crystal structure changes during the initial nucleation stage. They found that a ZrO_2 crystal nucleus has a periodic structure, composed of ZrO_n polyhedra and $(Si/Al)O_4$ tetrahedra bonded through edge sharing (yellow area in the illustration at lower left). This structure improves mechanical strength and heat resistance while maintaining transparency.

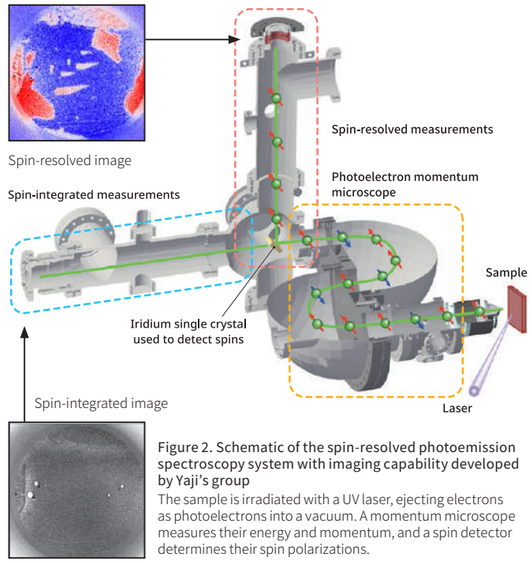


Figure 2. Schematic of the spin-resolved photoemission spectroscopy system with imaging capability developed by Yaji's group
The sample is irradiated with a UV laser, ejecting electrons as photoelectrons into a vacuum. A momentum microscope measures their energy and momentum, and a spin detector determines their spin polarizations.

"The experiments we conducted at SPring-8 were crucial to our discoveries," Kohara said. "However, an experimental approach alone won't give us a competitive edge in glass development. A data science approach is also vital. We are building a database to compile our extensive quantum beam diffraction data in coordination with data science experts and other scientists at the Center for Basic Research on Materials. We aim to create one of the largest glass-related databases in the world."

Nanoscale imaging of spin textures

Yaji, whose main focus is analytical technique development, has been advancing electron observation and measurement methods.

"I've primarily been working on UV lasers," Yaji said. "UV beams have lower energy than X-ray beams, making them suitable for measuring the electronic states involved in electrical conduction within materials. By improving measurement accuracy, I hope to contribute to next-generation spintronic and quantum device development. Our group has developed a spin-resolved photoemission spectroscopy system with imaging capability, small enough to fit in a laboratory space, reducing reliance on large-scale facilities (see the photo on the previous page)."

The electron's energy, momentum, and spin influence spintronic and quantum device performance. Measuring these typically requires several instruments, but Yaji's system can measure all simultaneously (figure 2).

"We spent about three years developing the system by adjusting and modifying a German-made photoemission analyzer," Yaji said. "Our spin-resolved photoemission spectroscopy system currently has spatial resolution and measurement efficiency far superior to those of any other similar systems available in Japan."

Yaji's system offers spatial resolution approximately 100 times greater than that of other existing systems, allowing it to measure areas smaller than one square micrometer. This significantly reduces the minimum measurable sample size.

"Our spin-resolved photoemission spectroscopy system has imaging capability and can make significantly larger numbers of simultaneous measurements," said Yaji. "Conventional systems determine spin at a single position, but our system's data collection efficiency is several million times greater, measuring spin at multiple positions. Additionally, it images spin textures within a material, providing a more intuitive understanding of the relationship between the spin and the material's electrical and magnetic properties (figure 3)."

In addition to these projects, Yaji has launched a new project: measuring spin states using a soft X-ray at NanoTerasu. He is attempting to measure spin states within a device under its actual operating conditions.

"I'm eager to make everything occurring in materials observable," Yaji said. "I want to master the full range of photoemission spectroscopic techniques so that I can recommend the most suitable ones to materials researchers for their specific objectives."

The growing range of quantum beam-based analytical techniques is opening new horizons in materials R&D.

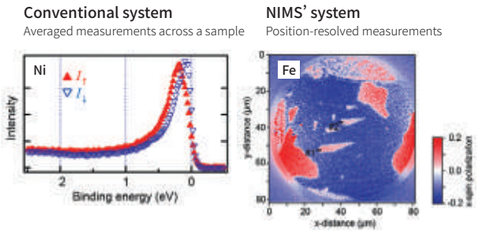


Figure 3. Comparison of photoemission spectroscopy data generated by conventional systems and the system developed by NIMS
NIMS' system can simultaneously perform photoemission spectroscopy at one million positions on a sample's surface—impossible with conventional systems. The image at the right shows the spin texture across an 80 μm × 80 μm surface of an iron polycrystal, with spin-up and spin-down states in red and blue, respectively. This spin texture map provides information on how the spin changes direction across magnetic domains.

RESEARCH 3

Aligning AI and Robotic System for Autonomous Automated Materials Explorations

Ryo Tamura
Team Leader,
Data-driven Algorithm Team

The electrochemical automated experiment robot "NAREE,"*2 controlled by NIMS-OS. Inside the glovebox at the top, the robotic arm tirelessly moves back and forth, advancing the exploration of electrolytes.

A robotic experimentation system can be used to automatically synthesize and evaluate materials. Artificial intelligence (AI) can then analyze the evaluation results and propose next areas for investigation. Ryo Tamura developed the NIMS-OS middleware to seamlessly integrate AI and robotic experimentation systems, contributing to the advancement of data-driven materials development.

Using AI to make materials research 72 times faster

Data-driven techniques can significantly streamline and expedite efforts to identify new materials with desirable characteristics. Tamura is one of the researchers who have been developing AI tools for data analysis since the early days of the integration of data science into materials science. He has also been helping other materials researchers at NIMS and elsewhere by providing them with AI tools, substantially speeding up their searches for new materials.

In 2023, Ken Sakaushi (NIMS Research Center for Energy and Environmental Materials) used AI tools developed by Tamura in his search for electrocatalytic materials capable of efficiently accelerating water electrolysis (see NIMS NOW Vol.22, No.1, p.15). Sakaushi succeeded in identifying a new, less expensive material as active as existing, platinum-based electrocatalysts in just a month. For reference, it has been estimated that the same comprehensive search process would have taken almost six years to carry out manually. This serves as a good example of how effective and powerful data-driven techniques can be when used in materials searches.

"AI-assisted materials searches require a variety of algorithms to achieve different objectives and identify different target materials," said Tamura. "I've collaborated with a number of materials researchers specialized in different types of materials—including alloys, magnetic materials, solar cells and electrolytic materials—to develop AI tools tailored to their specific needs. Currently, more than 10 different algorithms are posted on our Data-driven Algorithm Team's website,*1 making them freely available for public use.

What is NIMS-OS?

Although the AI tools developed by Tamura have already made materials searches significantly more efficient, he felt that there was still room for improvement.

Improving the prediction accuracy of AI models requires training them with large amounts of actual experimental and measurement data. In recent years, an increasing number of researchers have adopted automated robotic experimentation systems. These systems are perfectly suited for AI models as they generate large amounts of consistent data for training purposes.

An AI's ability to identify promising materials also enables a robotic experimentation system to conduct more efficient searches. Coordinating an AI with a robotic experimentation system can therefore produce significant synergistic effects. This "smart" materials search approach proved very effective in a joint research project conducted by Tamura and Shoichi Matsuda of the NIMS Research Center for Energy and Environmental Materials. Tamura's AI tools were used to analyze data generated by NAREE,*2 an automated robotic system.

Despite the great efficiency demonstrated by the AI-assisted NAREE system, it still required manual retrieval of experimental results from the robotic system and the entry of new data into the AI program after each round of experiments. Tamura felt that this human intervention created a bottleneck in the efficiency of the materials search process.

"Historically, AI and automated robotic experimentation technologies have been developed independently, making it impossible to seamlessly coordinate them," Tamura said. "To address this issue, I began to develop middleware to link them with the goals of eliminating human error and enabling experiments to be repeated continuously and without interruption around the clock. I thought that this would greatly expedite searches for promising materials while significantly reducing the amount of manual labor involved."

After about a year and a half of development, the NIMS-OS (Orchestration System) middleware was rolled out in July 2023. NIMS-OS recognizes the AI and automated robotic experimentation systems as separate modules, enabling it to be compatible with different combinations of AI and robotic systems. To use NIMS-OS, users must first install it on the PC used to control the automated robotic experimentation system. They then specify various parameters, including the type of AI tool*3 to use, the number of experimental cycles to run and the locations of input and output files. They then execute the programs to initiate their autonomous, automated experiments (figure).

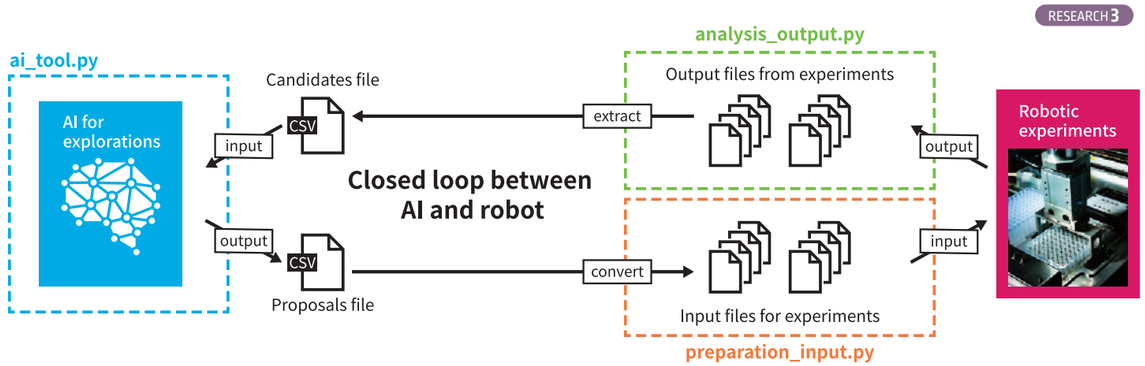


Figure. NIMS-OS middleware enables seamless coordination between the AI component and the automated robotic experimentation system.
NIMS-OS mainly consists of three types of programs. The first selects candidate materials for the next round of experiments based on a file of candidate materials, using AI to determine which candidates to test (blue box). The second program converts the data to a format readable by the automated robotic experimentation system and initiates the experiment (orange box). The third program analyzes the results upon receiving a completion signal from the robotic system and updates the candidate materials file accordingly (green box). By repeating this cycle, NIMS-OS enables autonomous and automatic material searches using robotic systems.

"I purposely designed NIMS-OS to be versatile. It is equipped with several AI tools that can be used in combination for various tasks," Tamura said. "Users can also add their own AI tools. It would be very difficult for researchers to create materials search algorithms from scratch without a great deal of experience in data-driven materials development. I wanted NIMS-OS to be user-friendly no matter the user's level of expertise and experience. These tools are posted on our website*1 as open-source software."

Successfully identifying high-performance electrolytes for lithium batteries

To verify the performance of NIMS-OS, Tamura installed it on NAREE—the automated robotic electrochemical experimentation system mentioned above—enabling the system to carry out fully automated searches for new, effective rechargeable battery electrolytes.

More precisely, this project aimed to identify high-performance electrolyte additives suitable for use with lithium-metal electrodes by utilizing an autonomous, automated system capable of performing all necessary steps—from dispensing and mixing compound quantities to conducting electrochemical measurements. 16 additive compounds were evaluated in groups of five, mixed into a base electrolyte in various combinations. In each round of the experiment, 32 different electrolyte formulations were evaluated simultaneously. After each experiment, Bayesian optimization was performed using the PHYSBO AI tool to select the next 32 formulations to be evaluated. As a result, a total of 384 different electrolyte formulations were evaluated in 11 experimental cycles from among 4,368 possible combinations.

"This proof-of-concept experiment demonstrated that NIMS-OS can enable a robotic system to perform autonomous, automated experiments without human intervention, except for one manual refilling of a solution," Tamura said. "I expect that autonomous, automated experimentation systems will become increasingly prevalent in the near future. I hope that NIMS-OS will help the many researchers who plan to use such systems. I'll add new AI tools to NIMS-OS if demand arises."

Contributing to the enhancement of materials databases

The data generated through autonomous, automated experiments is a valuable resource for other data-driven materials development efforts. As a leading materials science organization in Japan, NIMS has been developing DICE,*4 one of the world's largest materials data platforms, to fulfill its role as the core of data-driven materials development in the nation.

"NIMS-OS includes a feature that allows experimental data to be optionally transmitted to DICE through a data registration system called Research Data Express (RDE).*5 The transmitted data is then registered through RDE and integrated with other data within DICE," Tamura explained. "In fact, an increasing amount of data generated by NAREE has been registered by RDE and incorporated into the DICE data platform, making it a more reliable data source available for use by researchers searching for promising new materials. I hope to contribute not only to the individual needs of materials researchers but also to Japan's broader efforts to advance data-driven materials development."

With the aim of ensuring that all materials researchers can benefit from data-driven approaches, Tamura is committed to evolving his methods while considering the specific needs of materials researchers.

*1 Data-driven Algorithm Team's website: https://www.nims.go.jp/group/da/algorithm/

*2 NAREE is an automated robotic experimentation system mainly designed for searching promising electrolytes for lithium-air batteries. It can automatically and simultaneously prepare a large number of cell samples—each containing an anode, cathode, and separator—dispense electrolyte samples with different ratios of ingredients, inject differently formulated electrolytes into individual cells, and evaluate their performance.

*3 Three types of AI tools are available: PHYSBO, a Bayesian optimization tool; BLOX, a boundless objective-free search tool for identifying extreme mechanical properties in materials; and PDC, an active learning tool for constructing phase diagrams and state diagrams.

*4 DICE is an abbreviation for "Digital Innovative Collaborative Ecosystem." A pioneering system that seamlessly handles everything from the collection to the utilization of materials data.

*5 Simply by registering data online, it undergoes processes such as automatic formatting standardization and metadata extraction, and is stored within DICE as AI-readable data.

RESEARCH 4

Compiling a Dictionary to Enable AI to Analyze Polymer Data

Masashi Ishii
Chief Researcher,
Materials Modeling Group

Ishii, who is visually impaired, energetically engages in bridging the gap between the visually impaired and science, including hosting lectures on science behind braille.

PoLyInfo is a publicly available NIMS polymer database. Masashi Ishii has been working to make this database machine-readable, thereby enabling AI to understand the semantics of the data. He is also attempting to integrate PoLyInfo with biological databases, significantly more abundant data sources, to make it possible to address global-scale issues.

Creating a polymer dictionary with 25 million entries

NIMS' PoLyInfo database is a large collection of scientific literature-based data—a valuable resource in designing polymeric materials. The data was generated from about 170,000 polymer samples. In addition, more than 500,000 pieces of information related to the polymer samples (e.g., their properties, formation methods, conditions under which they were measured, monomers used to synthesize them and polymerization procedures) and their linkages are made available. The PoLyInfo database is a very rare, rich source of information on polymers—a group of materials that are difficult to categorize systematically due to their structural complexity and diversity.

Despite being a robust and comprehensive database, PoLyInfo is still challenging for AI to utilize effectively in discovering new polymers.

"AI recognizes PoLyInfo data merely as a series of meaningless characters and is unable to identify even basic terminology, such as what a polymer or a carbon is—fundamental knowledge for materials scientists," Ishii said.

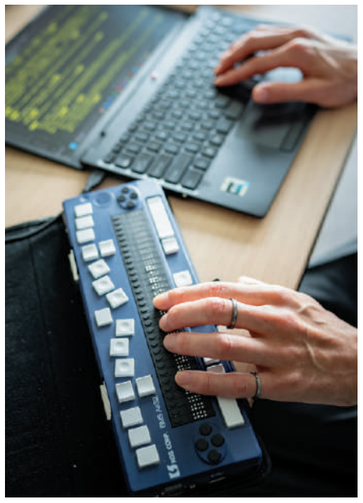
"It is therefore necessary to patiently teach AI all of the basics of polymers (e.g., that polymers are a type of material) so that it can correctly understand the semantics of the PoLyInfo data."

To achieve this, Ishii created a polymer dictionary, a compilation of "triples" consisting of three components: a subject, a predicate and an object. The triples describe a range of polymer-related information, from basic characteristics to advanced data derived from PoLyInfo.

"Ontologies are commonly used to describe concepts and relationships between entities using international standards, and have a hierarchical structure," Ishii said. "I compiled triples compatible with an ontology. I then used these triples to create a second set of triples by taking the object terms from the first set and employing them as the subject terms in the second set (figure 1). Data structured in this manner makes the database machine readable and compatible with AI analysis. For example, a series of triples can teach AI the following concepts: Tokyo is the capital of Japan, Ibaraki is located north of Tokyo and lotus roots are Ibaraki's main agricultural product. This machine-readable data structuring using triples is called the Semantic Web, and ontology is a vital tool in achieving it."

Ishii ended up composing 25 million triples using PoLyInfo data. Given that James Joyce's long novel 'Ulysses', which consists of approximately 10,000 sentences, we can comprehend just how colossal a number that is.

Although it may sound like a simple mechanical process, compiling triples to make PoLyInfo machine-readable was in fact very challenging. This is because there is considerable variation in the scientific literature from which PoLyInfo data is collected due to individual differences in expression and writing style. This variation is a major reason for PoLyInfo's poor machine readability. Ishii therefore revised the language used in the original literature containing the data to align it with international standards using specialized tools.



In both his research and daily life, Ishii makes full use of braille displays. When connected to a computer, the display raises braille dots according to the text information on the screen. This allows him to read and write data and save meeting notes in braille, among other things.

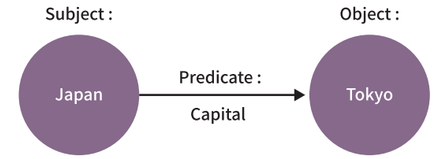


Figure 1. Example of information expressed in a machine-readable format (i.e., a triple)
An example of information expressed using a so-called triple, which consists of three components: a subject, a predicate, and an object. Among these components, the predicate is particularly important as it specifies the relationship between the subject and the object. Terms representing predicates are therefore aligned with international standards to enable the integration of PoLyInfo with other databases in different fields.

Solving social issues through integration with biological databases

Although PoLyInfo is an invaluable source of polymer data, its use has been limited to materials science. However, Ishii says that ontology has now opened the way to integrating and systematizing knowledge of PoLyInfo and other fields.

In addition to the 25 million-entry polymer dictionary Ishii compiled, many other data dictionaries that conform to international standards exist in different fields. Among these, biological databases are by far the most well-developed, both qualitatively and quantitatively.

"Biological databases contain substantially more data than PoLyInfo. For example, there are hundreds of millions of genome data points alone," Ishii said. "Moreover, efforts have already been made to link biological and non-biological databases, forming a huge information network. If PoLyInfo can be linked to this network, this integrated dataset could be used to address global-scale issues."

Marine pollution by microplastics is an example of one such issue. One promising approach to tackling this issue is to use bacteria capable of degradation polymers, the main constituents in plastics.

"A bacterial taxonomy database organized in the form of an evolutionary tree is available," Ishii said. "Closely related living organisms often share similar traits. Based on this assumption, a bacterium that is closely related to other bacteria capable of decomposing polyolefins—polymers with carbon-carbon double bonds—may also have the ability to decompose polyolefins. To investigate this hypothesis, I'm working to integrate polymer, bacterial taxonomy, and enzyme/protein databases. This database integration will enable a systematic approach to addressing key issues (e.g., identifying polymer-decomposing enzymes, bacteria capable of producing these enzymes and other bacteria that are closely related to these bacteria)."

Ishii actually integrated these databases, analyzed the data, created separate evolutionary trees for polyolefin-decomposing and polyester-decomposing bacteria, and combined the trees. As a result, he was able to identify bacterial lineages potentially capable of breaking down both polyolefin and polyester (figure 2).

Ishii hopes to understand plastic decomposition mechanisms exhibited by different bacteria around the world and provide information useful in designing new, environmentally friendly plastics.

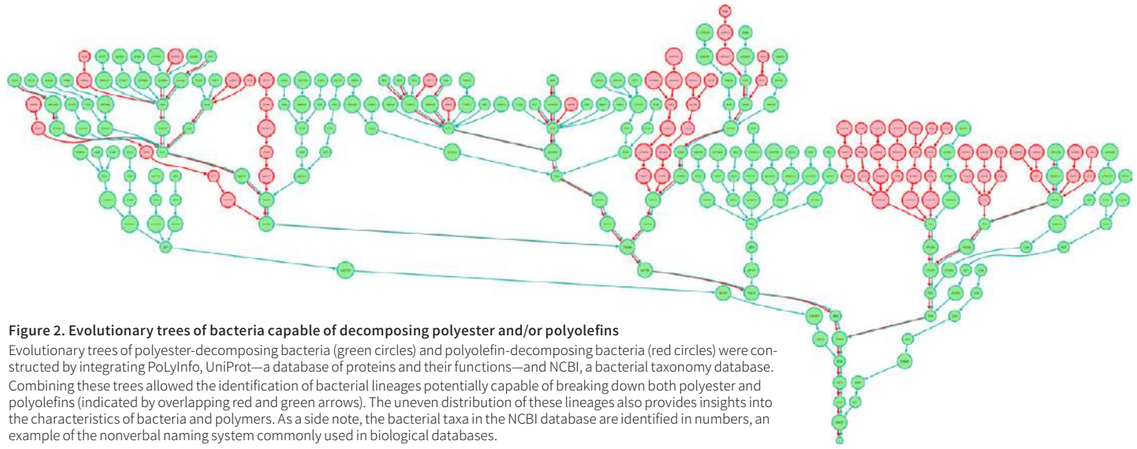


Figure 2. Evolutionary trees of bacteria capable of decomposing polyester and/or polyolefins
Evolutionary trees of polyester-decomposing bacteria (green circles) and polyolefin-decomposing bacteria (red circles) were constructed by integrating PoLyInfo, UniProt—a database of proteins and their functions—and NCBI, a bacterial taxonomy database. Combining these trees allowed the identification of bacterial lineages potentially capable of breaking down both polyester and polyolefins (indicated by overlapping red and green arrows). The uneven distribution of these lineages also provides insights into the characteristics of bacteria and polymers. As a side note, the bacterial taxa in the NCBI database are identified in numbers, an example of the nonverbal naming system commonly used in biological databases.

Bringing undiscovered information to light and passing it on to the next generation

Ishii has been working to derive useful information from materials science data and make it available to the public. However, he occasionally had doubts about his direction.

"I had doubts as to whether information generated in the past could be in any way useful in producing something positive and meaningful," Ishii said. "However, I first want to accurately recognize the fact that all the information generated in the past has formed the current knowledge, and then I hope for a future in which new knowledge is created on top of that. My mission is to make databases machine-readable, so that researchers can extract as much valuable information as possible from them."

What does passing information on to future generations mean? Ishii explained this using the example of the Rosetta Stone, an ancient inscribed stele discovered in Egypt.

"The Rosetta Stone was inscribed in three languages: the Egyptian hieroglyphic and Demotic scripts and Ancient Greek," Ishii said. "It was removed from a temple and its existence was completely unknown for hundreds of years until it was discovered by French soldiers. Extensive, long-term efforts to decipher it were then made. As a result, the term 'Rosetta Stone' is now used to refer to an essential clue to a new field of knowledge."

In addition to PoLyInfo, NIMS has various other databases. Making all of them machine-readable could help scientists develop new materials in the future. Ishii is leading this effort with the hope of promoting AI-based analysis by making large amounts of data accessible.

Leaders of the future

This article highlights the work of young researchers at NIMS who are establishing a foundation to support diverse materials development.

Yen-Ju Wu

Data Analysis Reveals Clues Hidden in Microscopic Images

Yen-Ju Wu

Researcher,
Data-driven
Inorganic Materials Group

I came to Japan after obtaining a postdoctoral position at NIMS. I then became a tenured researcher following my experience at the International Center for Young Scientists (ICYS). My research merges data-driven approaches with high-throughput experimental techniques to predict and validate the properties of inorganic materials, creating a highly efficient research cycle that accelerates the discovery process. Recently, I have focused on developing advanced materials with either superior thermal insulation capabilities or good ionic conductivity, aiming to understand the relationships between these materials' structures and their properties.

A key aspect of my research involves structural analysis of amorphous materials, which are difficult to analyze using traditional methods due to their lack of periodic atomic arrangements. Although it is possible to create amorphous materials with varying thermal conductivities, understanding the structural features that lead to these differences has remained difficult. In our recent studies, we used topological data analysis (TDA) to extract structural features from transmission electron microscope (TEM) images of amorphous germanium (a-Ge). TDA quantified the sizes of the "ring structures" (atomic chains) in a-Ge, revealing how deposition temperatures influence their formation and thermal conductivity.* Building on the insights gained through these data-driven methods, I will continue to challenge myself to develop materials that can address environmental issues and offer promising solutions to future needs.

*Related information: Press Release 2024.1.19
https://www.nims.go.jp/eng/press/2024/01/202401190.html

Naoka Nagamura

Naoka Nagamura

Senior Researcher,
Photoemission Spectroscopy Group

In-Depth Understanding of Surface and Interfacial States of Materials Using Innovative Measurement and Data Analysis Techniques

Microspectroscopy is a technique used to visualize surface and interfacial states in materials by irradiating these regions with light and measuring the interactions between the light and the material. I'm working to upgrade this technique to enable multidimensional spectroscopic measurements, such as simultaneous measurements of multiple parameters (e.g., spatial, temporal, and externally applied fields). At the SPring-8 synchrotron radiation facility, I quantified charge carrier mobility in an atomic-layer device during operation, using a modified scanning photoelectron microscope (SPEM). I'm also developing original laboratory instruments, including a multiprobe operando Raman microscope. I deliberately handle uneven, heterogeneous material specimens with variations in thickness and chemical element distribution. This approach allows me to collect data under conditions more closely reflecting those found in real industrial devices.

Besides conducting measurements, I focus on extracting useful physical property information from the collected data. I'm developing machine learning-based data analysis techniques to efficiently extract information from microspectroscopy data—a rich source. I developed an automated technique to rapidly analyze 1,000 to 10,000 spectral datasets generated by a SPEM, successfully detecting localized spectral changes that might be overlooked in manual analysis. Reflection high-energy electron diffraction (RHEED) is effective for characterizing crystalline material surfaces. However, RHEED data is complex and difficult to analyze. To address this, I developed an automated technique that analyzes RHEED data as accurately as data science experts. I'm skilled in taking measurements, analyzing data, and interpreting it to understand materials' physical properties and guide material design. I plan to develop diverse measurement techniques by approaching my research with curiosity and enjoyment.

Kosuke Nakano

Kosuke Nakano

Independent Researcher

Development of Next-Generation Computational Techniques for Higher-Accuracy Understanding of Material Properties

I've been developing a first-principles quantum Monte Carlo (QMC) technique—stochastic sampling-based methods for calculating electronic states—and making it useful for materials development. Today's widely used first-principles techniques (i.e., density functional theory (DFT) and wave function (WF)-based methods) have several drawbacks. QMC holds promise as a next-generation, high-accuracy computational technique.

QMC accurately calculates electronic structures but has struggled with estimating physical properties, while they are keys in materials design. I've been developing new methods and necessary codes to address this issue. For instance, I have recently established a new method to compute atomic forces and pressures using QMC. I'm also enhancing the versatility of our packages to benefit more materials developers. Our software package, TurboRVB, which implements QMC algorithms, is publicly available via a website.* I will continue this research to further contribute to the global community engaged in electronic structure calculations.

*http://turborvb.sissa.it

Yuichi Yamasaki

Yuichi Yamasaki

Team Leader,
Synchrotron Radiation Imaging Team

Development of New Synchrotron Radiation Microscopy Capable of Imaging the Invisible

I've developed many unique instruments using high-brilliance synchrotron X-rays to analyze materials with high resolution. My current focus is on analyzing antiferromagnets that can behave like ferromagnets under certain conditions. Typically, antiferromagnets do not exhibit a net magnetic moment. However, since discovering antiferromagnets with ferromagnet-like properties, I've investigated these antiferromagnets to understand the underlying mechanisms.

I've focused on X-ray magnetic circular dichroism (XMCD), which can analyze the electronic states of different chemical elements and magnetism. It was widely believed that XMCD measurements were not applicable to antiferromagnets. However, I've succeeded in theoretically clarifying the conditions under which XMCD is allowed in antiferromagnets, even without ferromagnetic order. Anisotropic magnetic dipole moments can explain why certain antiferromagnets display ferromagnet-like magnetic properties. I proposed this new theory and demonstrated its validity through experiments at a synchrotron radiation facility. Building on this success, I am now developing a soft X-ray microscope to image magnetic dynamics at the NanoTerasu. I believe many unexplored functions exist in materials, and I am eager to discover them by developing innovative measurement techniques.

*X-ray magnetic circular dichroism (XMCD) refers to the difference in X-ray absorption spectra of a material when irradiated with circularly polarized X-rays under different magnetic orientations. These differences arise from the alignment between electron spins in the material and the polarized X-rays.

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Institute Professor / Honorary Professor, Tokyo Institute of Technology
Distinguished Fellow, NIMS

Hideo Hosono

I have published a number of review articles and original research papers in STAM. I was pleased to learn that our 2010 review article on amorphous IGZO-TFTs has been cited 1,600 times. When we published our efforts to explore new superconducting materials in another review, I was even allowed to enlist unsuccessful materials thanks to STAM's unique policy. I find STAM to be a valuable resource for materials scientists.

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on the cover: TEM specimen holder for measuring thermal transport within materials (see p. 3 for details)

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