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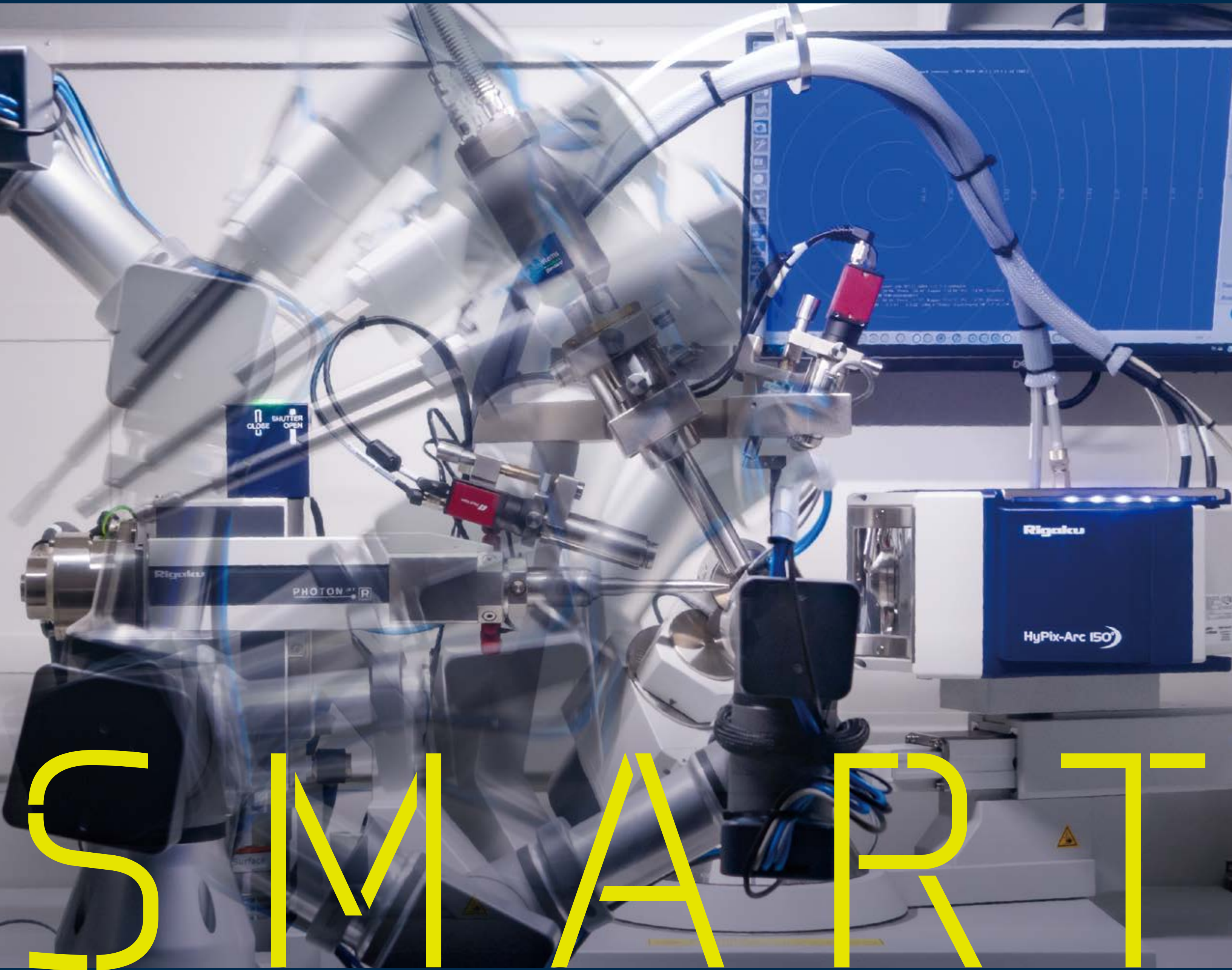


SMART LAB.

Advancing research automation

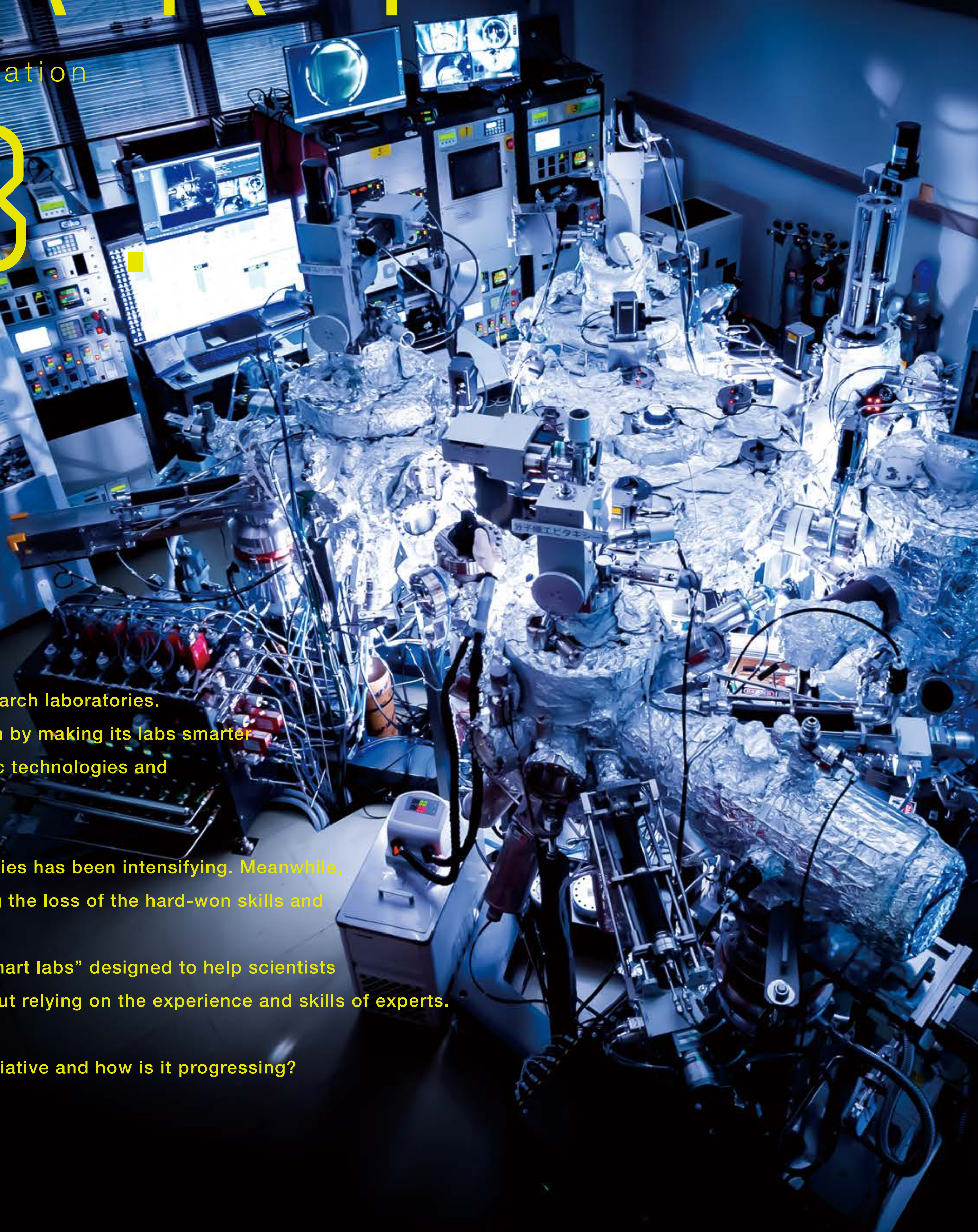
SMART LAB

Advancing research automation

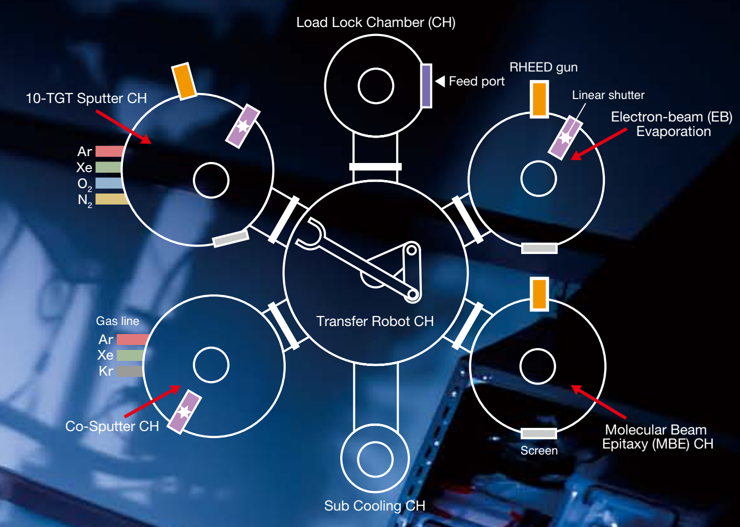
Major transformations are underway at NIMS research laboratories.
NIMS has been accelerating its materials research by making its labs smarter—some human labor has been replaced by robotic technologies and the "intelligence" of AI is being leveraged.

Competition in the development of new technologies has been intensifying. Meanwhile, the population of researchers is dwindling, risking the loss of the hard-won skills and expertise of retiring senior researchers.
These issues may be alleviated by developing "smart labs" designed to help scientists carry out high-quality research consistently without relying on the experience and skills of experts.

What NIMS strategies are behind its smart lab initiative and how is it progressing?
This issue will highlight the latest developments.



Fully-Automatic Cluster System for Magnetic Multilayer Deposition



Episode 1

Maximizing material performance using multiple film deposition techniques

NIMS has a system capable of making thin films using any combination of several different film deposition techniques. It consists of a group of machines taller than a person equipped with vacuum pumps housed in a high-ceiling lab. Shinya Kasai has been leading the development of this system.

The heart of the system is comprised of four film deposition machines: two types of sputtering machines, an electron beam physical vapor deposition machine and a molecular beam epitaxy machine. All of these machines are commonly used in materials development facilities. However, this system is unique in that they are all connected via a central chamber equipped with a substrate transfer robot (diagram above).

"The program-controlled substrate transfer robot automatically delivers individual substrates to the most appropriate film deposition machine depending on the type of material to be deposited on them," Kasai said. "This feature enables fully automated formation of thin films with complex multilayered structures." Kasai's group developed this integrated system to facilitate their efforts to develop effective magnetic tunnel junction (MTJ) devices—a key component of next-generation memory devices.

Exploring wide-ranging possibilities using various film deposition techniques

An MTJ device is composed of two ferromagnets separated by an insulator (i.e., a tunnel barrier). The device's electrical resistance changes with the changing electron spin orientation in the ferromagnets. This effect—called tunnel magnetoresistance—enables the device to store the binary numbers 0 and 1. MTJ devices are used in magnetic hard disk read heads. Integrated MTJ devices are also found in the cells of non-volatile magnetoresistive random access memory (MRAM), which is able to retain stored information even after the power supply is cut off.

An integrated MTJ device consists of a stack of approximately 60 thin films each with a thickness in the nanometer range. They include a tunnel barrier only three to four atoms in thickness sandwiched between ferromagnets. To greatly increase the performance of this device, new thin film materials need to be discovered. Heterojunctions in the device (i.e., interfaces be-

Film synthesis is controlled by a computer and film deposition status can be monitored on the display above. "Although automation allows us to conduct experiments more efficiently, it also makes fabrication processes incomprehensible—a so-called 'black box,'" Kasai said. "To overcome this disadvantage, we've installed a fabrication process monitoring device."

tween layers of different materials) need to be optimized by improving the quality of the thin film surfaces that form heterojunctions (e.g., crystalline orientations and surface evenness). To enhance heterojunction quality, the selection and use of optimum film deposition techniques are indispensable.

Sputtering is currently the most commonly used film deposition technique in the industrial sector due to its ability to form a film over a large surface area at low cost. "However, for the purpose of speedily evaluating the potential performance of new materials, the use of various other film deposition techniques, in addition to sputtering techniques, is crucial," Kasai said.

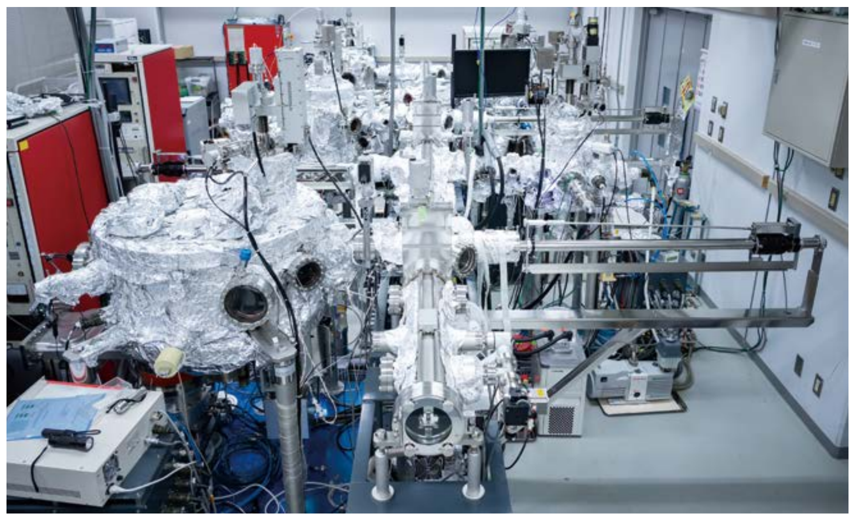
"Magnesium oxide (MgO) compounds are currently used as tunnel barrier materials in MTJ devices," Kasai said. "When MgO barriers are formed using sputtering on the surfaces of certain types of materials, their physical properties are severely inhibited. However, when they are formed by electron beam physical vapor deposition, they exhibit significantly more desirable properties. In basic research that aims to explore the wide range of potentialities of materials, all available types of film deposition methods should be considered."

Making a 20-year-old system smarter

Enhancing the performance of materials by combining different film deposition techniques is not a new idea. In fact, NIMS has spent many years achieving this. The previously developed film deposition system still exists in another NIMS lab. This system was initially equipped with only a single sputtering machine. It has since undergone a series of expansions to make it compatible with more diverse materials and film deposition processes. As a result, the system has grown to 8 m in length with a sample transport tunnel stretching down the middle and various film deposition machines placed on its sides (photo at left). A manually operable carrier inside the tunnel is used to transfer samples from one machine to another. This system is capable of forming heterojunctions using a combination of different film deposition techniques while keeping the interfaces clean.

"We've discovered many new materials and gained new insights about film growth processes using this older system," Kasai said. "However, operating it is very labor intensive. For example, in order to prepare a single sample, researchers need to manually transfer it to different machines every few hours to subject it to thermal treatment and film deposition."

Kasai's group has been working to automate the new system to the extent possible. Based on information and experience gained from operating the older system, Kasai designed every potentially useful process he could imagine to be programmable, enabling system users to preset various parameters (e.g., the distance between the substrate and the target material, types of gases to use and film deposition temperature).



First-generation film deposition system with a sample transport tunnel stretching down the middle. The system is equipped with three types of sputtering machines and an electron beam physical vapor deposition machine.

Kasai also designed the program to be expandable, making it easier to add new functions to the system. He actually has already added a series of new instruments to perform electron beam diffraction analysis, plasma-induced emission energy evaluation and other analyses.

"In line with NIMS' vision to develop smart labs, we built this system to be able to fabricate film samples significantly more efficiently and speedily—about four times faster than the previous system," Kasai said. "It can also deposit a thin film on a substrate using a combinatorial chemistry approach so that you can gradually change the film's composition along its length. This feature allows us to assess a number of different conditions using a single sample." This robust system is expected to facilitate Kasai's research and contribute to the development of next-generation memory devices.

Kasai has been working with Hiroaki Sukegawa (left), a NIMS principal researcher, on the development of new scientific instruments. "Dr. Sukegawa is the world's leading specialist in magnetic thin film deposition," Kasai said. "I regularly have discussions with him to take advantage of his expertise in developing technologies relevant to our research."

Ultimate goal: full automation of all processes

The new system has been in operation for nearly two years. Kasai is currently working to optimize the structures of ferromagnetic layers deposited using sputtering techniques. In this project, he uses a technique to deposit two-dimensional thickness gradient films. In addition, Kasai developed a probe system capable of automatically taking electrical resistance measurements from hundreds of microfabricated devices integrated with electrodes arranged in a grid on the substrate with the aim of expediting the evaluation of ferromagnetic layer performance when they are integrated into devices. Using this probe system, he measured the tunnel magnetoresistance (TMR) ratios (the most important indicator of MTJ device performance) of individual minute devices with different thicknesses and mapped the TMR ratio distributions across the substrate (figure). This project quickly found that an MTJ device into which CoFeB ferromagnets are integrated can yield a TMR ratio close to the world record. Kasai's group is currently attempting to break this record.

Some decisions related to film deposition processes (e.g., types of materials to be deposited and film deposition conditions) still need to be made by researchers, requiring a significant amount of human intervention.

"I'll continue to evaluate the transport properties of the micro-fabricated devices on substrates and feedback into the film deposition processes to accumulate data in the hope of developing an even more advanced film deposition system capable of automatically recommending new materials and optimizing film deposition processes," Kasai said. With this goal in mind, Kasai has already begun compiling evaluation data into a database and constructing analytical models.

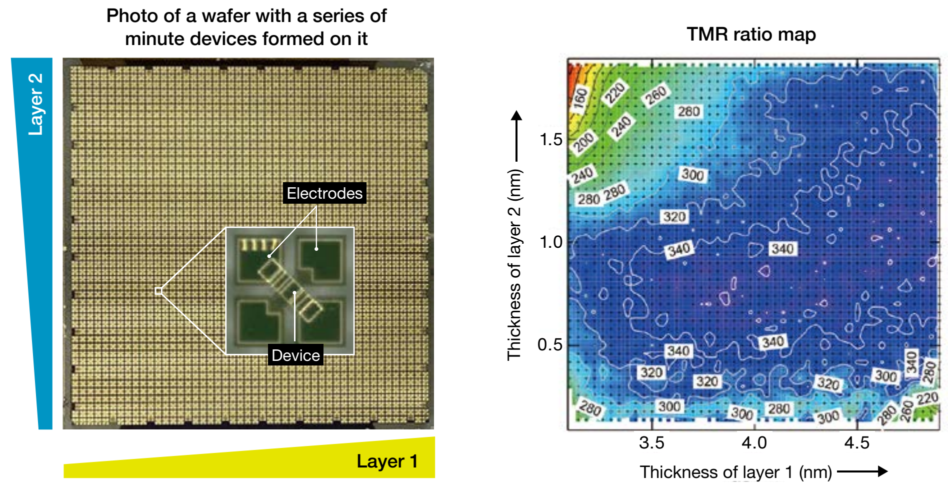
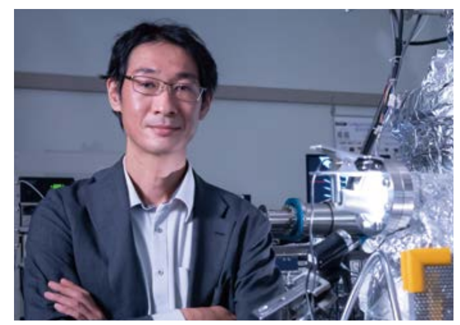


Figure. Multilayered thin film formed using a technique to deposit two-dimensional films of varying thicknesses

A multilayered thin film containing two layers of varying thicknesses (layers 1 and 2) was formed and then used to fabricate a series of minute devices arranged in a grid (left). The tunnel magnetoresistance (TMR) ratios of these devices were measured using an automated probe system and their distributions across the grid was mapped (right). The map indicates that TMR ratios depend on the thicknesses of layers 1 and 2.

"I plan to improve the current film deposition system and eventually automate all of the processes it performs, including various measurements. That's my ultimate goal," Kasai said. The use of a smarter system is expected to accelerate the search for effective MTJ device materials vital to the development of next-generation memory devices.

(by Kumi Yamada)

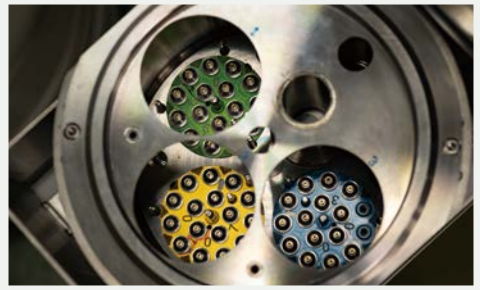


Shinya Kasai
Group Leader
Spin Physics Group
Research Center for Magnetic and Spintronic Materials

Sixteen glass capillaries placed on a disk approximately 7 cm in diameter. A single phosphor particle is bonded to the pointed tip of each capillary.

Step3

Particles with desired characteristics are individually examined to determine whether they are novel materials. For this purpose, their lattice constants are first determined using an X-ray diffractometer (XRD).



XRD sample holder (left) and single particle measurement in action (right). The sample holder can carry 48 glass capillaries at once. A robotic arm picks up one glass capillary with a phosphor particle attached to its tip from the sample holder and transfers it to the sample stage. The transferred capillary is then held in place magnetically on the stage. This new XRD has eliminated the need to manually affix a capillary to the stage using a screw. Automation has been implemented in all of the steps in the single particle diagnostic procedure, from focusing an X-ray beam on a particle to be measured to determining whether a particle is a novel material. If the particle is found to be potentially novel, more detailed analysis **Step 4** will be carried out.

Reproducing the skills of experts 1

Examining individual phosphor particles in search of novel luminescence properties

Light-emitting diodes (LEDs) have become a standard lighting technology. Sialon phosphors* developed by NIMS played a major role in this development. Earlier white LEDs were unpopular because the white light they emitted lacked an adequate red component and had an unnatural appearance. The integration of a red sialon phosphor into white LEDs improved their luminescence properties, rapidly increasing their popularity around the world as lighting devices and liquid crystal display backlights. Takashi Takeda has been leading the development of phosphors at NIMS. He believes that phosphors still have a lot of room for improvement.

"For 8K ultra-high-definition television to have a wider color reproduction range, more vivid green colors need to be added," Takeda said. "In addition, new phosphors need to be developed for use in laser lighting devices, which can emit more intense light than LEDs."

Takeda and his colleagues developed a single particle diagnostic procedure (the figure at lower right on p. 7) as a method of identifying novel phosphor particles in large quantities of sintered phosphor powder. In this procedure, phosphor particles are individually analyzed for their physical properties, including crystalline structures and chemical compositions. Particles with desired properties are then examined to determine whether they are novel materials. While earlier technology was able to analyze only large particles, more recent, improved technology can analyze smaller particles, including particles 10 microns in size. When phosphor particles are found to be novel, they are subjected to potential mass synthesis. Takeda's group has discovered more than 30 novel phosphors since incorporating data science techniques into the single particle diagnostic procedure.

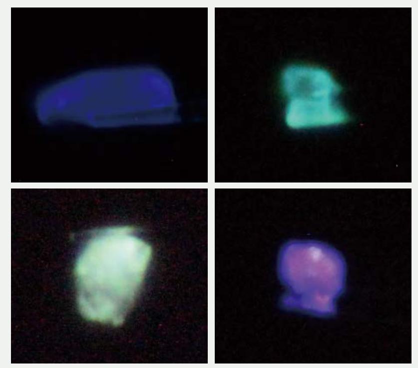
However, this procedure (i.e., selecting promising phosphor particles from large quantities of sintered phosphor powder and analyzing them individually) is extremely labor intensive. Technicians tasked with this role need to be highly focused and skilled.

To improve this, Takeda's group has worked to make each step of the procedure more efficient. Promising phosphor particles were previously selected manually under a microscope based on their luminescent colors and crystalline sizes and shapes. The group eased this process by introducing a system capable of automatically measuring the luminescence properties of individual particles and ranking them accordingly **Step 2**. The group also introduced an X-ray diffractometer (XRD) capable of performing simplified analysis of each particle **Step 3**, which was found to be particularly effective in expediting the diagnostic procedure.

"The previous XRD needed to be operated manually: a glass capillary with a single phosphor particle attached to its tip (photo above) was manually affixed to the XRD sample stage using a screw. An X-ray beam was then manually focused on the particle to be measured and the capillary on the sample stage was manually replaced with a new one after each measurement," Takeda said. "We replaced this XRD with a fully automated one which performs all of these tasks independently. We also constructed a computer program designed to determine whether a phosphor particle of interest is a novel material based on measurements taken by the XRD. The use of this program has greatly expedited our search for novel materials." Takeda's efforts to discover materials with groundbreaking luminescence properties will continue.

GALLERY

Some of the phosphors discovered through the single particle diagnostic procedure



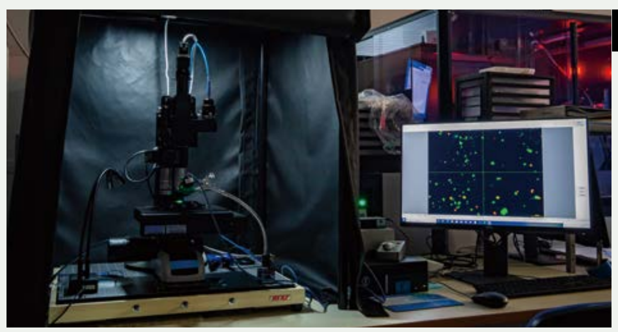
Takashi Takeda

Group Leader
Luminescent Materials Group
Research Center for Functional Materials

* A sialon phosphor is composed of a ceramic host material doped with light-emitting rare earth elements (e.g., europium and cerium). It absorbs light energy, converts it into light of a longer wavelength and emits it.

Step1

Sintered phosphors vary in luminescence color and intensity; some even emit no light at all. In the single particle diagnostic procedure, a number of phosphor powder samples are first measured for their luminescence spectra using a plate reader. Samples are then selected for further analysis.



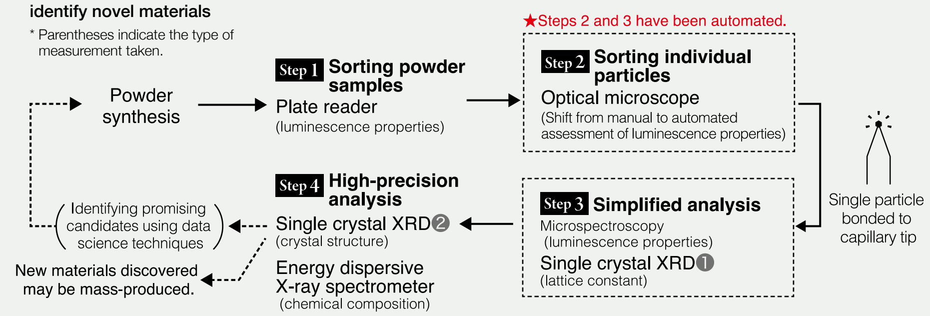
Step2

A selected powder sample is then placed on a glass slide and exposed to ultraviolet light. Individual particles with desired luminescence properties are then isolated. This new microscope capable of pattern recognition automatically focuses on individual particles and measures their luminescence properties.

"Human eyes are amazingly accurate and quick in determining the luminescence characteristics of particles, such as brightness," Takeda said. "However, visual abilities vary among individuals, and the chance of human error increases naturally with fatigue. This microscope is capable of replacing human labor and measuring each and every particle."

Figure. Single particle diagnostic procedure designed to identify novel materials

* Parentheses indicate the type of measurement taken.



Episode2

A close look at automated energy materials search systems

Hydrogenproduction

Next-generation rechargeable batteries

The NIMS Automated Robotic Electrochemical Experiments (1st NAREE) —featured in previous NIMS Now issues—leverage robotic technology for accelerating discovery of electrolyte for high-performance lithium–air batteries. While this system is efficient, NIMS has also developed even smarter materials search systems. Let's look at how they work.

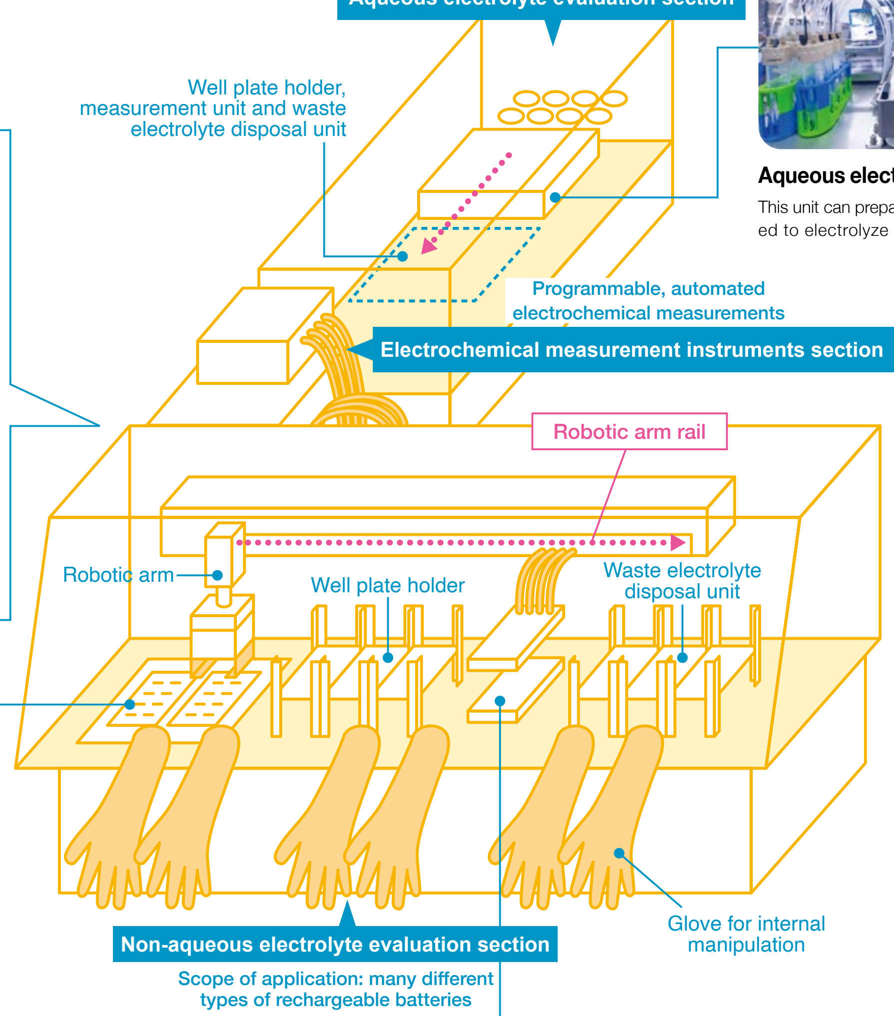
1st NAREE (quick review)

The main purpose of this system is to identify potential candidate of electrolyte exhibiting high-performance lithium–air battery. Each hole in the microplate (photo) contains an "electrochemical cell" composed of electrode sheets and a separator between them. The addition of an electrolyte into electrochemical cells make them tiny rechargeable battery cells. A plate of rechargeable cells is then transferred by a robotic arm to the measurement section where the cells' charge/discharge capacities are measured. 1st NAREE's ability to automatically perform this electrolyte preparation–measurement cycle enables it to evaluate about 1,000 electrolyte samples a day (see NIMS Now Vol. 20, No. 2).

Robotic arm carrying a microplate based electrochemical cells

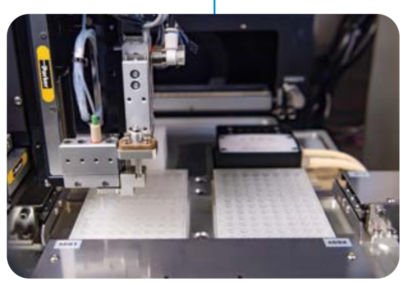
2nd NAREE

2nd NAREE is comprised of three sections developed using 1st NAREE as a model. It can take more types of measurements from a wider range of battery materials than 1st NAREE and is able to evaluate electrolytes for various types of rechargeable batteries in addition to lithium–air batteries. The system can also evaluate materials for non-rechargeable batteries, such as electrode materials needed to efficiently produce hydrogen by electrolyzing water. 2nd NAREE's various instruments enable more detailed study of battery mechanisms.



Aqueous electrolyte preparation unit

This unit can prepare a variety of solutions needed to electrolyze water or for other purposes.



Non-aqueous electrolyte preparation unit

The microplate on the left contains 96 different types of electrolyte ingredient compounds. Using suction devices, they are transferred to the plate of electrochemical cells on the right with individual cells receiving different combinations and ratios of the compounds.

Electrochemical measurement unit

Measurements are taken from electrochemical cell samples sandwiched between the top and bottom electrodes. The types of data collected can be modified by programming the electrochemical measurement instruments section.

Electrochemical measurement instruments section

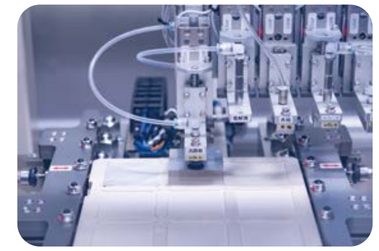
This section houses various instruments used to take measurements from samples prepared in the aqueous and non-aqueous electrolyte evaluation sections. These measurements—including charge/discharge capacities and other electrochemical measurements (e.g., impedance and linear sweep voltammetry (LSV))—can be programmed in various ways.

3rd NAREE

Although various kinds of promising candidate for electrolyte has been discovered by 1st NAREE, some of them do not showed expected superior performance in cell with practical laminated cell condition. To minimize such gap, NIMS developed **a laminated battery cell production system** (3rd NAREE) capable of evaluating electrolytes under conditions more similar to those found in practical use. A laminated cell is produced in a drying room by stacking a positive electrode, separator and negative electrode, injecting an electrolyte and laminating and sealing it. Even talented technicians can fabricate only six laminated cells a day at best. In contrast, 3rd NAREE can produce 80 cells a day.

Electrode rack

The rack at left holds plates containing positive electrodes, negative electrodes and laminate sheets. A separator—the long roll of white material at right—is automatically dispensed and cut into individual pieces.

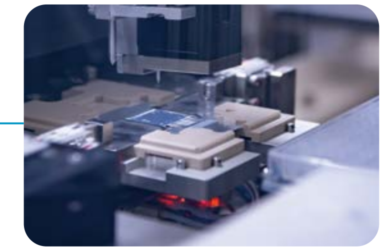


Electrode transfer arm

The arm lifts a material from the electrode rack using a suction cup and transports it to the stacking area at the center of 3rd NAREE. The suction strength of the arm is controllable, ensuring safe transfer of even extremely thin electrode materials.

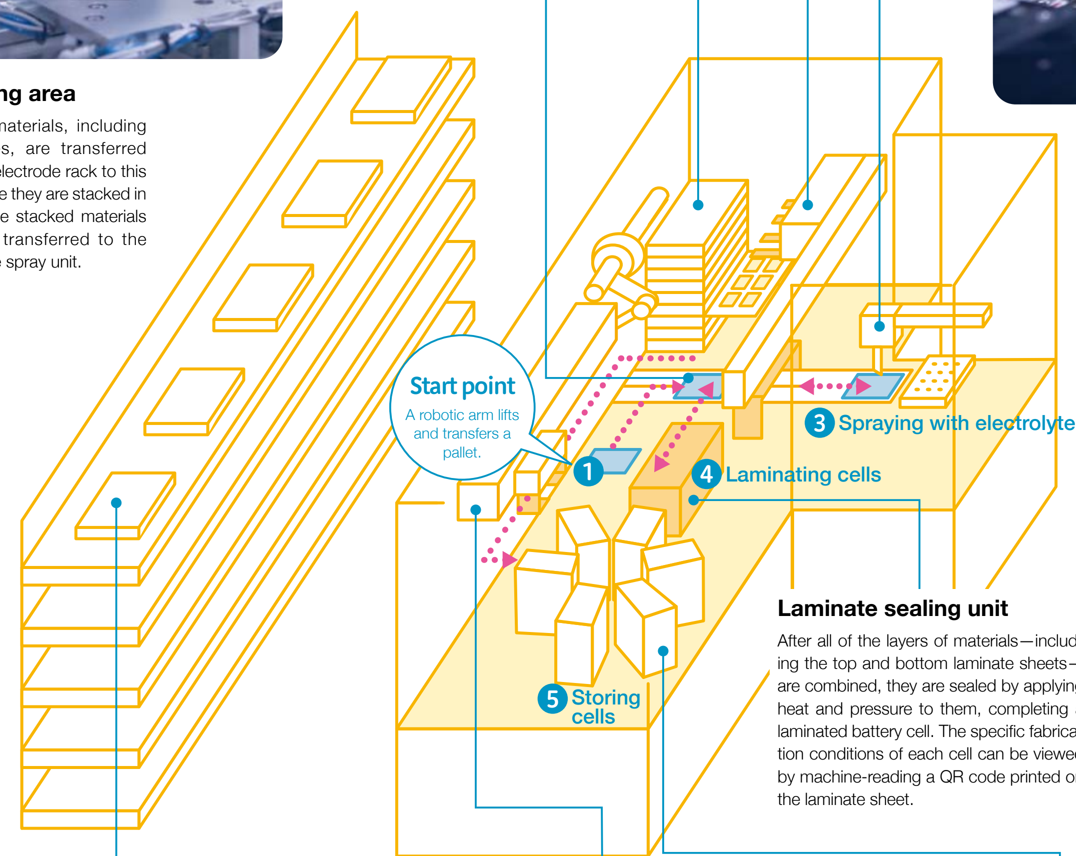
Stacking area

Various materials, including electrodes, are transferred from the electrode rack to this area where they are stacked in layers. The stacked materials are then transferred to the electrolyte spray unit.



Electrolyte spray unit

Each electrode transferred from the stacking area to this unit is sprayed with an electrolyte prepared beforehand in the form of a fine mist. A specified amount of electrolyte is applied evenly to every part of the electrode by slowly repositioning it while it is being sprayed.

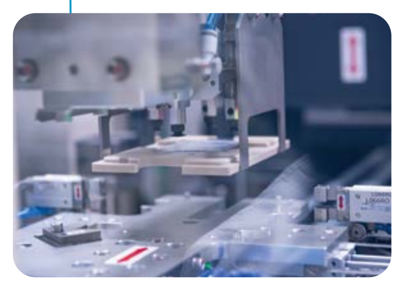


Laminate sealing unit

After all of the layers of materials—including the top and bottom laminate sheets—are combined, they are sealed by applying heat and pressure to them, completing a laminated battery cell. The specific fabrication conditions of each cell can be viewed by machine-reading a QR code printed on the laminate sheet.

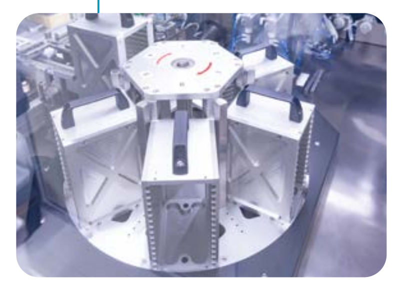
Battery performance evaluation area

The cells transferred to this area from the battery cell holder are manually connected to electrodes. The charge/discharge capacities of as many as 400 cells can be measured simultaneously.



Battery cell transfer arm

This arm transfers sealed cells to the battery cell holder.



Battery cell holder

Laminated cells are stored in these holders until they are used for measurements.

NIMS Automated Robotic Electrochemical Experiments (1st NAREE) —an high throughput screening system designed for searching high-performance electrolytes for lithium–air batteries—have been operating for nearly four years. In early days of 1st NAREE's operation, Shoichi Matsuda—a leader of battery-related systems development—often brought output data generated by the system to data scientists for consultation for deciding on the next experimental plan. The system has since been fully automated. "The integration of AI into 1st NAREE has enabled it to automatically and continuously update its prediction models as it generates more data," Matsuda said.

1st NAREE has produced some significant results, including the 2021 discovery of an electrolyte with a new chemical composition. The use of this electrolyte was demonstrated to double the cycle life of lithium–air batteries. This discovery was made possible by introducing Bayesian optimization-based AI prediction models into 1st NAREE. The enhanced system was able to identify the optimum composition out of more than 10 million candidate compositions by analyzing more than 10,000 electrolytes. This extensive analysis was completed in only about three months. The combined introduction of AI and robotic technologies has proven to be very effective.

Several issues with 1st NAREE remains to be solved, however. "Although 1st NAREE is capable of identifying high-performance multi-component electrolytes, it doesn't explain the mechanisms behind their high performance or how they will work within battery cells," Matsuda said. "I realized that more detailed measurements would be needed to find scientific answers to these questions."

Matsuda also faced other challenges. Several companies asked him whether he could evaluate materials other than lithium–air batteries. "There are many products and technologies other than rechargeable batteries whose performance is significantly influenced by electrochemical reactions taking place between solutions and electrodes," Matsuda said. "Because 1st NAREE was designed specifically to analyze electrolytes for lithium–air batteries, I was unable to accommodate their requests. This motivated me to build 2nd NAREE."

2nd NAREE: in-depth analytical capabilities

Unlike 1st NAREE , which specializes in measuring charge/discharge capacities, 2nd NAREE is capable of gathering a wide variety of information and can be programmed to perform any combination of several different electrochemical measurements. For example, electrochemical impedance measurements provide information about differences in electrochemical reaction rates at the interface between electrode and electrolyte, which is useful in identifying specific electrochemical reactions most influential to the performance of battery cells.

Moreover, 2nd NAREE is designed to evaluate electrode materials in addition to rechargeable battery electrolytes. One of Matsuda's current research interests concerns electrode materials that can be used to produce hydrogen by electrolyzing water. Although most water electrolysis research has focused on breaking down fresh water, Matsuda has been targeting seawater—a more abundant resource. "Even if an effective fresh water electrolysis technology is developed, its practical use would likely to be limited because fresh water is a scarce resource in many regions in the world," Matsuda said. "Seawater contains various substances (e.g., chloride ions) known to impact the catalytic activities of electrodes. I'm therefore looking for electrode materials capable of functioning stably in the presence of these substances. In addition, I need to address various other issues, such as additives able to improve electrodes' catalytic activities. In these efforts, 2nd NAREE is capable of rapidly assessing a huge number of samples very efficiently."

3rd NAREE: capable of mass-producing battery cells of consistent quality

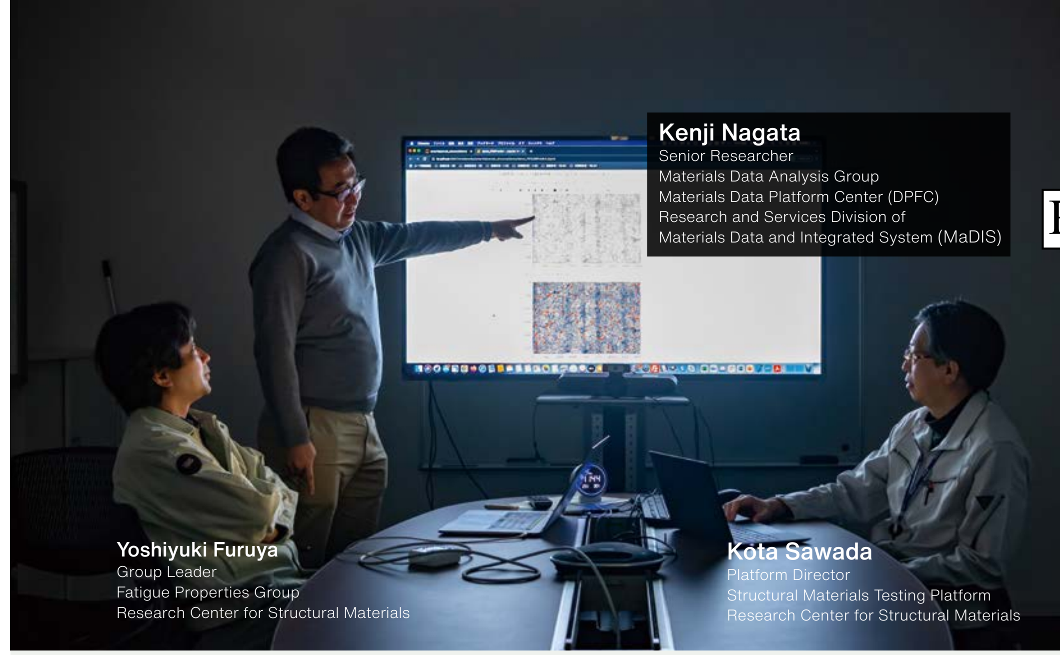
Unfortunately, many promising results found in material level is not easily transferred to high performance at practical cell level. This was also the case for lithium–air battery research. "Some of the promising electrolytes we discovered using 1st NAREE were unable to meet the strict conditions required for practical cell condition, such as the smaller electrolyte volumes permitted, the need for thinner electrodes and the need to generate larger amounts of electricity through electrochemical reactions," Matsuda said. "To evaluate the performance of electrolytes under conditions more similar to batteries in practical use, we developed 3rd NAREE—an automated laminated battery cell production system."

The production of a lithium–air battery—stacking a porous carbon based positive electrode, separator and lithium negative electrode and injecting an electrolyte—sounds deceptively simple and would seem achievable using commercially available equipment. However, there are some challenges and tricks to producing them. For example, they are composed of delicate materials: the porous carbon electrode is fragile and the metallic lithium electrode is thin and prone to twisting and bending. These materials need to be stacked in perfect alignment in a single step. Moreover, a very small amount of an electrolyte needs to be continuously and uniformly applied to the entirety of these materials. 3rd NAREE—which NIMS has developed using its accumulated cell production know-how—is able to carry out these high-precision processes.

3rd NAREE has been in full-scale operation since April 2021 to meet the need to mass-produce battery cells of consistent quality similar to those in practical use. Matsuda's goal is to discover battery materials that will perform outstandingly in practical use. Expectations are high for him to find new materials effective in resolving energy-related issues in the near future using the systems featured here.

Matsuda being interviewed next to 2nd NAREE

Shoichi Matsuda
Senior Researcher
Solid-State Battery Group
Center for Green Research on Energy and Environmental Materials



Kenji Nagata
Senior Researcher
Materials Data Analysis Group
Materials Data Platform Center (DPFC)
Research and Services Division of Materials Data and Integrated System (MaDIS)

Yoshiyuki Furuya
Group Leader
Fatigue Properties Group
Research Center for Structural Materials

Kota Sawada
Platform Director
Structural Materials Testing Platform
Research Center for Structural Materials

Reproducing the skills of experts 2

Sawada, Furuya and Nagata commented similarly about Endo, who has been engaged in building the AI for this project. Although he had no experience with metallic materials when the project began, he greatly exceeded their expectations by developing a great ability to assess the cross-sectional surfaces of fractured metal pieces. Scientists can use this diagnostic AI model to learn how experts diagnose metal fractures in an evidence-based manner. It is expected to become a very useful educational tool.

Determining metallic material durability and the causes of accidents using diagnostic AI

When investigating an accident involving metallic component fracturing, observing the microscopic patterns on the cross-sectional fracture surfaces can offer clues about the cause. These observations also provide useful information about the service lives of metals. Materials that are persistently exposed to high temperatures and pressure—such as steam piping in a thermal power plant—experience a type of deformation called creep. The remaining service life of steam piping can be estimated through microstructural observation by skilled experts during plumbing inspections. Developing successors to these experts is an urgent issue.

To address this issue, a "master's eye" AI development project was launched. In this project, two types of AI models are being developed. The first is an accident investigation AI capable of replicating the diagnostic skills of Yoshiyuki Furuya, who has helped find the causes of many of the accidents NIMS has been assigned to investigate. The second is a service life estimation AI capable of mimicking Kota Sawada, who has more than 20 years of experience monitoring creep damage. Akihiro Endo (third-year doctoral student) is responsible for constructing machine learning models under the guidance of Hayaru Shouno, a professor at the University of Electro-Communications. Kenji Nagata—a machine learning expert who has been assisting with data-driven materials development at NIMS—is serving as an "interdisciplinary translator" for this project. The project members first aimed to develop AI systems applicable to carbon steel and stainless steel because of their wide-ranging applications.

"Teaching AI how to assess the cross-sectional surfaces of metal specimens was challenging initially," Nagata said. Several different morphologies of fracture surfaces -including dimple- are relevant to accident investigations. They cause microscopic patterns to develop on the cross-sectional surfaces of fractured metallic components. "Sometimes, what appear to be dimples may not actually them. Their appearance is also sometimes influenced by material compositions and electron microscopic measurement conditions," Furuya said. Sawada brought up another issue: "The key to estimating the service life of a metallic component is precipitate measurements, such as their sizes and distributions (figure 1). These pieces of information allow us to estimate the temperature to which the metallic components had been exposed (i.e., exposure temperature). This information is vital to estimating their service lives. However, microstructures had traditionally been assessed visually by comparing them with a number of previously examined microstructures without quantifying the microscopic patterns."

To identify characteristics used by experts in their fracture diagnoses, the AI development team, including Endo, examined previously captured fracture surface images with marks left on them by diagnostic experts. After carrying out many case studies using texture analysis—an image processing method designed to quantify the perceived texture of an image—and other techniques, the team ultimately selected a set of "features"—individual measurable properties or characteristics of a phenomenon in machine learning. Using these features, the team was able to improve the estimation accuracy of the AI models.

The team also developed an effective AI diagnostic procedure. Before a fracture surface image is assessed by the accident investigation AI, it is first divided into a fine grid (i.e., sections) and the type of fracture associated with each grid section was then determined. Diagnosis is then performed using the entire image by compiling the results of the individual grid section evaluations and comparing the proportions of different fracture types associated with the grid sections (figure 2). Using this procedure enabled the AI to answer correctly nearly 100% of the time.

With regard to the service life estimation AI, Sawada said, "It's now able to estimate the temperature to which stainless steel components had been exposed with a margin of error of plus or minus 10°C." Adequate estimation of the exposure temperature and stress applied to specimens will enable their service lives to be estimated.

For these AI models to produce diagnostic results, they need only be fed a image and other associated data, including measurement conditions. The team plans to commercialize these models and expand their applicability to a broader range of materials. These AI models are expected to become available as successors to diagnostic experts or to provide second opinions in the near future.

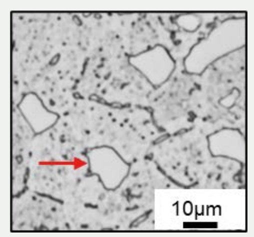


Figure 1. Extraction of features from images by the service life estimation AI

The parameters of each deposit (indicated by the arrow)—including its size and circumference, its distance from the nearest deposit and its degree of circularity—were measured and then fed into mathematical formulas used in machine learning.

Microstructure of a stainless steel after creep exposure

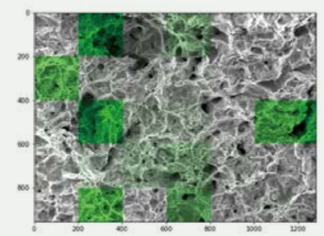


Figure 2. Diagnostic procedure by the accident investigation AI

A fracture surface image obtained using an electron microscope is divided into sections (a 200 x 200 pixel grid). Each section is then assessed for the probability of dimpling (lower probabilities are represented by darker green). Finally, the cause of the fracture is diagnosed based on overall statistics across all of the sections. The image above was used by the AI to diagnose the probable cause of the breakage as dimple fracturing.

Patterns detected on a fracture surface

Episode3

Combining combinatorial synthesis and AI to efficiently search for new electronic device materials

NIMS has been working to introduce automation into its materials research to expedite its efforts to discover novel materials out of a vast number of candidate materials. For example, in its search for new electrolytes, NIMS has adopted robotic technologies to perform a series of processes from materials synthesis to evaluation. NIMS has also introduced artificial intelligence (AI) techniques designed to select candidate materials. By introducing a combination of AI and robotic technologies, NIMS succeeded in developing fully automated electrolyte materials search systems (see p. 8).

Takahiro Nagata has been researching inorganic thin film materials for use in electronic devices. His specific focus has been to discover dielectric materials needed for the development of next-generation mobile communications systems beyond 5G and 6G technologies. "Because electronic device materials with a wide range of physical properties are required, we're taking a new approach to developing efficient materials search processes which differs from the approach used for electrolyte materials," Nagata said.

"Mobile communications systems beyond 5G and 6G technologies will require inorganic thin films capable of withstanding high temperatures and high-frequency electromagnetic waves," Nagata said. "Because these thin film materials have a variety of functions, they may also be effective in applications other than mobile communications. To explore this possibility, various types of measurements need to be taken from these materials. In our efforts to search for materials more efficiently, our group has long employed combinatorial synthesis, which enables us to rapidly produce large amounts of combination thin film samples of various elemental compositions. We've also been taking physical properties measurements from materials using several different lab instruments. We're currently seeking to further expedite our materials development using the materials search process we have developed without major modifications to our current measurement practices. In this effort, we're working to find ways of combining data generated by different measurement instruments and analyzing it using AI techniques (Figure 1 on p. 13) in collaboration with the NIMS Research and Services Division of Materials Data and Integrated System (MaDIS).

Automating measurement data collection and AI-based data analysis

Nagata's group has been working to combine combinatorial synthesis and AI in cooperation with MaDIS. Combinatorial synthesis enables the fabrication of a thin film which changes gradually in composition across a substrate (Figure 2 on p. 13). This technique allows quick and systematic materials evaluation. For example, a single thin film deposited on a three-inch substrate using this technique can be used to evaluate 400 different thin film compositions. Nagata also introduced some other technologies to accelerate thin film compositional evaluation, including a prober capable of automatically repositioning itself to take physical property measurements from various positions on a thin film.

For an AI system to efficiently analyze measurement data, large amounts of data generated by different instruments needs to be compiled. To achieve this, Nagata worked to automate lab data aggregation in collaboration with Hideki Yoshikawa and other data utilization platform developers at MaDIS.

"MaDIS has been developing a Research Data Express (RDE) system—part of its materials data platform (DPF)—capable of automatically collecting large amounts of data generated by measurement instruments at research labs and compiling them into databases," Nagata said. "Data formats and terminology vary widely between different instruments. The RDE system standardizes the formats and terminology of data generated by different instruments before transmitting the data and associated metadata (e.g., experimental conditions and instruments used) to the DPF server. To introduce an RDE system into our lab, we first needed to closely assess the characteristics of data generated by individual instruments with DPF developers so that the RDE would be able to properly process the data. As a result, we are now able to automatically transfer basic thin film physical property data (e.g., chemical compositions, crystalline structures, permittivity and electrical resistance) generated by our lab instruments and stored on dedicated PCs to databases on the DPF server."

According to Nagata, the installation of the RDE system—which compiles the measurement data generated by different instruments—has significantly improved the efficiency of research activities at his lab. The system is equipped with data visualization capabilities (i.e., representation of data in graphic form), enabling researchers to visually analyze data much more quickly than they can using software dedicated to individual

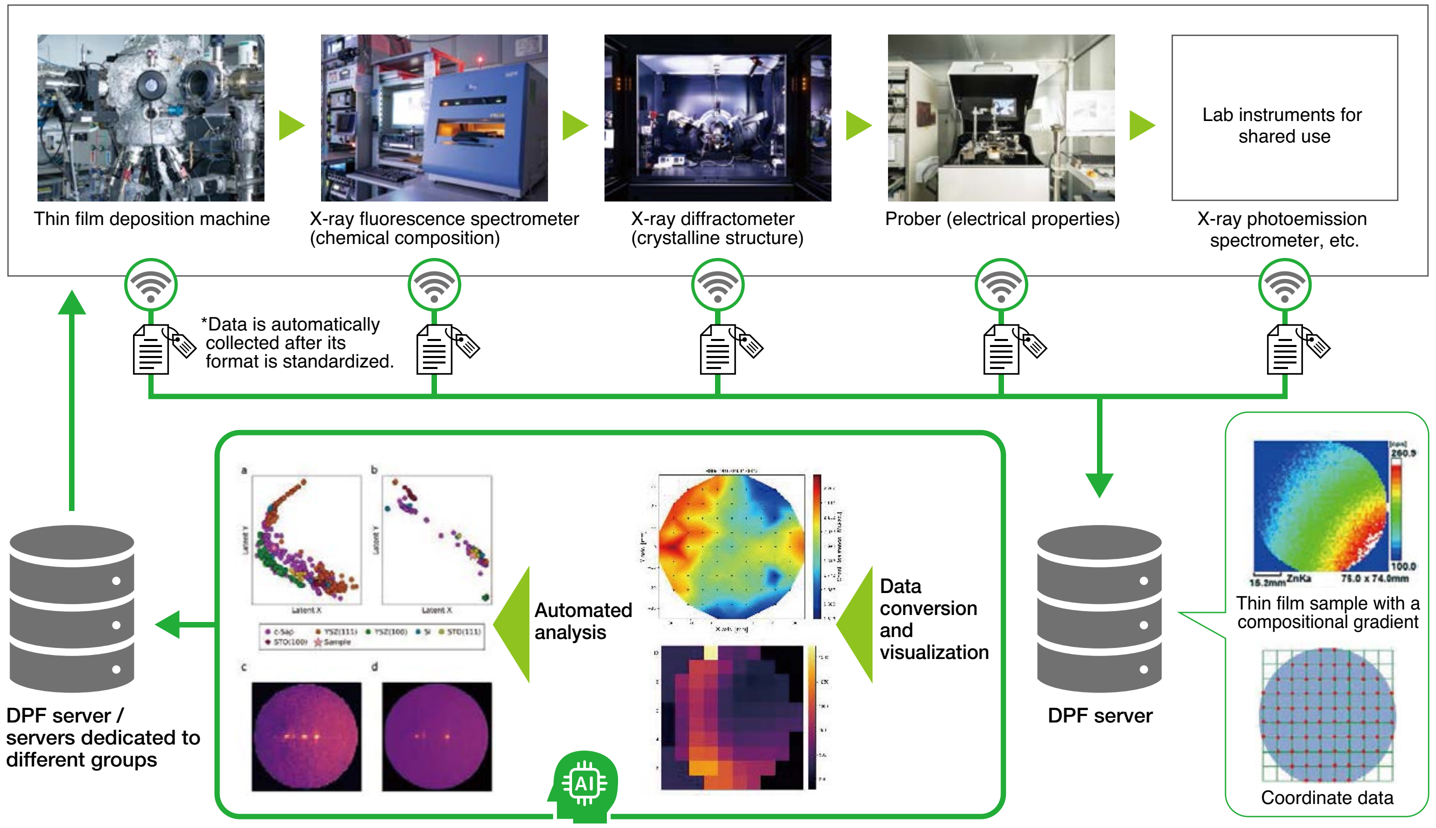
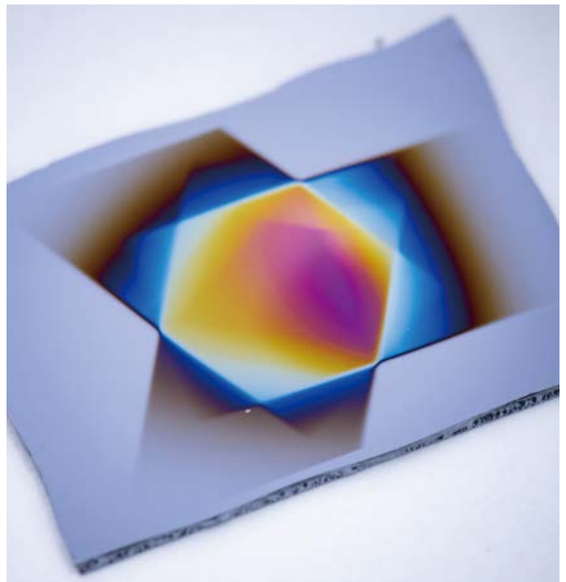


Figure 1. Integration of combinatorial synthesis and AI into thin film materials search processes



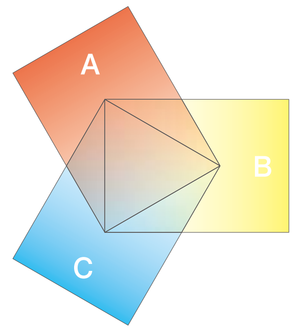
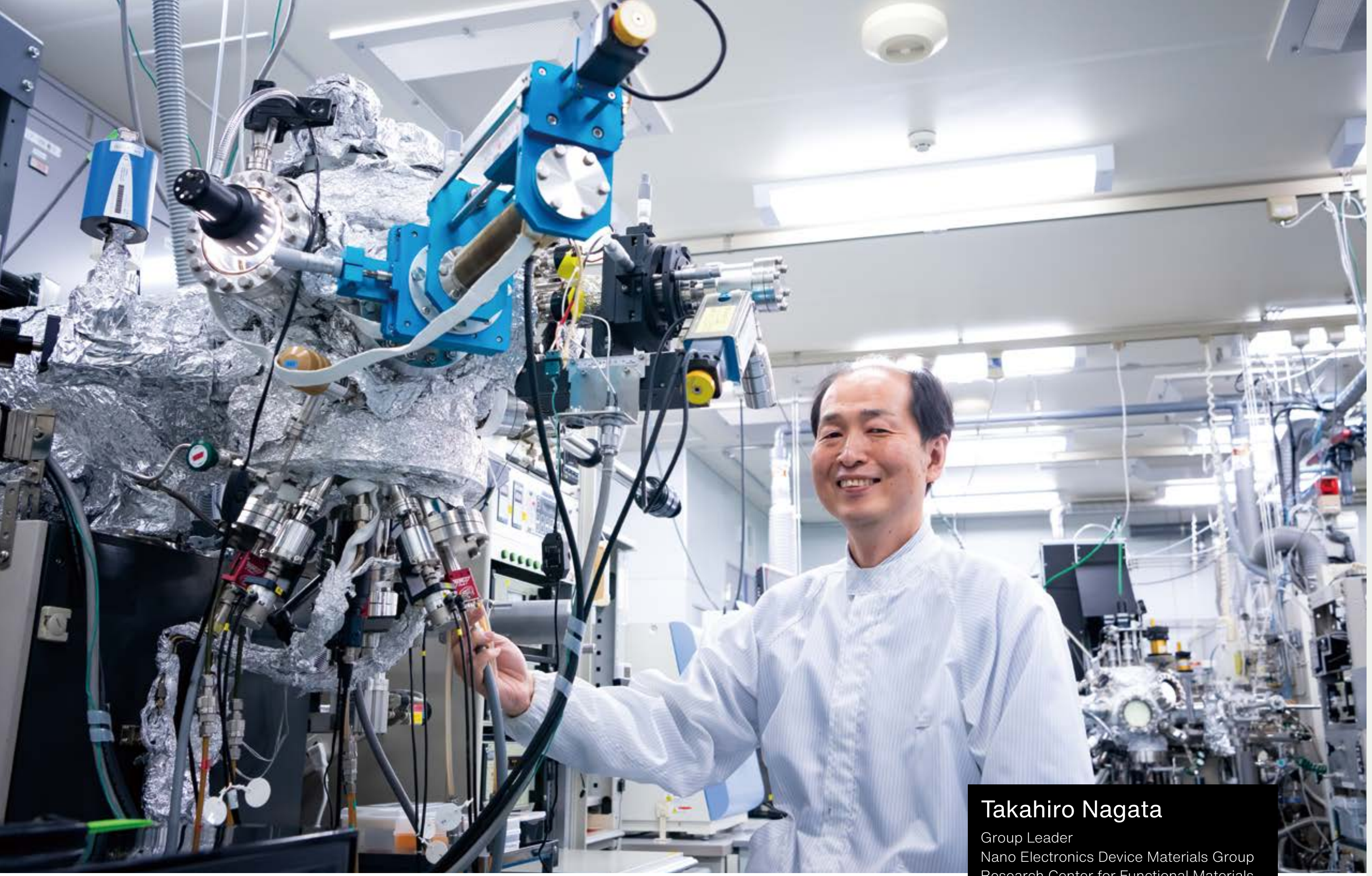


Figure 2. Thin film sample prepared using pulsed laser deposition (PLD)

This thin film is composed of three crystalline chemical elements A, B and C. These elements were vapor-deposited on the substrate surface while the mask covering it was slowly repositioned. The concentration of each element varies between 0% and 100%, enabling all possible compositional combinations to be created on a single substrate (i.e., a thin film sample with a compositional gradient).



Takahiro Nagata
Group Leader
Nano Electronics Device Materials Group
Research Center for Functional Materials

measurement instruments. Moreover, when the RDE system collects measurement data from lab instruments, it also collects associated coordinate information and standardizes data formats. With this feature, the system can produce images of consistent quality in scale and orientation regardless of the instruments from which data originates. Using these images, researchers can easily compare physical properties measured at different coordinates on the substrate surface.

"When we want to identify thin film regions with desirable crystalline and permittivity properties, we can do so by overlaying a crystalline property distribution image and a permittivity distribution image," Nagata said. "Although the use of the RDE system has made this type of data analysis more convenient, the time-consuming actual analysis still needs to be done by researchers. To make this process even more efficient, I plan to introduce AI-based image recognition technology. We're currently developing an AI system capable of automatically graphing the correlative relationship between two types of physical properties and identifying thin film regions with desirable properties."

Making the film deposition process more efficient

Nagata's group is also working to improve the efficiency of film deposition processes using AI. Film deposition has traditionally been done using either pulsed laser deposition (PLD) or sputter deposition as appropriate.

While PLD is unsuitable for forming a thin film over a large substrate, its ability to create high-precision thin films is useful for basic researchers searching for new materials. By contrast, sputtering is unsuitable for precisely controlling chemical compositional changes across a thin film, but is effective in depositing a film over a large surface area, making its use popular in industrial film deposition processes. Leveraging the advantages of both film deposition techniques would improve the efficiency of thin film basic research.

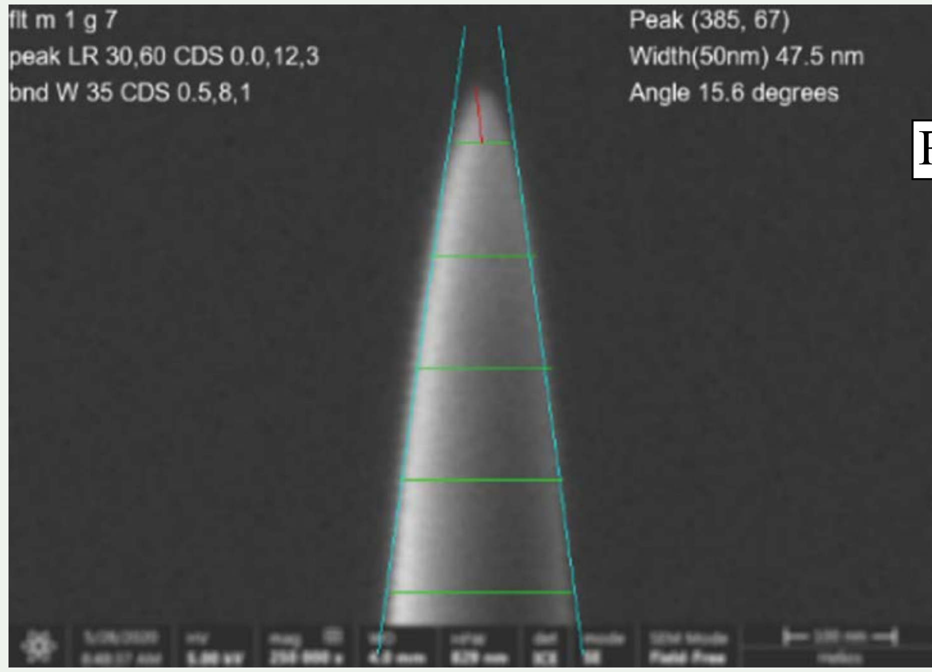
Nagata is currently developing an AI system capable of graphing the relationship between thin films' crystalline structures and their electronic properties. Based on this information, the AI will then identify crystalline structures with desirable physical properties. Thin films to be used for data collection and AI analysis will be fabricated using more precise PLD, while thin films with AI-determined crystalline structures will be fabricated using more efficient sputter deposition. This approach is expected to be applicable to various types of research.

"I hope to expand the range of analyses this AI system can perform," Nagata said. "For example, I want to design the system to be able to graph and analyze the correlative relationships between thin films' crystalline structures and their various other physical properties, including optical properties. These analyses may provide us with some clues to discovering new materials out of a vast number of candidate materials. These clues may also help us design the AI system to automatically suggest the optimum film deposition conditions for thin films, which will be put to practical use in device applications. To improve the accuracy of the AI system, we need to collect various types of data which will be used to train it."

The popularity of AI-based smarter technologies is increasing. Although Nagata developed the new data collection system to facilitate his materials search, he also found other advantages. "The various types of measurement data we store in the RDE system will also be available for use by other researchers," Nagata said. "For example, while I'm mainly interested in the electronic properties of thin film materials, others may be interested in their optical and mechanical properties. It's my hope that our measurement data will help many other materials developers."

(by Kumi Yamada)

Reproducing the skills of experts 3



SEM image of a 3DAP specimen during fabrication in a FIB-SEM system
The program developed by Ohkubo et al. automatically detects the tip shape as defined by its radius of curvature and taper angle (i.e., the angle formed by two imaginary lines drawn along the opposite edges of the tip (blue lines) which extend beyond the tip and intersect). Based on real-time monitoring of a specimen's shape, fabrication continues until the specimen reaches the desired shape.

Automated tip specimen fabrication for 3DAP analysis

A three-dimensional atom probe (3DAP)* is the only means of visualizing three-dimensional distributions of atoms in materials and devices. This analysis method requires the preparation of specimens with needle-like tips from regions of interest such as grain boundaries, interfaces and dislocations. Precise and reproduceable microfabrication is urgently needed to ensure the accuracy and reliability of the analysis. A focused ion beam (FIB)-scanning electron microscopy (SEM) system is commonly used to fabricate these specimens. Target materials or devices are fashioned into the desired needle-like specimens using FIB while the specimen shape is observed by SEM. The process requires very experienced and skilled FIB-SEM operators.

"The 3DAP technique requires very sharp, needle-shaped specimens with tip radius curvatures of less than 50 nanometers," said Tadakatsu Ohkubo, a 3DAP expert. "Highly developed fabrication skills are needed to prepare specimens with the sufficient precision and reproducibility. Experienced and highly skilled FIB-SEM operators therefore play an essential role in the 3DAP field."

Programmable FIB-SEM systems have recently been commercialized globally. Ohkubo's research group immediately began developing an automated 3DAP specimen fabrication method. Based on the experiences and skills of Jun Uzuhashi, an expert in specimen fabrication, Ohkubo and his colleagues developed a program to control FIB-SEM devices, enabling them to prepare 3DAP specimens while monitoring specimen shape in real time and giving active feedback on fabrication parameters.

Automated specimen fabrication was successfully demonstrated in March 2022. Once the fabrication script starts to run, a target material is automatically prepared with the desired, pre-programmed tip shape. Real-time SEM imaging gives active feedback on the fabrication process, thereby ensuring that 3DAP specimens of consistent shape can be prepared. As the result, the reliability and accuracy of 3DAP analysis has been improved, and this automated method is suitable for systematic data collection.

Ohkubo is also working on automated fabrication of thin-foil specimens for transmission electron microscopy (TEM). "We're still in the early stages of developing a prototype," he stated. "However, we've already succeeded in preparing TEM specimens of a quality sufficient for use in leading-edge TEM instruments." The development of automation based on the skills and experience of expert operators accelerates research activities.

* A three-dimensional atom probe (3DAP) is a unique means of visualizing three-dimensional atom maps. Voltage or laser pulses are applied to a needle-like specimen. Atoms at the apex of the tip are ionized in a process called "field evaporation." Ions that evaporate from the tip surface come into contact with a position-sensitive detector with a magnification of over 2 million times. In addition to the position (x, y), the time-of-flight determines the type of atom. By reconstructing the data obtained, three-dimensional maps can be created at atomic scale. This technique is applicable to a wide range of industrial materials and devices, including steel, magnetic materials, superalloys, semiconductors and insulators.

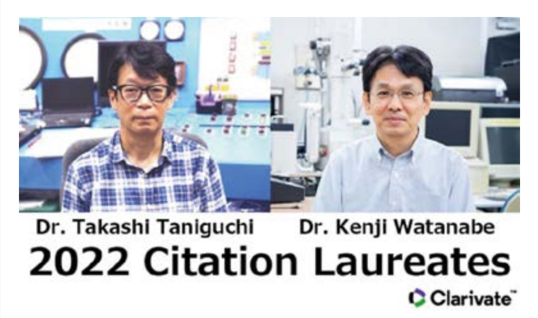
Tadakatsu Ohkubo
Deputy Director
Research Center for Magnetic and Spintronic Materials

Jun Uzuhashi
Senior Engineer
Research Center for Magnetic and Spintronic Materials

NIMS NEWS

1 Two NIMS researchers selected as 2022 Clarivate Citation Laureates

Clarivate Citation Laureates is a list of candidates considered likely to win the Nobel Prize in their respective fields based on the citation impact of their published research. Two NIMS researchers—Takashi Taniguchi (Fellow and Director, International Center for Materials Nanoarchitectonics (left)) and Kenji Watanabe (Chief Researcher, Research Center for Functional Materials (right))—were awarded this honor in 2022 for their fabrication of high-quality hexagonal boron nitride crystals, the availability of which enabled a revolution in research on the electronic behavior of two-dimensional materials.



* Their research will be featured in the next NIMS NOW issue.

2 NIMS researchers were selected as "Highly Cited Researchers 2022"

Five researchers of NIMS and three related researchers were selected as "Highly Cited Researchers in 2022" by Clarivate Analytics Inc.

Highly Cited Researchers have demonstrated significant and broad influence reflected in their publication of multiple highly cited papers. These highly cited papers rank in the top 1% by citations for a field or fields.

■ Highly Cited Researchers (NIMS)

Katsuhiko Ariga, Jonathan P. Hill, Takashi Taniguch, Kenji Watanabe, Jinhua Ye

■ Highly Cited Researchers (NIMS-related researchers)

Dmitri Golberg, Shinichi Komaba, Yusuke Yamauchi

Hello, my name is Akhilesh and I am from India. I came to Japan in August-2019. I was a JSPS postdoctoral researcher at Shizuoka University before joining NIMS as an ICYS Researcher in September-2021. Realizing the significance of biosensors and their influence on society piqued my interest in the biosensing sector, which has led me to this point. In 2020, when the COVID-19 outbreak occurred, it became a catalyst in the development of biosensors suitable for on-site and off-grid settings. It was with this mind, I joined the ICYS program at NIMS for developing heterostructured nanomaterials and their subsequent utilization for engineering the detection methodologies for virus sensing. We are trying to develop a novel self-powered photo-electrochemical virus sensing device.

NIMS is an international research center for nanomaterials with state-of-the-art advanced research equipment and a pioneer in the materials research field. The research atmosphere at NIMS is exciting, with opportunities to discuss ideas, share experiences, and be inspired by many excellent researchers. Japan is an awesome country and keeps surprising us with its culture, friendly people, and amazing sightseeing while providing a safe atmosphere for our family.

At Takayama in Gifu prefecture with my family.

Akhilesh Babu Ganganboina
(India)
ICYS Research Fellow

Research introduction of Dr. Ganganboina (NIMS NOW p.10-11)

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on the cover: X-ray diffractometer (see p.7)

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