MESSAGE FROM THE DIRECTOR

Steadily advancing research and development in the midst of the coronavirus pandemic

The Advanced Institute for Materials Research (AIMR) in October 2022 marks 15 years since its founding. On this occasion, I would like to offer my heartfelt thanks to all of you who have made this possible through your support.

AIMR was launched in 2007 as one of the centers established by the World Premier International Research Center Initiative (WPI), a Japanese government project. Since that time, it has been continually engaging in new system creation and research and development activities on the way to becoming a global center for materials science. In this process, we have sought to carry out the WPI’s four basic objectives, namely, advancing leading-edge research, creating interdisciplinary domains, establishing international research environments, and reforming research organizations. In 2012, AIMR began collaboration between mathematics and materials science, seeking to formulate new scientific principles that will enable the development of materials through prediction. It further develops devices and systems for putting to use in society the developed leading-edge materials, seeing as its mission helping to solve problems faced by humankind in such areas as resources and the environment.

In 2017, AIMR was recognized as a member of the WPI Academy, established to accelerate and expand global brain circulation. We intend to continue carrying out research at the top level worldwide, as a hub for global brain circulation.

A major feature of AIMR, the global center for materials science, is the above-noted collaboration between mathematics and materials science. By using the universal language of mathematics to describe materials science, which covers an extremely broad range, we aim to discover new phenomena and phenomena that are not yet known, and to pursue new topics to produce novel research results.

Dealing with the spread of the COVID-19 pandemic from March 2020 has forced us to place certain limits on how research activities are carried out. Yet even as the pandemic has become protracted, AIMR, while taking every measure to control the spread of infections, has been steadily developing new principles and creating new materials. This work is proceeding mainly in three advanced target projects putting into practice mathematics-materials science collaboration, namely, 1) Local Structure Control in Topological Functional Materials; 2) Integrated Control of Bond Variation and its time evolution; and 3) Improvement of Self-Organization Technology and Control of Biological Response.

To give further momentum to these projects, along with awareness of the AIMR’s strength of bringing researchers from different disciplines under one roof, I would like to call attention to the three Rs I have proposed as a way of maximizing this strength. The first R, Relief, refers to our guiding principle of not only giving priority to the safety and welfare of everyone involved in AIMR, but also giving importance to support of their mental wellbeing. The second R, Reform, means the mental preparedness for continuing to run at the forefront of research, aware that in being at the leading edge of our research field we are at the same time at the “edge” in the sense of precipice. The third R, Recognition, means actively publicizing research results, so that the necessity of AIMR will be recognized more than ever in Japan and abroad.

With the help of all those who support AIMR, as we overcome the difficulties posed by the ongoing pandemic, I would like us to give substance to our mission of “bringing advanced materials science to the world.” Along with becoming a core institute for building the global research environment of Tohoku University, we would like AIMR to contribute to the world’s advanced materials science and to society.

Shin-ichi Orimo, Director
Advanced Institute for Materials Research, Tohoku University

INFORMATION

WPI Advanced Institute for Materials Research

Established in 2007 as part of the Japanese government’s World Premier International Research Center Initiative (WPI), the Advanced Institute for Materials Research (AIMR) was tasked with pursuing world-class research and promoting global brain circulation. The AIMR has since been bringing together world-class researchers from Japan and abroad to carry out cutting-edge research in materials science through interdisciplinary collaboration among its four materials-related groups — Materials Physical, Non-equilibrium Materials, Soft Materials, Device/System — and the Mathematical Science Group.

In 2017, the AIMR became a member of the WPI Academy, which consists of WPI centers that have achieved world-premier status. The institute will continue to maintain its world-class research environment and further promote global brain circulation. Led by distinguished material scientist and director Shin-ichi Orimo, the institute promotes interdisciplinary research across the different groups. It also fosters young researchers through the Global Intellectual Insulation and Integration Laboratory (GIIL). This unique program, which is currently supported by the WPI Academy, promotes international joint research conducted in close cooperation with high-profile researchers invited from countries around the world.
The electronic states of 3D graphene are unaffected even by high curvatures.

Bending graphene on a nanoscale maintains some of the excellent electrical properties of flat graphene while giving others a boost, an AMR-led team has shown. By providing engineers with another parameter to work with—namely the radius of curvature—this discovery could lead to novel devices based on three-dimensional (3D) graphene.

Graphene, a one-atom-thick honeycomb array of carbon atoms, exhibits many desirable properties (e.g., high electrical and thermal conductivities and high electron mobility), making it attractive for a wide range of applications. However, graphene’s sheet-like structure is a problem because large areas are needed for devices, challenging compact device fabrication.

One approach to this problem is to bend graphene into 3D nanostructures, packing large areas of graphene into tiny volumes. However, packing often deteriorates the properties of graphene by introducing defects and by disrupting the crystalline structure. Until now, what role, if any, the radius of curvature plays in this deterioration remains unclear.

Here, a team led by Mingwei Chen from AMR fabricates porous graphene sponges with pore sizes ranging from 35 to 1,000 nanometers, and systematically investigates how the properties of graphene are affected by the radius of curvature.

The researchers discovered that curvature does not alter the electronic states of graphene. Importantly, it preserves the Dirac fermion nature of electrons from which many of graphene’s properties spring. “We showed that graphene with a radius of curvature between 50 and 1,000 nanometers maintained its Dirac fermion character,” says Yoichi Tanabe, the first author of the study.

Furthermore, the team has found that the curvature can be used to tune the electrical transport properties of graphene. “We expect that the properties of graphene can be amplified by nearly 1,000 times in our 3D graphene sponge,” says Tanabe.

Future directions will explore what effects varying the curvature-induced pseudo-magnetic field has on graphene. “We found that the pseudo-magnetic field acts as a control parameter for curved surfaces of graphene,” says Tanabe. “So, next we will try controlling the physical properties unique to 3D curved surfaces by tuning this field.”

Resolving atomic structure into polyhedra

The arrangement of atoms near the interface between two crystals can be represented in terms of polyhedra.

The atomic structures of regions near grain boundaries in polycrystalline materials can be characterized by certain types of polyhedra, as AIMR researchers have shown. This promise to make it easier to model these materials and to predict their properties.

Most metals and ceramics are three-dimensional (3D) messes of microscopic crystals of different sizes and orientations. The properties of such polycrystalline materials are strongly affected by how the atoms are arranged near the grain boundaries where two crystals meet. However, the breakdown of the regular structural order near these grain boundaries makes the structure-property relationship challenging to investigate.

For example, a crystal with a face-centered cubic (fcc) structure (e.g., aluminum, copper, and gold) can be considered to be made up of adjacent four-sided (tetrahedra) or eight-sided (octahedra) shapes.

Now, Kazutoshi Inoue of AIMR at Tohoku University and his co-workers have shown that it is also possible to analyze regions near grain boundaries in terms of atomic polyhedra (see figure). They did this by analyzing the 3D polyhedral structure of fcc crystals for certain types of grain boundaries.

"We discovered that regions close to grain boundaries can be packed only by the bulk polyhedral units (i.e., tetrahedra and octahedra) or grain-boundary-type polyhedral units that differ from the bulk ones," says Inoue. "This extends the current two-dimensional model framework to define 3D polyhedral units, covering all the grain boundaries in cubic crystals.

"The researchers also uncovered an unexpected connection between the grain-boundary atomic structures and the properties of fractions that can be represented by mathematical figures known as Farey diagrams.

"We found, a one-to-one correspondence between the 3D atomic structures of grain boundaries and the distribution of rational numbers," says Inoue. "It’s surprising that the grain-boundary hierarchy can be accurately described by a modified version of the Farey diagram, which also represents the mathematical structures of other physical and biological phenomena, including oscillations and phyllotaxy. This connection will enable researchers to readily estimate the 3D arrangement of grain boundaries.

The team intends to use this method in conjunction with microscopic observations to determine the conditions for realizing stable atomic structures at grain boundaries.


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Helical optics creates spin at interfaces

A new way to generate electron spin using light could lead to ultrafast memory devices with low power consumption.

Helical light beams can induce electron spins at the interface of a magnetic and non-magnetic material, three AIMR researchers have discovered. This effect could form the basis of light-driven magnetic memory devices that are much faster and have far lower power consumption than present memory devices.

Researchers are working on developing a new breed of devices that will combine the advantages of light and electricity. Such devices will realize rapid, low-energy information processing by using photoelectric integrated circuits, where light is used to perform many of the functions currently realized electrically.

"Photoelectric integrated circuits are being developed for future fast and energy-efficient information processing by using light, because light is a low loss and extremely fast," says Akira Ishibashi of AIMR at Tohoku University. "Many researchers are trying to implement optics in electronic devices.

Ideally, data would be stored in such devices by using light to switch the direction of magnetisation in nanoscale magnets, but this has yet to be realized due to the weak interaction between light and magnetic materials.

"Here, the researchers use helical laser beams to manipulate the magnetisation in metallic ferromagnets—a technique based on the optical Rashba–Edelstein effect that has never been used on the interface between a ferromagnetic film and a non-magnetic metal film (see figure).

"This way, the team has found a new method for generating spins optically at the interface of the two materials. The optical Rashba–Edelstein effect has not been demonstrated experimentally so far.

"We tried to observe the optical Rashba–Edelstein effect experimentally using the interface between a ferromagnet and a heavy metal, and we discovered that there exists a coupling between optical helicity and electron spins even at a nanoscale interface," says Ishibashi. "This is the first time this has been demonstrated, and it came as a surprise to us."

"This study could lead to energy-efficient photoelectric–magnetic memory devices, and it could also open up a new research field that combines nanophotonics and spintronics," says Ishibashi. "As a next step toward realizing photoelectric–magnetic memory devices, we are planning to try to further increase the nanoscale photon–spin coupling.


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**MATERIALS CHEMISTRY**

**Chemical dealloying synthesizes new porous-carbon anode**

A process that can fabricate a bimodal porous carbon and tune each modal pore size independently

Researchers at AIMR have developed a sequential chemical dealloying technique for synthesizing 3D bimodal porous amorphous carbon with well-defined meso- and micro-pores. With pore sizes and architecture tunable at the atomic scale, the amorphous-carbon product of this new, scalable process shows excellent performance as a sodium-ion battery anode.

The search for the next sodium-ion battery hard-carbon anode is a quintessential transport problem. While numerous studies have shown that graphic features such as surfaces, interlayers, and micro pores can ameliorate sodium diffusion and storage, the sheer variety of hard-carbon defects makes the mechanism difficult to elucidate.

That is, unless the improvement strategy targets a higher-performance hard-carbon, whose 3D porous features are directly tunable by the novel fabrication method from the start.

"The main obstacle to exploiting dealloying for porous carbon synthesis is the strong chemical bonds and the high chemical stability of most carbide precursors," says Jinhai Han, the first author of the study. "We overcame this obstacle by using a special Ni₃C precursor that is in a metastable phase at room temperature."

Using a two-stage dealloying process, the AIMR team produced bimodal-porous amorphous carbon. The first stage removes Mn from a Ni₃Mn alloy, replacing it with carbon through carburization. The second stage removes Ni from the resulting metastable NiC alloy, yielding an amorphous carbon with coexisting meso- and micro-pores.

One key advantage of this method is that individual steps of the process control specific pore sizes; both at the micro- and at the meso-scale.

The team tested the performance of the new bimodal porous carbon as a sodium-ion battery anode. "They found that not only does the product have rapid diffusivities, excellent rate capacities, and long cycling stability compared with known hard carbons, but its transport properties are affected by specific features that are process-controlled," says Han.

Future plans will utilize different metal-carbide precursors to expand the tailoring capabilities of this process. "Our DFT calculations of bond strength predict a broad range of metal carbides as potential precursors for preparing porous carbon by dealloying," says Han. "We will explore these metal carbides in the future."

**MAGNETIC MATERIALS**

**Hydrodynamics explore a soft material’s magnetic handle**

Tracing the nanoscale origins using mesoscale observations of a magnetic liquid

An AIMR-led team has used hydrodynamics experiments to investigate the magnetization of ferromagnetic liquid droplets. The results show that the droplet remanent magnetization stems from the magnetostatic short-range order (MSRO) between jammed magnetic nanoparticles, opening a new way to tailor the extraordinary material.

Ferromagnetic liquid droplets possess both solid-state magnetic properties and fluid-like shape-morphing capabilities thanks to the jamming of paramagnetic nanoparticle surfactants at liquid-liquid interfaces. This mechanism lies in the nanoparticles MSRO, generating a remanent magnetization that makes bio-compatible liquid robots a real possibility.

However, bio-compatible capabilities such as targeted drug delivery or functional microfluidic channels entail the controlled shape changes and interactions with the environment at exquisite scales. Currently, few is known about the MSRO origin of the droplet magnetization for such control.

"Magnetic short-range orders are difficult to measure directly," says Xuefei Wu from Shanghai University of Chemical Technology. "They require magnetic imaging or scattering experiments that only provide local information."

Instead, the team’s approach to studying MSRO is to design hydrodynamics experiments that can directly measure the angular accelerations and velocities induced to spherical droplets by external magnetic fields.

"Our experiments are designed to inform us on how much magnetic torque the droplets experience," says Robert Strzelecki of the University of Nebraska Lincoln. "Choosing spherical paramagnetic nanoparticles also enables the desired reversible transformation between paramagnetic and ferromagnetic liquids."

With this approach, the team has provided some intriguing evidence for MSRO, such as the pH of the droplet’s aqueous phase controlling the amount of torque applied to the droplet, or the jammed nanoparticle enhanced magnetization by coupling with dispersed nanoparticles via magnetic interactions—the same coupling that enables the additional assembly of non-magnetic nanoparticles while retaining substantial magnetization.

"The surprising thing about the remanent magnetization is that it emerges with the formation of the ferromagnetic liquid droplets," says Thomas Russell from AIMR. "This is the reason why droplet magnetization only requires low concentrations of magnetic nanoparticles and is preserved through droplet deformation."

Future directions will combine numerical modeling with hydrodynamics experiments to determine how the patterning of the interfacial layer can tailor the droplet magnetization and functionalization.


When microscopy and theory stretch beyond two dimensions

A synergistic approach that can elucidate complex interfacial structures

A collaboration between AIMG, the University of Tokyo, the University of Cambridge, and the University of Yokohama has demonstrated a new approach to investigating grain-boundary structures. Combining scanning transmission electron microscopy (STEM) with ab initio random structure searching (AIRSS), the team has isolated a grain boundary with strong anastase character embedded in bulk rutile TiO₂, taking one more step toward bridging the structure-property gap in ceramic research.

Grain boundaries are ideal platforms for tailoring ceramic materials such as TiO₂. Ubiquitous in all but single-crystal solids, these interfacial structures are believed to be the sites of many processes such as dopant segregation, charge recombination, and domain migration—all potential solutions to engineering desired macroscopic properties of bulk materials. However, while the traditional approach involving high-resolution imaging followed by computational optimization can elucidate simple grain-boundary structures, it cannot address more complex interfacial structures—where interesting physics happens—due to the 2D image limitations.

Here, the team led by Yuichi Ishihara and Chris Pickard from AIMG, and Keith McKenna from the University of Yokohama takes a synergistic approach to broaden the scope of the grain-boundary structure investigation. Starting with imaging the grain boundary of a rutile TiO₂ bicrystalline with high-resolution STEM, the team then uses AIRSS to explore the chemical environment adjacent to the imaged 2D structure.

"Our initial local energy minimisation methods failed to find a structure consistent with experimental STEM and electron energy-loss spectroscopy (EELS) observations," says McKenna. "Hence, we applied AIRSS, a global optimization method, to uncover the structure of the more complex TiO₂ grain-boundary structure."

This way, the team has predicted the local chemical environment of the imaged grain-boundary structure, calculated its EELS signals, and confirmed these signals experimentally. The results demonstrate that the grain boundary of the imaged bicrystal strongly resembles bulk anastase TiO₂—an impossible feat using the traditional modeling approach.

"Our study shows that the structures of complex interface systems are now within reach," says Georg Schmertmann from the University of Cambridge. "We will use our method not only to elucidate these structures, but also to calculate their electronic, thermal, and mechanical properties for future interface designs."

CALCIUM BATTERIES

A step toward a high-energy density battery using an abundant, non-toxic alkali-earth metal

A designer electrolyte from concept to testing

A collaboration between AIMG, the Institute for Materials Research at Tohoku University, and the Laboratory of Materials for Renewable Energy at Ecole Polytechnique Fédérale de Lausanne has designed, synthesized, and tested a new fluoro-free calcium monocarbonate (Ca₂(CO₃)₂) electrolyte. With improved conductivity and electrochemical stability, the new electrolyte is a significant step toward more reliable rechargeable calcium batteries.

Calcium batteries are prime candidates for replacing their lithium-ion counterparts because of calcium's abundance, non-toxicity, lower reduction potential compared to Mg or Al, and higher (Ca²⁺) ionic charge capacity compared to that of Li²⁺.

However, the lack of a suitable electrolyte hinders the realization of calcium batteries with the above advantages. Currently, cutting-edge model calcium batteries use fluoro-containing, weakly-coordinating electrolytes (e.g., Ca(BF₄)₂ or Ca(B(Ph)₄)₂) (BF₄⁻ hexafluorophosphate) in organic solvents to maximize Ca²⁺ conductivity at the expense of electromechanical stability. Moreover, the presence of fluorine also results in the formation of a passivating CaF₂ film that prevents the reversible calcium plating/stripping at the anode surface.

To address these challenges, the team led by Kazuaki Kiryu (currently affiliated to the Institute for Materials Research) and Shin-ichi Orimo from AIMG has designed a new electrolyte using monocarbonate counterions (tiosalenidrons in Figure). Like the Ca(BF₄)₂ example, the new counterion is a weakly-coordinating anion that maximizes Ca²⁺ conductivity, but unlike Ca(BF₄)₂, it is free from CaF₂ film formation.

"The monocarbonate cluster is a stable, weakly-coordinating counterion, but it is not soluble in many solvents," says Kiryu. "We overcame this problem by using a mixture of desired solvents at specific ratios in this way, the team has synthesized the new electrolyte using a known scalable route. Preliminary tests of the electrolyte performance show promising results including high conductivity, wide electrochemical window, and reversible calcium plating/stripping without CaF₂ film formation."

"The test results from the new monocarbonate electrolyte open a new path for other nonfluorine rechargeable battery systems," says Orimo. "We are currently exploring the inclusion of other metals such as magnesium and aluminum."

Redesigning a fabrication method around a better template

How to make a proof-of-concept experiment scalable

An AIMR-led team has developed a scalable template-assisted method for fabricating edge-free, single-layer graphene mesospore (GMS). Combining experiments with theory, the team has redesigned a known method using magnesium oxide (MgO) as template material instead of alumina (Al₂O₃). The use of MgO enables scalability and produces GMS with superior transport and mechanical properties compared with Al₂O₃.

Proof-of-concept experiments often need major redesign before they are scalable. One such example is the recent template-assisted fabrication of an edge-free, single-layer GMS that can outperform carbon nanotubes as supercapacitor electrode materials. While this method produces a robust GMS, the removal of its Al₂O₃ nanoparticle template requires the drastic use of hydrofluoric acid that precludes its commercial application.

Here, scalability entails redesign of the fabrication method around a different template—one that can both form the desired GMS and be removed in a cost-effective way. The team selects MgO nanoparticles. MgO is a commonly used template material in commercial porous-carbon production samples are easily removed by dilute acid solutions,” says Hirotomo Nishihara, the principal investigator from AIMR. “The trick is to make sure it can form GMS with the right features.”

To this end, the team has used a combined experimental/theoretical approach to elucidate the GMS-formation mechanism and to establish the optimal conditions conducive to desired features such as single-layer graphene walls.

In so doing, the team has determined not only that the graphene-sheet formation likely occurs at oxygen vacancy sites on stepped MgO(110) facets, but also that the formation of the first graphene layer is much faster than that of subsequently stacked layers. This latter finding enables fine control of the reaction time for obtaining single-layer GMS.

Direct property comparisons indicate that the GMS produced using MgO is not only a good supercapacitor electrode material, but it is also more mechanically flexible than the GMS produced using Al₂O₃.

“We are investigating the use of other metal oxides and their effects as template materials beyond MgO and Al₂O₃,” says Nishihara. “Ultimately, we are aiming for the development and the mass production of precisely controlled graphene nanomaterials.”


Representation of the graphene mesospore synthesis via catalytic methane decomposition on magnesium oxide.

Making a link between graphene geometry and properties

A mathematical approach that connects experimental and theoretical observations

A research team led by AIMR has developed a mathematical model called standard realization with repulsive interaction (SRRI) that uses the geometric information of graphene to simulate its defect-induced curvatures and properties. Experimental comparisons with the simulated properties suggest the possible use of SRRI as a complementary pre-screening tool for both density functional theory (DFT) and experimental measurements.

Scientists are still trying to understand how geometric changes in the graphene honeycomb structure can produce exotic properties.

For example, the introduction of topological defects by chemical treatments of graphene is known to both alter graphene geometry and enhance graphene catalytic properties. However, how these two observations are related to each other remains unclear.

Here, the team develops SRRI—a mathematical model that takes into account the attractive and repulsive interactions between graphene defects and their carbon-atom neighbors—to both simplify the model of interest and predict the curvatures and catalytic properties induced by graphene topological defects.

“Our approach refines an existing mathematical model to describe the geometric changes in systems such as graphene and investigate their properties,” says Yoshikazu Zuo (from the University of Tokyo). “The resulting SRRI gives insight into the link between the geometry of the material and its properties.”

Direct comparisons indicate that while SRRI predicts graphene defect-induced properties in qualitative agreement with DFT, it does so a billion times faster.

To demonstrate the significance of these results, the team synthesizes graphene samples with similar defects and uses scanning electrochemical microscopy to find a link between curvature and catalytic properties similar to those predicted by SRRI.

“Our results show a fast mathematical modeling method that not only can pre-screen model systems for in-depth DFT studies, but also could be used to guide experimental measurements in real time,” says Motoke Kotani from AIMR.

Future projects will use SRRI to investigate the structure-property links of other carbon networks such as the Mackay crystal.


Representation of a graphene section with curvature induced by topological defects.
Deconvolving the many-body problem with numbers

First-time demonstration of a Bayesian approach to surface spectroscopic data analysis

A team led by AIMR has developed a new technique for interpreting surface electronic state characteristics by modeling angle-resolved photoemission spectroscopy (ARPES) data using a Bayesian approach. The team demonstrates this technique by confirming the size and by elucidating the microscopic origins of the Ti8(S,Se)5 Dirac gap.

In condensed matter physics, the many-body problem often complicates experimental measurements.

For example, the surface electronic states mapped by techniques such as ARPES are a convolution of the bare-band dispersions (E, single-electron dispersions) with the electron self-energies (E, multiple-electron correlations). This convolution makes graphene band gaps and dispersion kinks of high-temperature superconductors difficult to explain by ARPES measurements alone.

That is, unless one of the two components can be statistically extracted using known quantities.

Here, the team demonstrates an approach to investigating surface states by applying Bayesian modeling to the ARPES data acquired on a Ti8(S,Se)5 surface—a material whose Dirac cone characteristics have been controversial due to the many-body problem.

The Bayesian approach is very good at inferring the probability of any event, provided that quantities relevant to the event are either known, or can be modeled from real data,” says Takafumi Sato, the principal investigator.

To this end, the team formulates a semiparametric Bayesian analysis expressing all relevant spectral quantities (Q), which include E and Δ, to model an ARPES map of Ti8(S,Se)5. The modeling provides estimates for all Q, enabling the team to ask physical questions such as whether the Ti8(S,Se)5 Dirac cones are gapped.

Using Bayes’ theorem (see Figure), the team calculated that the probability P(A|Q,Qs) of having a gap of size Δ = 0 is negligible, concluding that the bare-band Ti8(S,Se)5 Dirac cones must be gapped.

“With this technique, we were not only able to calculate the Dirac gap size with high precision, but we were also able to narrow down the microscopic origins of the gap,” says Sato. “This would not have been possible without the ability to deconvolve the many-body problem.”

Future directions will apply this technique to other quantum materials to demonstrate its applicability.

HETEROGENEOUS CATALYSIS

Redirecting surface atom migration to the (100) facets of a ceria nanocatalyst

A new dissolution/re-precipitation method for recycling facet-controlled nanocatalyst

An AIMR-led research team has combined the use of an organic modifier with supercritical hydrothermal treatment techniques to design a strategy for regenerating faceted nanocatalysts. The team demonstrates this strategy by regenerating degraded cubic ceria (CeO2) nanoparticles back to their original cubic shape.

All heterogeneous catalysts degrade over time: poisoned by chemicals, fouled by carbon deposits, eroded by heat and pressure, or eroded by wear and tear. For this reason, the design of new catalysts often includes catalyst regeneration, aiming for regeneration methods that are clean, easy, and cheap.

However, while conventional regeneration approaches can ameliorate the above degradation mechanisms, they cannot address a mechanism particular to nanocatalysts: degradation through changes in surface facets.

Here, the AIMR research team designs a regeneration approach aimed at reorienting active surface facets through partial dissolution/re-precipitation with a facet-selective organic modifier under supercritical hydrothermal conditions.

CeO2 nanoparticles are unique in that their shapes, exposed facets, and site distributions are controlled using carboxylic acids,” says Takahiro Tomai, first author of the research. “To reorient the (100) facet of the nanocatalyst, we use the decahydroquinoline surface modifier to control the exposed facet, and the formation of a Ce organometallic complex to promote dissolution in supercritical water.”

Monitoring the particle size, shape, and catalytic activity by transmission electron microscopy (TEM) and by oxygen storage capacity measurements, the team uses non-cubic CeO2 nanoparticles to determine the optimum conditions for cubic nanoparticle regeneration. The team then demonstrates the approach by degrading pristine cubic CeO2 nanoparticles in air and restoring the cubic shape to these same particles.

“Because our design involves complex particle-to-facet interactions through partial dissolution and re-precipitation, some of us were sure that it was going to work,” says Tomai. “We were very excited to see the cubic shapes on the first TEM images of the regenerated nanoparticles.”

Future directions will develop this simple nanocatalyst recycling approach to target several of the United Nations’ 17 Sustainable Development Goals, including clean water and sanitation, affordable and clean energy, responsible consumption and production, and conservation of life below water.


2. https://dx.doi.org/10.1002/chem.202103731
Isolation of a robust room-temperature two-dimensional Mott insulator

Using low dimensionality to enhance Mott-transition temperature

An AIMR-led team has isolated a room-temperature (RT), two-dimensional (2D) Mott-insulator phase of a single-layer 1T-TaSe₂. Using angle-resolved photoemission spectroscopy (ARPES), the team has determined that the enhancement of the Mott-transition temperature stems from the interplay between excitonic quantum features and low dimensionality. These results open a pathway toward the realization of RT Mottronic devices and high-temperature superconductors.

Superconductivity occurs in complex systems such as doped copper oxides (at T \approx 200 K) and layered graphene bilayers (T \approx 1 K). According to the Hubbard model, these superconductivity examples are associated with the Mott insulator—a phase that emerges when the electron correlation (U) of a material significantly exceeds the width of its partially filled band (W).

The Hubbard model also implies that the Mott transition of a material is triggered when physical conditions (e.g., temperature, pressure, and doping) maximize the material’s effective Coulomb interaction (U/W), suggesting a strategy toward RT Mott-transition: maximizing U/W using low dimensionality.

Here, the team demonstrates this strategy using 2D transition-metal dichalcogenides (TMDs) such as the single-layer 1T-TaSe₂. "Because the bulk 3D 1T-TaSe₂ undergoes Mott transition at low T \approx 200 K, reducing its dimensionality to 2D should raise its U/W value, enhancing the Mott-transition temperature," says Takafumi Sato, the principal investigator. "The key lies in designing a set of experiments that can demonstrate this possibility."

To this end, the team combines ARPES and other spectroscopic techniques to monitor U/W-defining electronic features. While the simultaneous observation of the Mott gap and the charge-density wave (CDW) gap at T \approx 40 K suggests that the 2D 1T-TaSe₂ is a Mott insulator, the observation of the lower Hubbard band up to T \approx 450 K indicates the Mott-insulator phase is robust at RT and beyond.

"We have discovered a unique, 2D RT Mott insulator phase," says Sato. "This is only possible thanks to an exquisite interplay where U is enhanced by excitonic features such as CDW, and W is reduced by 2D confinement."

Future work on 2D TMDs will aim at demonstrating the Mott-insulator to metallic phase transitions at RT and at detecting high-temperature superconductivity.