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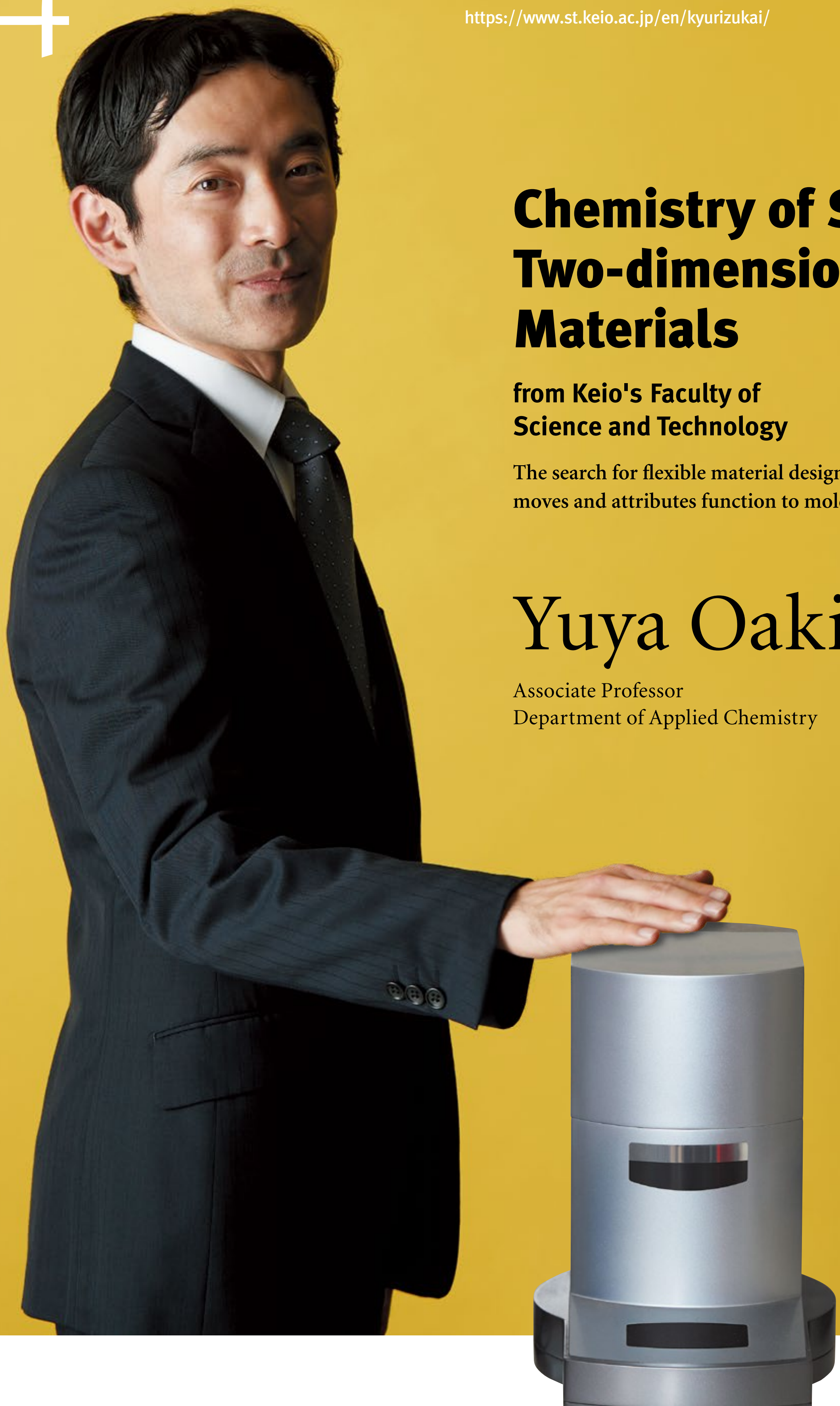


Chemistry of Soft Two-dimensional Materials

from Keio's Faculty of Science and Technology

The search for flexible material design that moves and attributes function to molecules

Yuya Oaki

Associate Professor
Department of Applied Chemistry

This issue introduces Associate Professor Yuya Oaki, a scientist who enjoys chemically designing materials and finding useful functions for them in the world.

The pursuit for functional and soft 2D material structures

Having curiosity making new things and new ways to use them.

The term "applied chemistry" refers to the science of manipulating atoms and molecules to create substances that are useful in everyday life. There are endless possibilities for potential creations and their functions.
Associate Professor Oaki of the Department of Applied Chemistry focuses specifically on and finds joy in crafting "soft two-dimensional materials."
He says that they hold the possibility for "unexpected discoveries."

The blackened sample bottle "eureka" moment

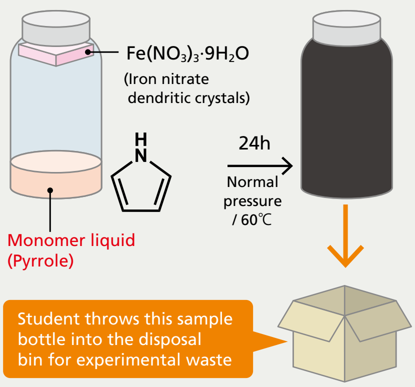
Throughout the research world, unexpected discoveries are often described as "serendipitous." Chemistry is no exception, with results from failed experiments sometimes leading to unexpected findings and useful data. Oaki of the Department of Applied Chemistry showed us a blackened sample bottle (Fig. 1, left) he says led to an "unexpected discovery" 10 years prior, explaining "I have been doing research for about 20 years, and have had an experience that I could call serendipitous on two occasions."

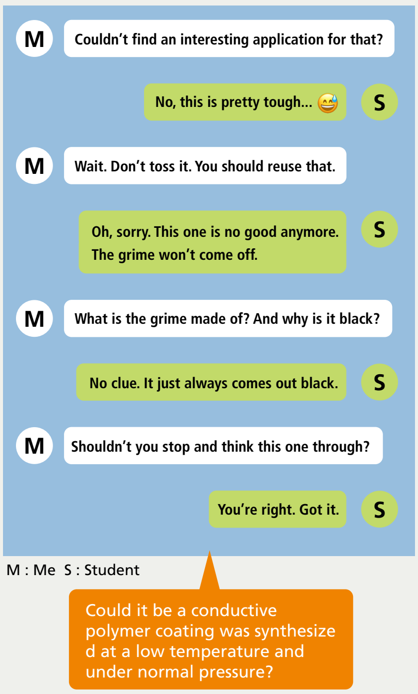
In university laboratories, students conduct research under the guidance of faculty members such as professors and associate professors. The faculty and students discuss and decide the direction and method of research together, but it is the students who carry out the experiments. Oaki likes to communicate with (or something like that...) how his students conduct their experiments and the results of their collected samples and then engage students in discussion. On one particular occasion, his attention was drawn by a student who was conducting an experiment to coat the surface of a dendritic crystal of iron nitrate with a conductive polymer in order to apply it to an electrical circuit. The student, thinking he had failed in his attempt, proceeded to dump the blackened sample bottle into the disposal bin for experimental waste.

"I instructed the student to reuse the bottle, given that normal grime can be removed from glass when washed. The student replied that it wouldn't come off," Oaki recalls. "I knew intuitively that something interesting was afoot as a coating on a glass bottle so black, even, and strong, should not have been produced by the planned experiment."

Following his instincts, he examined the black substance. The polypyrrole film they expected to form on the crystal surface of the iron nitrate had also adhered to the entire inner wall of the bottle. "Polypyrrole, which is a series of pyrroles (see the figure), is a 'conjugated polymer' in which double bonds and single bonds are alternately linked. They are incapable of mixing with solvent due to their rigid chemical structure. In other words, it should not be possible for them to form a paint-like coating. This means that while many conjugated polymers are highly functional in terms of conductivity, heat resistance, and redox activity, applying the substance can prove quite a challenge.

However, this failed experiment showed that conjugated polymers could easily be coated on various substrates and substrate surfaces," Oaki explained, emphasizing that the black sample bottle was a discovery, not a failure.





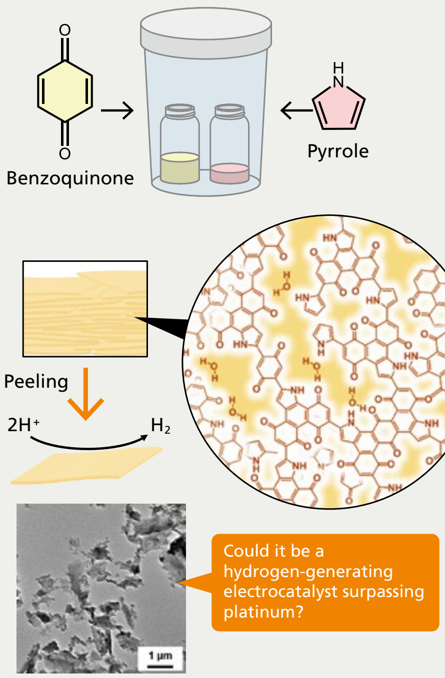
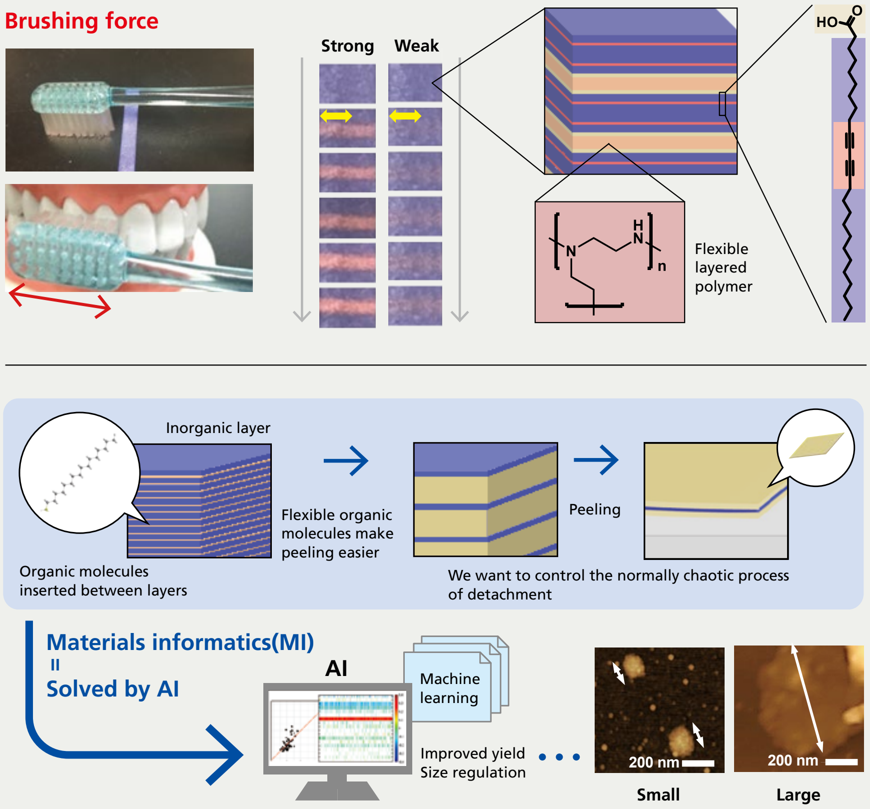


Fig.1 Two serendipities
Left side, middle: Reproduction of interaction with student at the time of the first "unexpected discovery," which is used when giving lectures on the topic. The student attached iron nitrate crystals to the lid of the sample bottle, added the monomer liquid which serves as the base of the coating, and closed the lid. It was thought that when this was allowed to stand at 60°C, the monomer liquid would evaporate and a polypyrrole coating would form solely on the surface of the iron nitrate crystals. In reality, the monomer vapor reacted with the iron nitrate, filling the bottle and creating a solid coating. Right: The second "unexpected discovery." A network structure is formed from a reaction between pyrrole with benzoquinone (middle). This could be easily delaminated (photo), and the produced nanosheets had the property of acting as a hydrogen-generating electrocatalyst.

Fig.2 Example of soft 2D material for which research and development is in progress

Above: Sensor material using layered polydiacetylenes that creates a visual of brushing force.
Bottom: Material development using "materials informatics (MI)" into his research since 2016 (Fig. 2, bottom).
Artificial intelligence is used to work out factors (e.g., the properties required of a solvent used for dispersion) that are important for achieving a goal (in this case, wanting to control the exfoliation process).



A large step toward the development of a catalyst for hydrogen evolution reaction

The "coatability" of polypyrrole film is not the only factor to its viability; its quality also comes into question. At first the film was not sufficiently conductive to be used, but this problem was solved by changing the strength of the oxidant (a benzoquinone derivative) combined with pyrrole during its formation. "With that I thought that my research had reached its ending point, until one of my students said that they would attempt to synthesize the film by means of combining pyrrole and benzoquinone without substituents. Drawing from my own chemical knowledge, I warned the student that the reaction would not be feasible due to insufficient oxidizing power, but they proceeded with experiment anyway," Oaki remembers.

Their effort succeeded, resulting in the creation of a new polymer material consisting of a loosely stacked random network formed of pyrrole and benzoquinone (Fig. 1, right). Not only was it easily peeled off to produce thin nanosheets, but it was also found that the nanosheets function as a catalyst to electrochemically convert protons (H^+) into hydrogen (H_2). At present, hydrogen is of interest to researchers because it is an energy source that does not generate carbon dioxide. However, in general, since hydrogen production requires a platinum catalyst, there are inevitable cost and resource complications. This nanosheet has received a large amount of interest because it may present a possible alternative as a metal-free organic compound.

While Oaki claims that these two incidents were serendipitous, the story hints at Oaki's deep insight, chemical expertise, attentive ear when interacting with students, and ability to ignore "common sense" when pursuing an interesting lead that was necessary to capitalize on the fortunate turn of events.

Soft 2D materials are fascinating!

This is how Oaki's research focusing on conjugated polymers began. His methodology is to give flexibility (molecular mobility) to generally rigid conjugated polymers by various chemical methods, creating two-dimensional materials such as layered structures and nanosheets, and then explore their functionalities. Oaki's mindset is that "it is important to first enjoy creating substances; figuring out the new material's specific characteristics comes later." In the aforementioned case of a two-dimensional material made from polypyrrole and benzoquinone, "creating" was the important part of the process—its ability to catalyze hydrogen was a coincidental byproduct.

Oaki is also developing sensor materials that quantitatively detect heat, light, and force (Fig. 2, top) using a similar research methodology. When the diacetylene molecules arranged in layers are irradiated with ultraviolet light, the triple bond moieties polymerize with each other and turn blue. When this polydiacetylene is stimulated with heat, force, light, etc., the molecular chain is twisted and other subtle changes occur in the structure, causing the color to change. By manipulating what guest ions and molecules are inserted between the layers, scientists can adjust the material's responsiveness to external stimuli.

One of the materials developed using this process changes color depending on the frictional force created when brushing teeth. This then can be used to create a sensor that can gauge, based on color, an appropriate level of strength when using a toothbrush.

A new perspective on material development using artificial intelligence

Oaki due to his enjoyment of making new materials, has been incorporating "materials informatics (MI)" into his research since 2016 (Fig. 2, bottom). MI is said to accelerate research and development by utilizing informatics for material research. Specifically, he feeds the artificial intelligence a large amount of data about materials, and tasks it with finding possible factors necessary to discover new substances and improve their performance. However, in many cases, it is difficult to interpret exactly what the AI has done after processing the data.

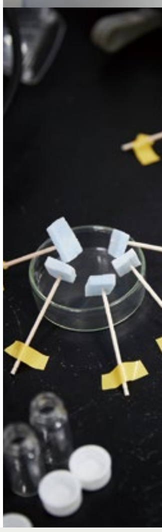
Oaki is trying to prevent his AI from becoming a "black box" (an AI system that gives no view of its inner workings) by judging whether the computer-generated factors are important or correct based on his own chemical knowledge. "It is important for researchers to know when to take the lead," he says, "But artificial intelligence can also accomplish a large number of tasks, so it is our job is to make good use of it."

I am looking forward to the next "unexpected discovery," and how AI innovation will aid in sharpening Oaki's insight.

(Interview and text writer: Akiko Ikeda)

Enjoying chemistry research to the fullest

In recent years in the world of chemistry, where applied and practical research tend to garner the most attention, Associate Professor Oaki spends his days developing new materials, a pursuit that he finds genuinely rewarding and engaging. "I'm the type who doesn't do something unless I'm convinced it's worth it," he says, revealing his stubborn side, but also the source of stalwart determination that has guided both his life and research. And now, moving forward, he has decided to devote his energy to sharing the joy of chemistry with the next generation.



What kind of childhood did you have?

I don't have a strong recollection of when I was little. I remember playing the flute in brass band during junior high school and high school. It may have also been due the influence of my mother who was involved in music, but I became totally absorbed in band. Even though I hated being forced to play piano when I was little, I think I was able to try my best with flute because it was something I chose by myself.

I have such fond memories of grueling practices with my band mates as we tried to get into the Kanto regional championships. In retrospect I think it was a bit of the Showa-era die-hard spirit, but it made me mentally tough. My music teacher was incredibly strict, but I am grateful for the lessons they taught me about etiquette, navigating relationships, and teamwork.

When did you become interested in chemistry?

All the way until around junior high school I resented my studies, not knowing why I had to do them. The first subject that I actually liked was physics. I was first inspired by a single-sheet printout that my high school physics teacher gave me. The handout organized all of the formulas from our physics textbook and explained their meanings and relationships so that they were easy to understand.

And yet, even though my father specialized in it, I wasn't very interested in chemistry. It wasn't until I studied under my preparatory school teacher who taught me that chemistry also held meaning beyond simple memorization that I started to find it interesting. It just goes to show that no matter what you do, you can't enjoy something until you understand it.

And from there you decided to major in applied chemistry at Keio University and become a researcher. Would you share some of your thought process?

There were times when I really wasn't sure about it. Actually, I had trouble deciding between physics, chemistry, and mechanical engineering when it was time to take the university entrance exams. The reason I chose Keio University was because students weren't required to declare a major until starting their second year. Before settling on chemistry, I even considered entering the School of Medicine and there was a time during the second year of my master's program when I thought about joining the corporate workforce. However, when it came time to search for a job, I started to feel like I was leaving my research incomplete. That was the turning point for me when I decided to pursue a PhD.

What is the environment like in your research laboratory?

When I return to my office I sometimes drop in on the students. They probably find it annoying, but for me to see students conducting experiments and their samples is just as fun as conducting experiments myself.

I consider my students to be colleagues who share in our collective goal of "enjoying the best research in the world."

Above all, I want them to personally invest in and enjoy their research. In order to get there, I believe it is a teacher's role to help students have their "lightbulb moment." Every student is different in what leads them to that moment, but once they light up, their progress always exceeds my expectations.

Tell us about what happened 10 years ago that has drastically changed your research.

As I mentioned during my research introduction, I am always learning from my students, and together we have come across quite the variety of "unexpected discoveries." As a result of these outcomes, I decided about 10 years ago to change my research to focus on such discoveries.

By telling the students of these past happenings, they feel compelled to come and show me samples when something unusual occurs during an experiment. "Unexpected discoveries" are not so common, but we find something interesting in about one in 20 reports. Humans naturally tend to treat things as failures when they don't go as expected. In those moments, in addition to asking my students to think about what could be done to make the original experiment a success, I want them to consider the possibility that a change of perspective might lead to something new and to challenge the notion of what is "expected" in the first place.

There was another reason I changed my research. It was right around that time when I was told by fellow researchers that there was "nothing of interest in your research lately," that "you should change your research so that it differs from your PhD," and other statements along the lines of "you should focus on things that

you're actually capable of." This is what got me to think I actually should pursue an independent course of research, blaze new trails in research based on my own discoveries, synthesize unique materials, and aim to develop special uses for them.

Even though you have you plate full with your research, you also seem very active in planning academic conferences and engaging in various outreach programs. Why is this?

I consider nurturing the next generation through promoting interactions between industry, academic, and government institutions to be another important aspect of my work. I want my students to share in this idea. I always tell them, "you've gone through so much effort to learn about chemistry, it would be a waste to not help bring up the generation after you through engagement in multi-discipline outreach." This may just be my way to get them to help me out, but I believe that it is also a worthwhile experience for them.

As I said, I didn't know how interesting chemistry could be until I started attending preparatory school before taking university entrance exams. That was because I was never exposed to an environment where chemistry could be perceived as interesting during elementary and junior high school. Many children would have curiosity in chemistry if they just had the chance. I want to be a part of helping those kids experience the fun of chemistry. I also hope that by being involved in the planning of new lectures and academic journals, chemistry in Japan will flourish and attract more public attention.

Would you say that Keio University represents a place where, in addition to research, professors are encouraged to invest in and foster the minds of the next generation?

I would agree, though I haven't done anything special myself. If you are doing research alongside students, it will naturally become an educational experience.

In recent years, interdisciplinary and joint research projects that involve other fields have increased, so solitary research that I do all alone has given away to a more collaborative approach with co-researchers. I first want my students to learn how to interact with researchers from other fields who don't speak in the same technical language, how to enjoy while collaborating to obtain the best results, and how to communicate not only as a researcher but also as a well-rounded human being.

How do you feel about Keio University as a place to conduct research?

While you can't say that Keio's faculties are particularly large-scale, I would say that they are appropriately sized. I used to think that the bigger the university, the better, but at some point, after conversing with teachers in the School of Medicine, I realized that what makes it so easy to communicate with teachers in other disciplines is that the university isn't too big. There is also a sense of unity and community at Keio.

I hope that myself and others able to use this great advantage while conducting research here.

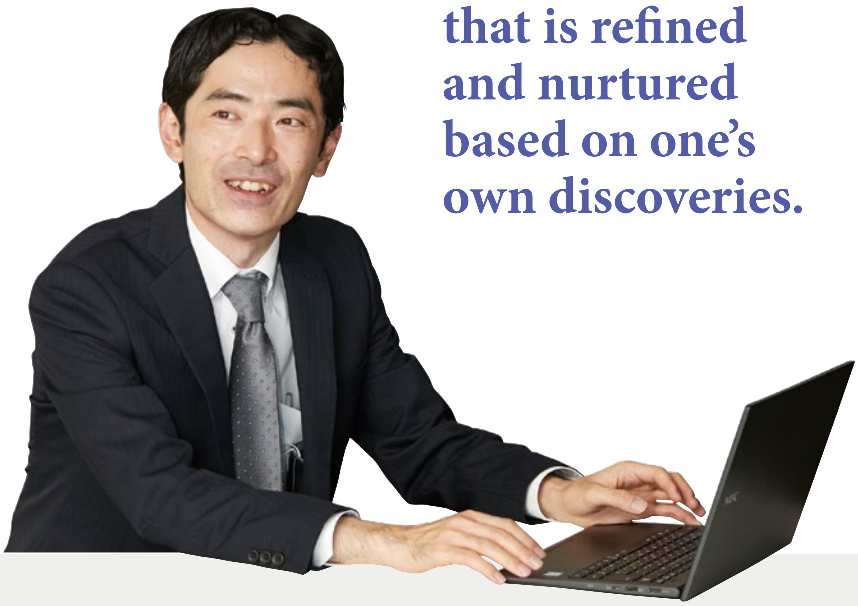
◎ Some words from students ...◎

● In a variety of fields there is a growing demand for the development of visual indicators for stimuli such as light, heat, and force. I've taken over research handed down from upperclassmen on materials that change color from external stimuli. The main reason that I chose to study with Professor Oaki was that the research felt very future-oriented, but another decisive factor in choosing his laboratory was the welcoming atmosphere. Here, students from different academic backgrounds share knowledge and push each other while enjoying their research. After completing my Master's degree I plan to enter the work force, but I will cherish the research and communication skills I learned here and intend to use them to expand my horizons. (1st-year master's student)

(Interview and text writer: Akiko Ikeda)

For the full text of this interview
https://www.st.keio.ac.jp/en/kyurizukai/

I realized that research is something that is refined and nurtured based on one's own discoveries.



Yuya Oaki

Department of Applied Chemistry

Specializes in conjugated polymer materials, layered materials, nanosheet materials, and materials informatics. Graduated in 2002 from the Department of Applied Chemistry, Faculty of Science and Technology, Keio University. Completed doctorate program from the same faculty in 2006. Ph.D. (Engineering) After finishing his position as a Research Fellow (PD) of the Japan Society for the Promotion of Science, he has worked as an Assistant Professor in the Department of Applied Chemistry at Keio University since 2009, a Senior Assistant Professor since 2012, and an Associate Professor since 2016. From 2016 to 2020 he was a JST PRESTO researcher, and from 2018 to 2020 he was also a Senior Scientific Research Specialist at the Ministry of Education, Culture, Sports, Science and Technology (MEXT).

Mingling with young Chinese and Japanese researchers

I participated as a member of the Japanese delegation to the "1st China-Japan Joint Symposium for Young Polymer Scientists" held in Xi'an, China, in October of 2019. It was an amazing opportunity to get firsthand insight into China's high level of research that has been making headlines across a variety of fields.

Presentation of research results.

A reception with Chinese presenters and hosts
On the right is Professor Zhang Xi, the chairperson of the organization.

Promoting and raising awareness of chemistry

I have been an active organizing member in the Chemical Society of Japan's "CSJ Chemistry Festa," which aims to deepen exchanges between industry, academia, and government; the Chemistry Daisuki Club for elementary and junior high school students; and other groups for nearly 10 years. The organization hopes to encourage the enjoyment of chemistry within the next generation and promote collaborations between in industry, academia, and government. I personally enjoy gathering at the receptions after committee meetings, talking with researchers from all over the country about various topics, and hearing stories from company insiders behind their popular products.

Yuya Oaki's ON and OFF

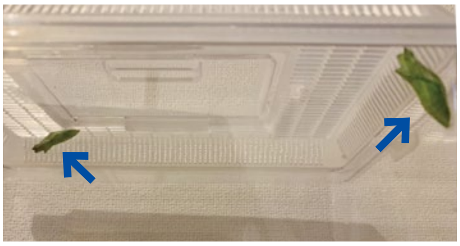
Looking to have fun while both "on" and "off"

Research conferences

The JST PRESTO (one of the research programs of the Japan Science and Technology Agency) organized discussions at a camp-style research conference. Stretching from early morning to late evening, the event provided a wonderful opportunity to focus on topics at hand through research reports, Q&As, and other discussions. After dinner, there were sessions led by veteran professors who were advisors to the project research which were filled with laughter and personal perspectives on different careers and research. I found it to be a very enlightening experience.

Raising butterflies with my family in the summer

We used to find larvae and keep them until they transformed into butterflies.

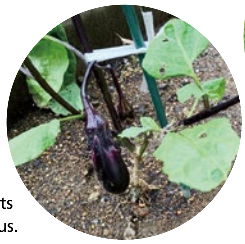


When they are in their pupa stage they keep still.

And when their wings have dried, they leave their cocoon behind.

Growing tomatoes and eggplants at home

We were late to plant this year, so it wasn't a good harvest.



The eggplants were delicious.

The tomatoes, on the other hand, were slightly sour and I wish we could have let them ripen more.

A scene of me playing with my daughter

Here we have a tea party set up in a play house attended primarily by pandas (note: this was before COVID-19).

Playtime often involves blocks and toys that can be stacked.

私の本棚

My favorite books

Native Speakers' English Grammar

A book my father gave me and the first installation of the Native Speakers Series. When writing papers about research findings in English, I still find myself constantly worried about definite/indefinite articles, singular/plural usage, tense, prepositions, and which word to select from a list of synonyms. The book is divided into several parts such as "grammar" and "prepositions," with each section introduced depicting a native speaker's impression and sense of the subject with illustrations. I'm bad at English, but even I am able to absorb and learn the material.

Polymer Electronics

A comprehensive breakdown of the synthesis, structure, and characteristics of conductive polymers. I find it particularly helpful as it also describes various methods for synthesizing conjugated polymers including specific reagent amounts and reaction temperatures, a rarity among reference books. When I started researching conjugated polymers, this kind of information was very helpful. Because the information on properties etc., is written in such a detailed and accessible way, I think it is a perfect primer for learning about conjugated polymers. This is one of the books I purchased at the recommendation of my supervisor in the laboratory when I was a postdoctoral researcher.

A House Of 100 Stories

This is one book from the "A House Of 100 Stories" series. The books are organized around themes such as *sora* (sky) and *chika* (underground), with residents differing every 10 floors. In this picture book, people and animals invited to the house make their way to the one-hundredth floor. The residents vary depending on the theme, but examples include squirrels, ladybugs, wind, rainbows, and thunder, all living within a 10-stories in their own original and imaginatively designed rooms. I happened across this book while traveling a couple of years ago and was riveted by the novelty of the content and illustrations, the exuberance, and the format which keeps you wanting to find out what is on the next page. I honestly wish that I could delivery my research as well as this book tells stories.

Doraemon

I think that some of Doraemon's gadgets have already come into existence, with mobile phones as the *ito-nashi ito-denwa* (string phone without a string) and automatic translators as the *honyaku-konnyaku* (translation gummy). I believe that other gadgets may also be invented in the future. When I think about how the authors planned these gadgets, I am both amazed by their ingenuity and feel as though they have presented scientists with a vision for what the end product of our research could be. While I have stressed the more serious side of Doraemon, I do simply enjoy getting lost in its story and the adventures that the characters go through. One of my personal favorites is the *baibain* (multiplication liquid) from volume 17. Here Doraemon teaches us how incredible "2^n" is in an episode in which a *kuri-manju* (a Japanese confectionery containing chestnut paste) infused with *baibain* doubles every five minutes ($n = 1, 2, \ldots$).

Quarterly Review of Chemistry No. 42: Inorganic-Organic Nanocomposites

When I was an undergraduate, I couldn't even imagine the combination of inorganic chemistry and organic chemistry. Then, as a graduate student I picked this book up at the library and was impressed with idea that the organic and inorganic could be combined, fusing together the positive aspects of both. In particular, I found it impressive that functional molecules were introduced (intercalated) between the layers of the layered inorganic compound, and the molecules were aligned in the nanoscale space. At that time, I was lucky enough to get an autograph from the professor who edited this book, who in turn encouraged me to work hard so that I could make a similar book in the future.

Does synergistic effect only exist in layers?

Yuya Oaki

While it may be overly ambitious, for me the synergistic effect is more significant than 1+1 = greater than or equal to 2. I believe that its multiplicative potency is as powerful as the '*n*' in 2^n, like the *baibain* from Doraemon that I introduced earlier from my bookshelf.

As you've mentioned in this publication, I am conducting research with the aim of creating soft and functional two-dimensional materials. In order to move and work the incorporated molecules, we are synthesizing materials with a soft, two-dimensional anisotropic structure. To explain more concretely, we take the "ingredients" of heteroaromatic polymers such as polypyrrole, quinone derivatives, layered polydiacetylenes, and layered inorganic compounds, then shape them into two-dimensional "structures" such as nanosheets or flexible network polymers, thus enhancing molecular motion for the dynamic functions. These products can then be used in various applications such as in lithium ion secondary batteries, hydrogen-generating electrocatalysts, light/heat/force sensors, etc. Using these avenues I hope my research can make contributions to medical care, environmental needs, energy issues, and resource management.

By combining the synergy between materials' properties (derived from its molecules and compounds) and the two-dimensional anisotropic structure, we hope improve performance and develop new functions that were not previously possible in the modern world. Under the framework of the aforementioned structural/functional research topic, my students are tasked with the thorough investigation of what makes this research new when compared to other studies, what materials can only be made via these methods, and what unique performance/functions are made possible by the developed materials. Young students have extraordinary brains and abilities that I lack. Like my research, I have discovered a "synergy" when I have involved students in the process, leading to major breakthroughs and unexpected discoveries.

When I think back on it all, there have been all kinds of serendipitous moments that have occurred because of this synergy—whether it was through a student bringing me their experiment samples as I passed by, an experiment that I personally thought was pointless but then gave surprising results, students taking ownership of difficult programs or principles, or the simple power of countless experiments and sheer will. My job very well may be to create an environment that produces that synergy, harnessing the collective power of my students.

I also find that synergy when working with researchers inside and outside the university. For example, in "Materials Informatics," which accelerates the research and development of materials based on data, we are indebted to a professor who is an expert in data science. Collaboration with researchers from different fields such as data science, doctors, and researchers in industry, not to mention collaboration with researchers in the closely-related fields like polymer chemistry, garners new insights for both students and myself every time we interact, resulting in a great synergistic effect on research. Using the synergy from all of these areas, I hope to harness the power of the "*n*" in 2^n to conduct the best research possible.

理 工 学 Information

KEIO TECHNO-MALL 2021 22nd Annual Keio Science and Technology Exhibition "Beyond imagination: encouraging advances to the future"

The KEIO TECHNO-MALL is an event showcasing the research findings of Keio University's Faculty and Graduate School of Science and Technology and the Keio School of Medicine, as well as a platform to facilitate collaboration between industry, government, and academia through joint research projects, technology transfers, and more. We pride ourselves that the event is the one of the largest exhibitions held by a science and engineering university, with numerous participants, ranging from private corporations, government entities, and other universities, attending each year.

In order to prevent the spread of COVID-19, this year's proceedings will be held online as they were last year. The event will utilize online tools and platforms and deliver interactive sessions to recreate the experience of visiting booths at the actual event. While you won't be able to experience the real thing, we hope that you will enjoy the features unique to the revamped online KEIO TECHNO-MALL We also hope that those who have faced physical difficulties attending previous events will seize the opportunity to join us online. See you at KEIO TECHNO-MALL!

Date and time: 10:00–18:00, Friday, December 10, 2021
Details: Research Exhibition
(featuring research explanations, introductory videos, and other materials)
-Live events and panels will be offered throughout the day
-Introduction of online research
Host: Keio Leading-edge Laboratory of Science and Technology (KLL)
(Keio University Faculty and Graduate School of Science and Technology, School of Medicine)
For more details: www.kll.keio.ac.jp/ktm/

Editor's postscript

Associate Professor Yuya Oaki says that he learns a lot from the watching the interactions of working researchers and that he appreciates the very "human" element of the event. Listening to stories from his pupils, it appears that Oaki himself has become a respected figure in their eyes. Considering the exemplary behavior he showed to our staff while gathering materials for this article, the care he took in providing drinks for everyone, and the professionalism he displayed in proofreading the manuscript, we hope that our article will highlight his attention to detail and courteous personality.

This issue marks the first publication since the COVID-19 pandemic began. As such students are pictured wearing masks while conducting their research. We hope that by the time of the next publication, things on campus will have returned to normal. (Yurina Tomohisa)

Cover of current issue : Professor Oaki standing next to an atomic force microscope for examining nanosheets.

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