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Doctoral Dissertation

Study on New Adhesion and Friction Based on Stress Dispersion Effects of Surface Microstructure of Organisms

(生物表面微細構造の応力分散効果に基づく新たな接着・摩擦に関する研究)

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Chapter 1

Intoroduction

1-1 Biomimetics

1-1-1 Two approaches to engineering design

Manufacturing is an excellent problem-solving technique that humans have. Humanity has developed by creating various things over the past hundreds of thousands of years. When it comes to solving problems with materials, which are manufactured products, functionality is the key. There are two approaches to expressing new functions in a material¹. One is to utilize the properties that the material has or to give it new properties, and the other is to realize it by new structuring processes, giving it structure, or controlling it. Humans have carried out this engineering design (expressing the required functions) as needed, but in recent years, engineering design that learns from living things (biomimetics) has been attracting attention. Humans have carried out this engineering design (expressing the required functions) as needed, but in recent years, engineering design that learns from living things (biomimetics) has been attracting attention.

1-1-2 The History and Importance of Biomimetics

The term biomimetics was coined in the late 1950s by German-American neurophysiologist Otto Schmitt. However, the importance of "learning from nature" is common knowledge in various fields of science and technology, and the idea of biomimetics existed before the term became widespread. A representative example is the flying machine developed by Leonardo da Vinci, a painter and scientist from around the 15th century, which focused on the flight mechanism of birds. In recent years, Schmitt's trigger and Velcro have also been developed. In the 1970s, biomimetics research appeared in the field of chemistry as Biomimetic Chemistry (molecular-level imitation of enzymes and biomembranes). Around this time, X-ray structural analysis revealed the chemical structure of the reaction site of enzymes, which are biocatalysts, making it possible to use organic chemistry techniques to elucidate biological reactions from a molecular perspective. Meanwhile, in the 1970s, biomimetics research also developed in the fields of mechanical engineering and fluid dynamics, leading to the development of robots that mimic the flight of insects and the swimming of fish, as well as sonar and radar that mimic the echolocation of bats and the sensory hairs of insects, which came to be called mechanical biomimetics. Research on mechanical biomimetics has been carried out mainly in industrial fields such as the military industry, railways, ships, and aircraft industries, and has also influenced cutting-edge technological fields such as micromachines and MEMS (Micro Electro Mechanical Systems). Furthermore, in this century, with the invention of the electron microscope and the advancement of nanotechnology, morphologists and taxonomists have revealed the hierarchical structure of the body surface of living organisms on the nm to μm scale. The hierarchical structures of these organisms have given us a hint, and by artificially creating these structures and adding them to materials, it has become possible to express functions due to these structures. These are called material biomimetics, and are a new trend in biomimetics². While the aforementioned biomimetic chemistry was born from the "combination of biology and organic chemistry" using X-ray structural analysis, material research, which is a "new generation of biomimetics," can be said to have been born from the "combination of natural history, biology, and material nanotechnology" based on electron microscope observation and microfabrication technology. European nanotechnology aims to integrate and collaborate with other fields, as symbolized by the catchphrase "Nano meets Bio." In addition, the fact that interdisciplinary collaboration is a

prerequisite for obtaining research funding in this field at German universities shows that interdisciplinary collaboration is an essential element in biomimetics.

In this way, the importance and benefits of learning from living things include the large amount of information. When a problem occurs, humans find a solution by trial and error using experience and inspiration. In addition, humans solve problems by circulating theories from many solutions, which can be used to solve the next problem. On the other hand, many living things have used evolution to solve various problems such as securing energy sources, reproduction, the struggle for survival such as predation and prey that began with the Cambrian explosion, and adapting to the environment. The evolution of living things in the history of the Earth is said to have lasted for several billion years, which is extremely long compared to the history of humanity, which is said to be hundreds of thousands of years since *Homo sapiens* became a species and several thousand years since civilization was established. Therefore, living things have information that is incomparable to the knowledge accumulated by humans, and can be said to be a treasure trove for researchers. Therefore, it is very inefficient to solve problems in human society by learning and theorizing only from human wisdom, and I believe that we are not making good use of the information resources that are overflowing around us.

1-2 Surface Microstructure Biomimetics

1-2-1 Representative examples of surface microstructure biomimetics

There are many living creatures that serve as models for the material (surface microstructure) biomimetics that utilizes the information possessed by living organisms in manufacturing^{3,4}. Examples include geckos (adhesion)^{5,6}, morpho butterflies (structural color)^{7,8}, lotus leaves (Superhydrophobicity)⁹, and moth eyes (anti-reflective)^{10,11} (Fig. 1-1). Each of these creatures has a microstructure that allows them to exhibit excellent functions, and many materials that mimic their principles have been developed and are being used¹²⁻¹⁷.

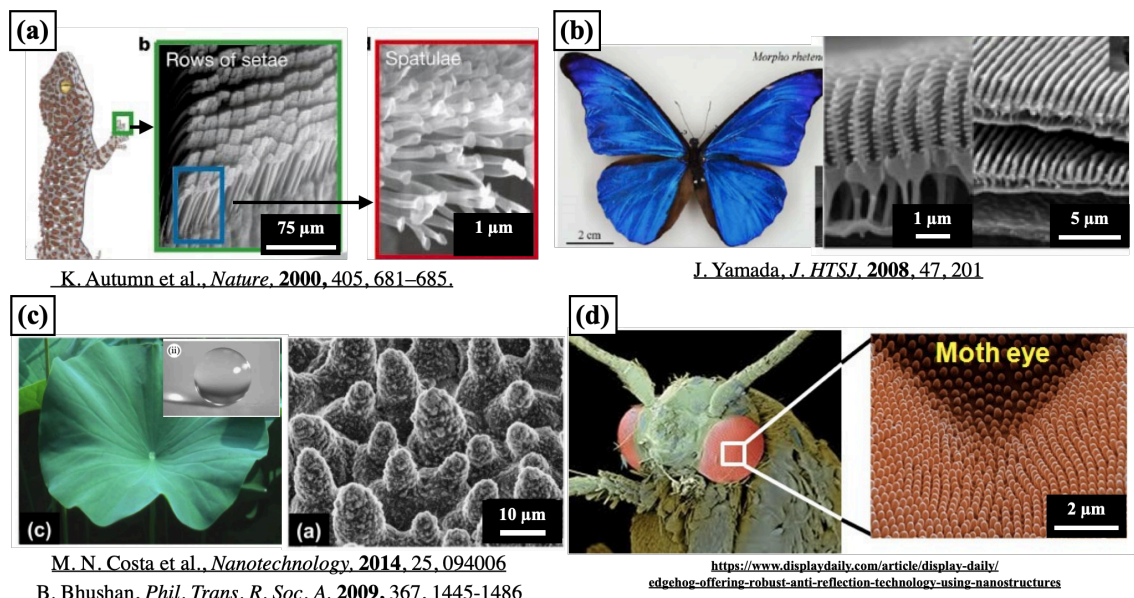


Fig. 1-1 Photographs and surface microstructures of (a) gecko, (b) morpho butterfly, (c) lotus leaf, and (d) moth eye

Geckos are one of the most famous and well-studied organisms that produce properties through surface microstructures. Geckos can adhere to any surface (wood, glass, rock, etc.) quickly and repeatedly and move freely. In the past, geckos' adhesion was thought to be due to the strong frictional forces caused by the hair-like structures present on their fingertips that fit into the gaps between surfaces¹⁸. However, the friction theory proved to be false, as geckos were able to climb even on smooth surfaces¹⁸. On the other hand, Kellar Autumn et al. stated that van der Waals forces were responsible for adhesion^{5,6}. This is because the microstructure of gecko fingers, with a density of

approximately 5000 hairs/mm², 30~150 μm in length, and 5 μm in diameter (seta), and a 200~500 nm spatula structure at the tip of the hair-like structure, increases the contact area when the finger touches the surface and generates large van der Waals forces. The force is 10 N/m per cm². The force is 10 N/cm² = about 100 kPa, enough to withstand a force of about 200 μN per hair^{5,6}. This is calculated to generate enough force to support the gecko's body, which weighs 80 mg per hair and 40 kg in total for the four limbs, so that adhesion is possible using van der Waals forces alone¹⁹. However, Sun and Waite et al. proposed that capillary forces create the adhesive force because the humidified environment affects the adhesive force of the spatula to the hydrophilic substrate¹⁸. They also supported the capillary force theory with data showing that increasing humidity on hydrophilic substrates increased the adhesive force of spatulas¹⁸. Subsequently, Puthoff et al. and Prowse et al. conducted experiments to resolve the question of whether gecko adhesion forces are produced by capillary or van der Waals forces. Thereby they found that humidity affects the mechanical properties of the keratin that makes up the spatula's structure. They found that high humidity makes the spatulas more flexible and increases the interaction (contact area, van der Waals forces) between the spatula and the surface it touches. Through these studies, it was demonstrated that van der Waals forces, rather than capillary forces, contribute to the adhesive strength of geckos¹⁸.

Since the theoretical elucidation, many mimetic structures using synthetic polymers (polydimethylsiloxane (PDMS), urethane polyacrylate, polymethyl methacrylate, polyethylene, etc.) have been created (Fig. 1-2) and their properties have been investigated. The results showed that the adhesive properties of mimetic adhesive materials vary depending on the hair density, hair orientation, elastic modulus and surface energy of the hairs, and hair shape (length, diameter, aspect ratio, and tip shape)²⁰. It is said that adhesion by these hair-like structures (dry adhesion) increases the adhesive strength due to the interaction between the two contacting surfaces and the increase in contact area (contact partitioning) caused by the hierarchical hair-like structure, as mentioned above . On the other hand, because it is difficult to synthetically mimic the setae and spatula structure of geckos, the adhesive strength of these mimetic adhesive materials is often inferior to that of geckos²⁰. However, in recent years, gecko-mimetic tapes with adhesive strengths comparable to that of real geckos have been created by using aligned carbon nanotubes (CNTs) with diameters of 1 to several nm (Fig. 1-3)²¹. Aligned grown CNTs are difficult to produce and expensive, so they are not yet used in

everyday products. However, in March 2020, Uesugi et al. of the Japan Synchrotron Radiation Research Institute (JASRI) published a paper on using CNT Gecko Tape as a sample support in solid-state material structure analysis²². Materials imitating lotus leaves have also been actively researched, and as a result of the advancement of the principle elucidation, they have been applied to yogurt lids and other products. A superhydrophobic surface like that of a lotus leaf prevents yogurt from sticking to the lid, which makes it possible to reduce food waste²³. In this way, surface microstructures and their functions can be reproduced in artificial materials by mimicking the structure based on the principle elucidation, and depending on how they are used, they can provide a clue to solving problems faced by humans. Therefore, research that learns from living things - discovery, elucidation of the principles, and application of surface microstructure functions - is very important. However, most biomimetics research to date has been conducted by people in the engineering field or industry, and in the absence of research showing the functions and structures of living things revealed by researchers in the biology field, it was necessary to wait for new research results on biological functions²⁴. Alternatively, research has been conducted based on inspiration only from the structure of living organisms, with the function of the organism remaining unclear. Since this is an idea based on human experience, it may differ from the actual function of living organisms, making it less valuable as an information source. In addition, research inspired only by the structure and using materials different from those of living organisms has sometimes been called biomimetic. Even if the above problems are overcome and it is possible to investigate the function of living organisms, microstructures can sometimes exhibit multifunctionality, so different functions may be hidden, or multiple functions may not be reproduced simultaneously. In fact, it is said that it is difficult to repeatedly adhere CNT tape like geckos because the CNTs are crushed.

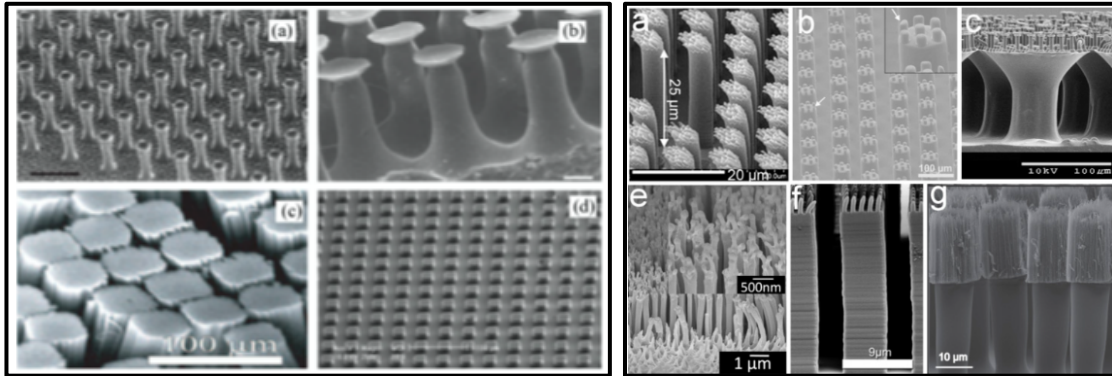


Fig. 1-2 Images of various gecko-mimetic adhesive materials (left)²⁰ (right)¹⁴

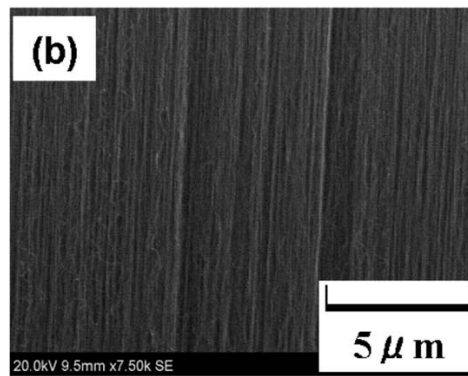


Fig. 1-3 SEM image of highly aligned CNTs²¹

1-2-2 Effect of structure on mechanical strength

It has been reported in various fields in the past that living organisms have better knowledge of material design than humans, and among them, the impact of structure on mechanical strength is extremely large ^{1,25}. In fact, from the perspective of engineering design, biomaterials are inferior to artificial materials in terms of single material properties such as strength and elastic modulus¹. Material properties that are preferable are light, strong, and have small deformation, but the material indices of specific strength (material strength/density) and specific elastic modulus (elastic modulus/density) of biomaterials are lower than conventional materials such as iron and aluminum, as well as advanced composite materials. However, biomaterials such as wood, shells, and bones are known to have better multifunctionality than artificial materials and are optimally designed for the environment. In fact, when pillars are made from wood, the properties are reversed, and the material (structural) index is several times higher than that of artificial high-tensile steel ¹. This means that living organisms can express superior functions through the structure even if they use materials with inferior material properties. Until recently, in human society, it was considered preferable for the structure of materials to be as homogeneous as possible from the perspective of strength, chemistry, and other functions. However, when looking at biomaterials and their structures, it is difficult to say that the composition, tissue, and structure of the materials are homogeneous. For example, the cross-sectional structure of bamboo is a composite of multiple materials. This results in a form that is well suited to the external load conditions and the intended use, and past researchers have shown that this fiber distribution exhibits maximum bending strength and bending rigidity with the minimum amount of material ¹. Materials that exhibit excellent functions by being heterogeneous rather than uniform can also be seen in wood, ivory, shells, etc. other than bamboo, and humans also create various materials as composite materials^{3,26}. For example, advanced composite materials are artificial composite materials that exhibit high specific strength and high specific elasticity by adding fillers (e.g. carbon fiber, glass fiber, etc.) to synthetic plastics. In addition, while composite materials basically have a constant distribution of dispersed phases (fillers, etc.), there are some materials that exhibit even more excellent functions by dispersing the dispersed phase non-uniformly (continuously or locally from one side to the other). The distribution of vascular bundle sheaths in bamboo is not uniform, and increases toward the outside. This makes it possible to combine the seemingly

contradictory properties of flexibility and high strength. This is a good example of how the latter approach described in 1-1-1 can be used effectively to achieve excellent functionality.

On the other hand, there are not many studies on the strength of materials based on the surface microstructure of living organisms, and most of the research is focused on the internal structure of materials such as turtle shells, bamboo, and bones^{3,27–29}. Conversely, metamaterials, which are a completely artificial material, have recently been studied as a material strength change because the way stress is transmitted differs depending on the shape of the surface structure^{30,31}. Therefore, it should be certain that the structure affects the strength of materials, but there has been little linkage with the surface microstructure of living organisms. In addition, most research on metamaterials, turtle shells, bamboo, and bones is also focused on material strength. Since material strength is related to the distribution of deformation and stress from the knowledge of structural mechanics, it is thought that it will also be effective in contact forces such as friction and adhesion, which are forces acting on the interface between two contacting surfaces, outside the field of material mechanics, but there is very little linkage with these.

1-3 Biomimetics for a sustainable society

1-3-1 Human impact on the global environment

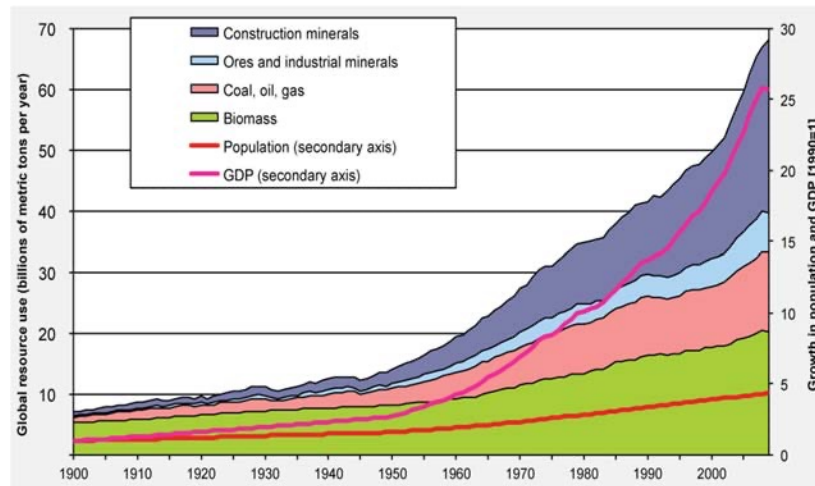


Fig. 1-4 Growth consumption of certain resources vs. population and GDP growth³³

In 1-1 and 1-2, we discussed the functionality of the microscopic surface structure of living organisms and its application to human manufacturing. In this section 1-3, we will discuss the common problems that humanity must solve in manufacturing in the first place. Humanity has solved many problems by using its intelligence to create and use superior materials. In particular, the development of industry and the economy in the past few decades has been remarkable, and the world population has increased 2.5 times due to advances in medicine and agriculture. However, it is said that along with this, human resource consumption has also exploded^{32,33} (Fig. 1-4). According to reference³⁴, in the 50 years from 1950 to 2000, the number of automobiles increased 10 times, oil consumption increased 7.3 times, natural gas increased 14.5 times, coal increased 3.6 times, and power generation capacity increased 21 times. Until now, humanity has held the belief that "nature is infinite and free, and that economic growth can continue in perpetuity by making effective use of resources obtained from nature," but in recent years this has been deemed incorrect, and the belief that the Earth is finite is becoming mainstream. In fact, there is an index called the "ecological footprint" that compares the production capacity of the Earth's ecosystem with the Earth's resources that humans use and utilize. According to the trend of the ecological footprint of the entire human race calculated from this, it can be seen that human consumption exceeded the Earth's

production capacity around 1987 and is still increasing. In other words, since 1987, human beings have not been able to live on the products provided by the Earth's environment, and have been consuming non-renewable resources such as fossil fuels that have accumulated in the past. This excessive consumption activity has caused various environmental problems. Major global environmental problems include deforestation and desertification, depletion of mineral resources, marine pollution, reduction of biodiversity, and acid rain. It is said that the intensification of human economic activity has caused various environmental destructions, which will ultimately threaten the economic foundation of human beings and ultimately the foundation of their survival.

1-3-2 Sustainability and the SDGs

Due to the circumstances described in 1-3-1, the stance of mass production and mass consumption as in the past was reviewed, and a new guideline for human society was established worldwide. It is called sustainability. Sustainability is said to be "the ability of the global environment to maintain an environment in which we can live" ³⁴. This term was first used in 1987 when "Sustainable Development" was taken up as a challenge for humankind in the report "Our Common Future" issued by the United Nations Commission on Environment and Development (commonly known as the Brundtland Commission), where it was defined as "development that meets the needs of the present generation without compromising the ability of future generations to meet their own needs." This led to widespread recognition around the world. Later, at the Earth Summit held in Rio de Janeiro, Brazil in 1992, it was agreed worldwide that "as a common goal for humankind, we must shift from the current economic growth-first approach to development that takes into account the Earth's ecosystem (the Earth's environmental carrying capacity)."

Nowadays, the word "sustainability" does not only mean the sustainability of the global environment, but also implicitly includes the sustainability of human socio-economic systems. International organizations, in particular, have come to strongly recognize that before the collapse of ecosystems, the distortions in human society, such as the widening gap between rich and poor on a global scale and the worsening poverty problem in developing countries, may threaten the survival of human society. At the United Nations Millennium Summit held in September 2000, the United Nations Millennium Declaration was adopted as a measure to improve these problems, setting goals for the international community in the 21st century. These Millennium Development Goals (MDGs) were mainly focused on supporting developing countries, but in reality, there was strong opposition from developing countries, and some goals were not achieved. In response to this, the United Nations Sustainable Development Summit was held at the United Nations Headquarters in New York in September 2015, and the 2030 Agenda for Sustainable Development was adopted as a long-term development guideline from 2015 to 2030. The "Sustainable Development Goals" at the core of this document are called the SDGs, and unlike the MDGs, they call on all countries to pursue prosperity while protecting the earth (Fig. 1-5)³⁵. In addition to protecting the global environment from climate change, marine pollution, and deforestation, these goals

also include the elimination of gender and national inequalities, disparities, and discrimination, urban development, and industrial and economic development. The SDGs were created to preserve the global environment, but unless the elimination of gender and national inequalities, disparities, and discrimination is achieved and a society in which everyone feels safe is realized, environmental protection could become a new burden for developing countries, the poor, and poor countries. If this happens and dissatisfaction and distrust towards environmental protection itself grows, the future will be far removed from the original purpose of preserving the global environment. Therefore, environmental protection should not be the only thing that shifts resource consumption to other ecosystems, poor countries, societies, and companies, but rather environmental protection must promote economic activity, industry, or national and regional development that transcends ecosystems and borders.



Fig. 1-5 The 17 SDGs³⁵

1-3-3 Differences between human and biological technological systems

In 1-3-1 and 1-3-2, we mentioned that the current activities of human society have a negative impact on the earth and ecosystems that far exceeds their capacity, and that goals such as the SDGs have been established to improve this. In the world before modern capitalism, required functions were individually achieved, and even if some waste or burden on the environment occurred, it was not excessive because productivity was low. However, in recent years, thanks to the development of science and technology and industrial society, the excessive use of exhaustible resources, disposable products, and the discharge of waste into the environment have become causes that threaten the survival of the global environment and, ultimately, the human race. In contrast, living things and humans have clearly different ways of solving problems. As mentioned above, since the industrial revolution and the IT revolution, humans have used "fossil resources" as materials and energy sources, and have used "high temperature and high pressure" to create things, and for microstructures, "lithography and etching" which are high energy, high cost, time, resources, and special equipment. In contrast, living organisms use the same carbon compounds, but mainly "biodegradable materials," to create objects and microstructures through "molecular assembly and self-organization at room temperature and pressure, which is a natural phenomenon and therefore has a low environmental impact because it uses natural phenomena" ². In addition, Professor Barthlott and his colleagues have commented on the multifunctionality of the hierarchical surfaces of living organisms with regard to their surface microstructures⁹. For example, the moth-eye structure is found not only in moths but in many other insects, and is known for having various properties such as anti-reflective, super-water repellency, and antibacterial properties in a single structure. (Fig. 1-6 ⁹). Furthermore, these excellent functions are realized by the structure, such as the surface microstructure, so the properties of the

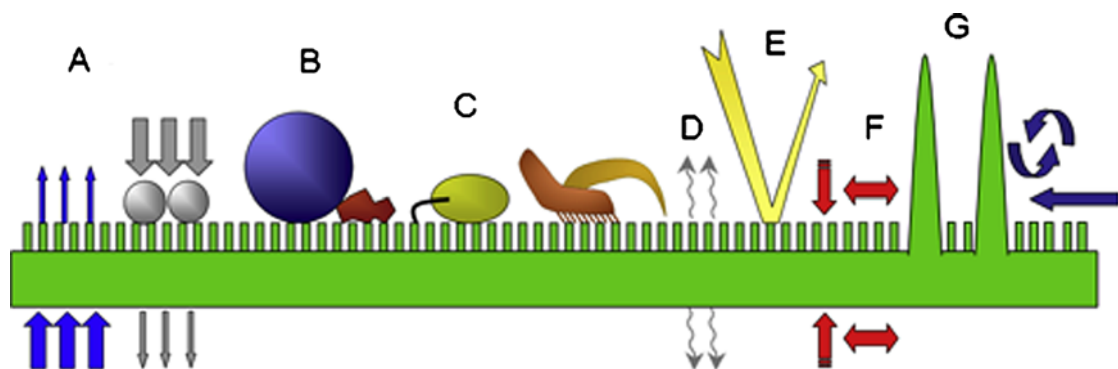


Fig. 1-6 Multifunctionality of surface microstructures⁹

material are not lost. The reason why the CNT gecko tape in 1-2-1 was used as a sample support in the analysis of the structure of solid materials is that the structure does not require a polymer adhesive like a normal adhesive tape, and clean adhesion is realized without contaminating the adhesion interface, and attractive functions such as antistatic properties and heat dissipation properties derived from carbon can be simultaneously realized without losing them. Sometimes, like the lotus leaf, the structure enhances the properties (wettability) of the material, and new functions that cannot be achieved by the material properties alone, such as the self-cleaning effect due to super-water repellency, can be created. In other words, living organisms simultaneously realize and utilize the two approaches to material design mentioned at the beginning (① material properties and ② structure) by using the surface microstructure. This can be said to be a great advantage of the surface microstructure. Behind such multifunctionality of biological surfaces lies a design concept that can be thought of as "energy-saving design," in which one structure fulfills two or more functions without losing the properties of the material. By skillfully combining materials and structures, living organisms are able to express multiple excellent functions with the smallest possible structure without compromising the properties of the materials, thereby achieving the harmony between the environment and technology required by the SDGs.

In biomimetics as described in 1-1 and 1-2, what we have learned from living organisms is mostly about functions and structures, and they have mainly been used only for human society. Even in surface microstructure biomimetics, while learning from nature, the fabrication process is often lithography and etching, which are costly and wasteful, and many of them use more energy than living organisms^{2,36-40}. This is due to the gap in technology between humans and living organisms, because humans are still inferior in the technology to fabricate complex structures at low cost to produce superior functions like living organisms^{2,36}. Even if we can precisely imitate shapes and express high functions using lithography and etching, which are superior human technologies, the gap in the fabrication process between living organisms and humans will directly lead to a gap between the research field and the industrial field. Trying too hard to get close to the structure of living organisms will actually move us away from sustainable prosperity, which is the meaning of the structure of living organisms. This is particularly noticeable when we only express functions without clarifying the principles and neglecting why the structure produces functions, and it becomes difficult to apply and becomes research for

the sake of research, which makes industrial application difficult. Although a great deal of research has been done on gecko hair structures and they have penetrated society, they have hardly been put to practical use in daily life or industry, largely due to the gap between the technological systems of these organisms and humans. In other words, no matter how clean and highly functional the adhesion using hair structures is, if there is a problem with the manufacturing method, the manufacturing cost and the actual function (benefit) obtained will not be profitable. CNT tape also has the problem that CNTs are difficult to manufacture and expensive, and that the CNTs themselves are harmful to humans. In addition, once CNTs are bonded, they collapse, which significantly reduces their adhesive strength. For these reasons, CNT tape is used as a special sample support rather than as an everyday item. In addition, problems will arise if we lean too much toward the side of organisms. If we place too much importance on the low environmental impact technological systems of organisms and forcefully push for the clean adhesion of geckos under the guise of environmental protection, consumers will have no choice but to use expensive adhesive tapes made using high-cost methods. Even if companies or governments shoulder the burden, this is merely a shifting of the problem away from fundamental solutions, and is far from a sustainable society (SDGs), which requires harmony between human society and the environment. In this case, it will only increase distrust of environmental impact. Therefore, even if we can mimic the structure of living organisms and reproduce their functions, it is not necessarily the case that we will be able to learn the meaning of living organisms' structure and apply it to human society.

1-3-4 Ecomimetics and “perspective of living things”

In contrast, even if living organisms act only for the prosperity of themselves or their own species, they end up maintaining a sustainable ecosystem. Plastic materials made by humans are poorly biodegradable and may cause marine pollution when they are released into the environment, but the metabolites of plants and animals, such as fallen leaves, shells, and feces, are known to be useful resources for other organisms. The manufacturing process, processing at room temperature and pressure and self-organization used by organisms, can produce materials or structures with low energy. This is not because organisms acted with sustainability in mind, but because their actions to reduce energy loss as a survival strategy result in sustainability. From these facts, we believe that we can learn not only about engineering design from organisms, but also about sustainable development, which is the greatest common challenge for human society. In other words, rather than only considering function and the environment in manufacturing, new material designs that take into account the impact of these resources will have a positive impact on both industry and the environment. This is not simply biomimetic function or structure, but a concept that should be called "ecosystem mimicry = ecomimetics" ⁴¹, which includes not only individual organisms but also the material circulation system that surrounds them. In addition, in recent years, the term "perspective of living things(蟲觀学)"⁴² has also emerged. This is an academic field that breaks away from the manufacturing that is only from the human perspective described above, and values the perspective of organism to solve problems. The reason we feel that the structure and function are wonderful in insects is because we look at them from a human perspective, but from the insect perspective, it is merely part of a survival strategy, and the meaning of the structure and function is a combination of many factors such as function, manufacturing cost, harmony with the environment, and hostile organisms such as predators. In other words, organisms have structures not to express functions, but to advance their survival strategies with an advantage through the functions created by the structure. Therefore, while sustainability is important, it would be counterproductive to simply express functions through mimetic structures and have problems with manufacturing costs and safety. Furthermore, since there is a large gap between the technological systems of humans and living organisms, and humans are far inferior to living organisms in terms of sustainability, manufacturing that only adopts the perspective of living organisms will not be profitable in some areas. Therefore, I believe

that what is important in “perspective of living things” is not only to have many perspectives, but also to know humans well. If humans cannot understand that they are inferior in terms of technological systems, they will only be able to imitate in a distorted way, and the problem will simply shift elsewhere. Conversely, I believe that if we can apply what we have learned from living organisms to human society by using the diversity and understanding of principles of technology, in which humans are superior to living organisms, we will be closer to realizing a sustainable society.

1-4 Purpose of this study

1-4-1 Reconciliation and progress of human society and ecosystems through understanding of nature

Based on the above background, this study aimed to conduct research that would lead to reconciliation (harmony) and progress of human society and ecosystems through understanding of nature, and conducted research on the discovery of functions of surface microstructures using organisms as information sources, elucidation of principles, and application to human society. In manufacturing, organisms have evolved to have much more information and problem-solving methods than humans, so research on biomimetics, which learns from them, is very important. In particular, in recent years, the expression of functions due to the surface microstructures of geckos and other creatures has attracted new attention due to advances in electron microscopes and nanotechnology. In addition, in the field of organism microstructures, mechanical strength has not been given much attention, and there has been little integration with adhesion and friction fields, where deformation and stress distribution caused by mechanical strength have a large impact. In addition, in previous biomimetics research, when the biological function to be imitated was not clear, the function was determined by waiting for research results on new biological functions or inspiration obtained from human knowledge. Since this is an idea based on human experience, it may differ from the actual function of organisms, and it is difficult to say that it is being used appropriately as an information source. In addition, there have been cases where research inspired only by structure and using materials different from organisms was called biomimetics. Even if the above problems were overcome and it was possible to investigate the functions of living organisms, it is possible that the multifunctionality of microstructures would go unnoticed, or that multiple functions would not be reproduced simultaneously due to distorted imitation. In addition, as mentioned above, the capacity of the Earth is reaching its limits due to human manufacturing, and it has become essential to consider sustainability in the field of engineering. Therefore, the manufacturing of highly sustainable organisms is attracting attention as a source of information, but there is still a large gap in the technological systems between organisms and human manufacturing, and especially in the field of surface microstructures, there is a large gap in the methods of microstructure fabrication. Therefore, simply faithfully imitating the structural shape of organisms may provide functionality, but there is a high possibility that the costs will be too high.

Therefore, in order to extract information that living organisms possess and utilize it to solve problems in human society, I discovered the surface microstructure of living organisms and their functions, and then used simulations to elucidate the principles and extract the elements necessary for the expression of functions. Based on the information obtained, I combined it with the wisdom cultivated by humans to create highly functional materials in human society from various perspectives such as functionality, applicability, energy cost, and low environmental impact, using the technology that living organisms possess. This research will eliminate the need to observe living organisms and discover their functions, so there is no need to wait for new research. In addition, by elucidating the principles, there will be less need to forcibly imitate them using human technology at high energy and high cost, and they can be changed to forms that are easier for humans to make, easier to apply, and less energy-intensive. It is not just a simple imitation of functions and structures for humans and researchers, but also an imitation of sustainable survival, which is the meaning of living organisms having surface microstructures. In addition, since the surface microstructure functions of living organisms are adaptations to the environment, they are expected to also have biological information. Therefore, in this study, we provided feedback to the field of biology by comparing them with other organisms based on the functions and principles obtained. If the information that living organisms possess can be used to create things, and if biological knowledge can clarify the significance of their structures to the survival of living organisms, then living organisms and the environment that surrounds them can become resources that humans should protect. In such a case, I believe that this will lead to environmental protection for the sake of living and for profit, rather than self-satisfaction or volunteer activities. This means that sustainability like that of living organisms can be applied to human society, so I believe that this type of research is extremely important.

1-4-2 Investigation of the influence of structural mechanics properties of surface microstructures on contact forces

In this study, we have taken the above viewpoints into account in our biomimetic research, and have combined the viewpoint of structural mechanics to investigate the effect of surface microstructures on contact forces such as friction and adhesion. There are not many studies on the material strength of organisms with respect to their surface microstructures, and most of the research is focused on the internal structure of materials such as turtle shells, bamboo, and bones. On the other hand, metamaterials, which are completely artificial materials, have been actively studied in recent years because the way stress is transmitted varies depending on the shape of the surface structure, which is why it is certain that the structure affects the material strength, but there has been little collaboration with the surface microstructures of organisms. In addition, many of the studies on metamaterials, turtle shells, bamboo, and bones focus on material strength. In fields other than material mechanics, especially in friction and adhesion, which are forces acting on the interface between two contacting organisms, deformation has a large effect, so changes in material strength may affect the contact force. Therefore, we have identified organisms that are likely to express contact-related functions through their surface microstructures, and research to evaluate their functions, elucidate their principles, and develop applications of friction and adhesion. In the functional evaluation, functions were investigated by measuring the surface of the living body directly as well as mimicking structures made from similar materials. In the elucidation of the principle, the parameters and shapes of the microstructure were changed and measured, and the stress was visualized and quantified by simulation to extract the elements necessary for the expression of functions. In the application, materials were created that express functions that lead to solving problems faced by humans based on the obtained principles. In this case, the materials were created using a microfabrication method that has a low environmental impact, just like living organisms. Finally, as feedback, they discovered biological insights by comparing with organisms that have a similar structure and factors such as their habitat. As models, they chose the clingfish, which has been reported to have high adhesive function and microstructure, but whose function of the microstructure is unclear, and the carrion beetle, which they newly revealed to have a hierarchical microstructure, whose function is also unclear. The effect of the microstructure of the clingfish on adhesion was investigated by creating mimicking

structures with and without microstructures and measuring the adhesive strength. After that, the principle was elucidated by simulation, and a material with similar functions to the clingfish was created using a simple method. The friction function of the carrion beetle was investigated by directly measuring the body surface, which was predicted based on the habitat and previous papers. After that, an imitation structure was created to investigate the effect of shape, and friction measurements were performed. The principle was clarified by simulation, and metal materials with friction-reducing surfaces and elastic deformation materials with structures were created by self-organization, applying the principle.

Chapter 2

**A novel adhesion mechanism
learning from the surface
microstructure of clingfish
suckers**

2-1 Introduction

2-1-1 Adhesive properties of clingfish

In addition to the gecko mentioned in 1-2-1, there are other creatures that have excellent adhesive properties due to their surface microstructures. We focused on the clingfish, a type of fish that lives in the ocean, and can adhere to rough surfaces such as rocks using the suction cups on its abdomen (Fig. 2-1 ⁴³). The adhesive strength of the clingfish has been investigated by D. Wainwright et al. ⁴⁴ and P. Ditsche et al. ⁴⁵. Both showed that the clingfish had an extremely strong adhesive strength of around 40 kPa on flat surfaces, which is 80 to 230 times its body weight. In addition, D. Wainwright et al. showed that the clingfish showed the same adhesive strength on a surface with a roughness (Ra) of 269 μm as on a flat surface ⁴⁴, and P. Ditsche et al. showed that the clingfish had the same adhesive strength on surfaces contaminated by bacteria, algae, and invertebrates as on uncontaminated surfaces ⁴⁵. It is believed that the high adhesive properties of the clingfish are due to the hair-like structures and mucus on the surface of the suction cups.



Fig. 2-1 A clingfish lifting a rock ⁴³

2-1-2 Surface structure and function of clingfish sucker

A field emission scanning electron microscope (FE-SEM) image of the sucker is shown in Fig. 2-2. The photograph and sample of this clingfish (*Aspasma ubauo*^{46,47}) were provided by Dr. Eri Katayama and Dr. Gento Shinohara of the National Museum of Nature and Science (Tokyo). Clingfish (*Aspasma ubauo*) is the name of a unique species of clingfish in the clingfish genus. What was previously considered to be *Aspasma minima* was recognized as a new species called *Aspasma ubauo* in 2019 by Kyoji Fujiwara & Hiroyuki Motomura⁴⁶. *Aspasma ubauo* lives in the temperate waters of Japan, and no specimens have been confirmed anywhere else⁴⁶. The clingfish investigated by D. Wainwright and P. Ditsche is a species called *Gobiesox maeandricus*, which lives on the Pacific coast of North America from Baja California north to Revillagigedo Island Alaska, so the species and habitat are different^{44,45}. Although they are both in the same family, Gobiesocidae, the genera *Gobiesox* and *Aspasma* are separated. As a supplementary note, although there is a high possibility that they are different because they are beetles and fish, rhinoceros beetles and sawtooth stag beetles are different families, and sawtooth stag beetles and Miyama stag beetles are in the same family but different genera. Japanese sawtooth stag beetles and Giraffa sawtooth stag beetles are in the same genus but different species. Therefore, in the broad sense, both are included in clingfish, but there are differences. The edges of the sucker of clingfish have protrusions called papillae, and when the cross section of the papillae was examined, a hair-like structure covered with mucus was confirmed (Fig. 2-2 (d, e)). According to D. Wainwright et al., in clingfish, the hair-like structure is caught on the surface of the substrate, increasing the friction between the sucker and the surface, resulting in high adhesive strength⁴⁴. In addition, it is thought that mucus also affects the increase in suction strength. The mucus that coats the clingfish body prevents the suckers from being detached because it provides adhesion through Stefan adhesion (the force required for the surrounding air or water to expel the mucus)⁴⁴.

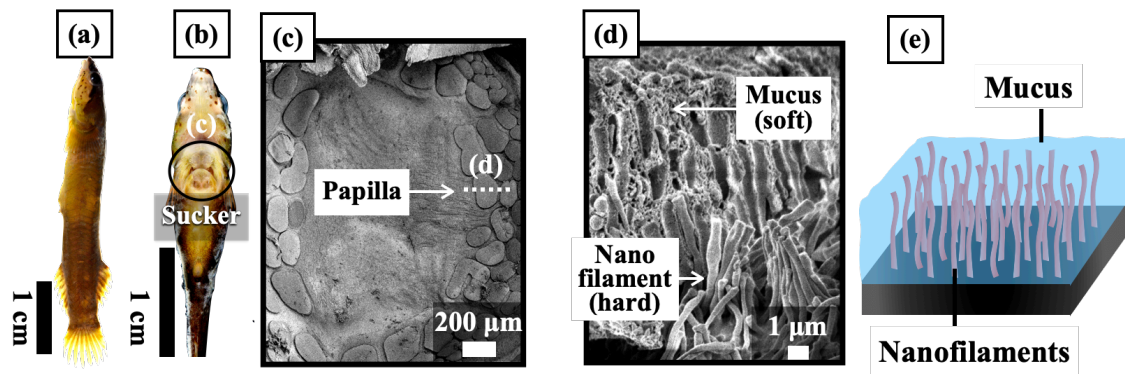


Fig. 2-2 (a, b) Photographs of clingfish (*Aspasma ubauo*), (c) FE-SEM image of sucker, (d) cross-section of papilla, (e) schematic diagram of microstructure

However, it is difficult to imagine that the hairs of clingfish generate adhesive forces, as in the gecko and other organisms with hair-like structures mentioned above. The hairs of clingfish are shorter than those of other organisms (gecko: 30-130 μm ⁵, clingfish: about 5 μm), and they lack the spatula structure (Fig. 2-2 (d)), which is said to be necessary for high adhesive forces from hair structures alone¹⁴. In addition, adhesive forces are generated when the hairs and the adhesive surface come close to each other at the molecular level, but adhesion is generally difficult in the presence of water or mucus, because water or mucus penetrates between the adhesive surface and the adhesive surface, inhibiting the generation of adhesive forces⁴⁴. Therefore, we speculated that the hairs of clingfish may have a different function from those of other organisms.

2-1-3 Basic principles of adhesion

Adhesion is a force generated by the contacting interfaces between two objects, and adhesive materials are used in many fields, including daily necessities, medicine, and architecture. Adhesive materials are also frequently needed underwater in many fields^{48–52}, including surgery, offshore work, and daily necessities. However, underwater adhesion is more difficult than on land because water molecules penetrate between the interfaces and form a hydration layer, making it difficult for adhesive forces to form between the interfaces. In recent years, the development of underwater adhesive materials based on the morphological characteristics and adhesive mechanisms of aquatic organisms, such as sessile marine organisms⁵³ and octopus⁵⁴, has attracted attention, and the creation of biomimetic adhesive materials has been reported^{49,55}. According to the basic principles of adhesion^{56–60}, strong adhesion requires the compatibility of two opposing properties: softness (increased contact area, adhesion) and hardness (resistance to peeling, cohesion). As shown in Fig. 2-3, soft materials can deform to fit the unevenness of the other material, increasing the contact area, which generates adhesive forces such as van der Waals forces at the interface. However, soft materials have low resistance to deformation stress, so when an external force is applied to peel them off, they easily deform and peel off from their edges. As a result, the overall adhesive strength is low. On the other hand, hard materials have high resistance to deformation stress, so they are difficult to peel off. However, since they cannot deform themselves, the contact area is small to begin with, and almost no adhesive strength is generated.

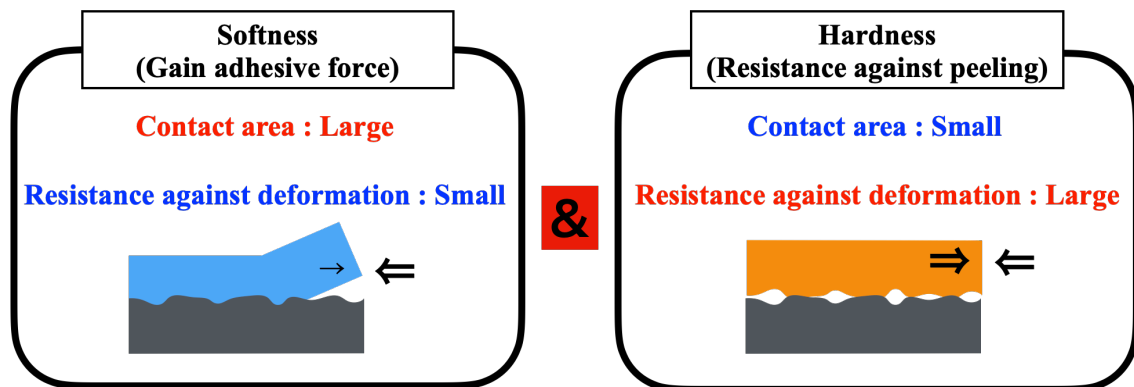


Fig. 2-3 Schematic diagram of the basic principle of adhesion

These adhesion principles are expressed by A. J. Crosby et al. as follows^{59,60}

$$F_c \sim \sqrt{G_c} \sqrt{\frac{A}{C}} \quad \cdot \cdot \cdot (1)$$

where F_c is the force capacity (maximum sustainable force), G_c is the critical strain energy release rate, A is the contact area, and C is compliance (the inverse of stiffness). According to the formula, a large contact area A and a small compliance C (increased stiffness) are required for strong adhesion.

Conventional adhesives (glue) solve the contradiction of being both soft and hard by chemical reactions. They start out in a liquid state, increasing the contact area, and then harden by applying heat or over time, creating hardness while maintaining the large contact area (Fig. 2-4). Such adhesives exhibit very high adhesive strength after hardening (cyanoacrylate adhesives have a strength of about $1,000 \text{ N/cm}^2 = 10,000 \text{ kPa}^{61}$), but they cannot be softened again after hardening, making them difficult to use repeatedly. Among living creatures, barnacles^{62,63} and snails⁶¹ use a similar adhesive mechanism. The adhesive proteins and mucus they secrete harden after coming into contact with the adhesive surface, creating a strong adhesive force. In addition, barnacles apply a water-repellent substance to the adhesive surface before secreting adhesive proteins, which creates strong adhesive strength even underwater, and snails have mucus that softens with water even after adhesion (hardening) (shear modulus: 1 GPa when hardened, 100 Pa when softened), allowing for reversible adhesion, and research into mimicking adhesive materials is also being actively conducted^{20,61}. However, these adhesive materials have the weakness that they require time and energy to harden and soften. In adhesion using surface microstructures such as those of geckos, the relatively hard hair-like structure (Einh $2\text{--}4 \text{ GPa}^{14}$) that is very densely branched at about 5000 hairs/mm^2 (gecko^{5,6}) can deform in response to the unevenness of the other surface, increasing the contact area (contact division). In addition, the spatula structure achieves

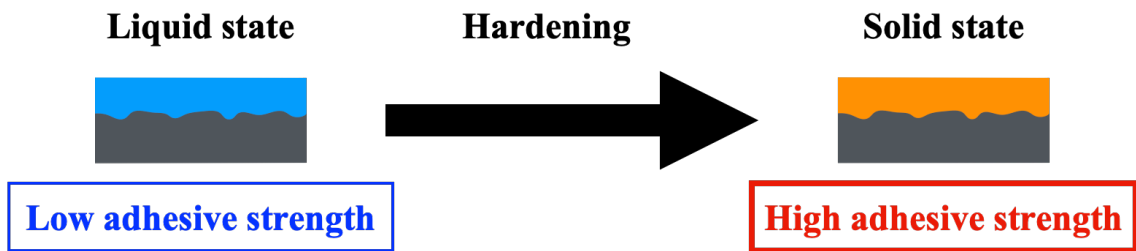


Fig. 2-4 Adhesive mechanisms of common glue

compatibility by using the difference in Young's modulus between the tip and base of the structure¹⁴. However, as mentioned in Chapter 1, it is difficult to completely mimic this complex structure, so the adhesive strength of adhesive materials with surface microstructures rarely exceeds 30 N/cm², and the adhesive strength often decreases when they come into contact with rough surfaces because the contact area cannot be secured⁶¹. In addition, the microstructure is easily contaminated and destroyed, making it difficult to use repeatedly. In addition, it is difficult to generate adhesive strength in water due to the effect of the hydration layer. Therefore, although it is possible to achieve strong adhesion like clingfish, underwater adhesion, and adhesion on rough surfaces individually, it is very difficult to achieve all of these, and it is still difficult to incorporate perspectives such as low environmental impact in the fabrication process.

2-1-4 Proposal of a new adhesion mechanism learning from the surface microstructure of clingfish

Based on the basic principle of adhesion described in 2-1-3, I thought that clingfish could achieve both softness and hardness by using soft mucus and a hard hair-like structure (Fig. 2-5). The soft mucus increases the contact area with the contact surface, generating adhesive force, and the hard hair-like structure resists the deformation stress during peeling, strengthening the adhesive force of the mucus. In other words, the deformation stress on the soft mucus is distributed to the hair-like structure, preventing peeling and generating high adhesive force. If this principle is correct, high adhesive force can be obtained without chemical reactions, time, heat, etc. Clingfish adheres by suction cups, but Stephan adhesion by mucus occurs even on uneven and dirty surfaces, unlike suction cups and hair-like structures, so the excellent adhesive force and functions described in 2-1-1 may be caused by the combination of this hair-like structure and mucus. Since this adhesive mechanism is a new adhesive mechanism different from that of geckos, barnacles, snails, etc., I used an imitation structure to evaluate its function and clarified the principle using finite element method (FEM) simulations. We then applied the principles we had discovered to create materials that could be easily applied to human society, and also provided feedback to biology by comparing them with other organisms.

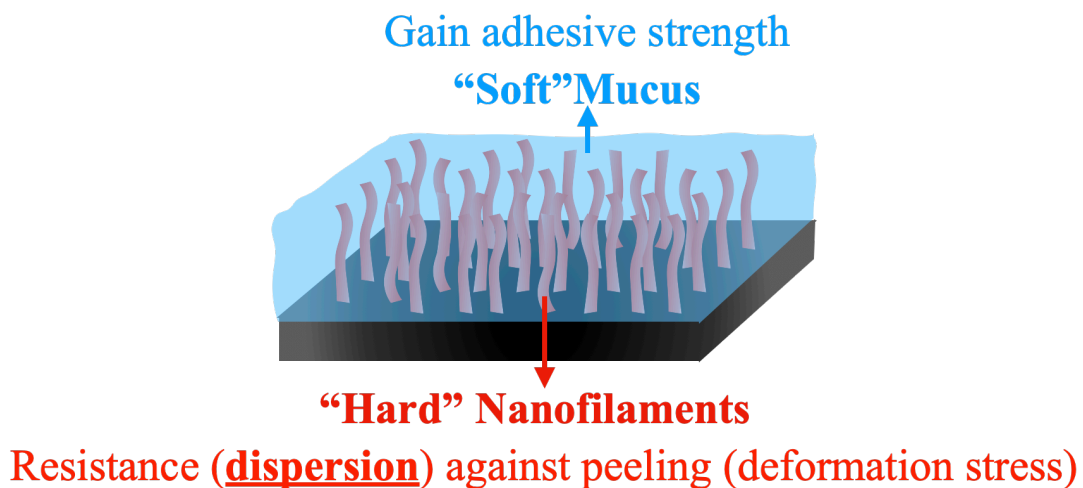


Fig. 2-5 Adhesion mechanism learning from surface microstructure of clingfish sucker

2-2 Reproducing the function

2-2-1 Mimicked structure preparation

• Nanofilament fabrication method

A PSt film was prepared by placing 50 mL of a 100 g/L polystyrene (PSt, Sigma-Aldrich Co. LLC., USA.)/chloroform solution in a 10 cm diameter petri dish and allowing it to dry naturally. An anodic alumina oxide (AAO) with vertically long pores was placed under the prepared PSt, and the AAO was pressed between a fluorine sheet (500 μm thick) and an iron plate by heat pressing at 210 $^{\circ}\text{C}$ to press the PSt into the AAO. This heat pressing was performed in two stages, first by pressing the PSt at ~ 1 MPa to soften it (annealing, 2 min), and then by applying a pressure of 2.5 MPa (3 min). After pressing, the PSt was removed and cooled by blowing cold air from a hair dryer with a 1.2 kg metal weight placed on the iron plate. After cooling, the PSt with the AAO was immersed in a 1 mol/L sodium hydroxide aqueous solution and stirred for the first time to dissolve only the AAO. The PSt was thoroughly washed with pure water and then allowed to dry naturally.

The FE-SEM image of the obtained nanofilament is shown in Fig. 2-6. Nanofilament was formed in the transferred area, and the hair length was about 30 μm and the thickness was about 200 nm.

• PDMS filling method

The nanofilament film prepared by the above method was cut to 1 x 1 cm and attached to a spin-coating platform using double-sided elastomer. After mixing the base resin of polydimethylsiloxane (PDMS, DOWSILTM, SILPOT 184 W/C, DuPont Toray Specialty Materials K.K., Japan) and the curing agent (mixing ratio 10:1), the mixture was thoroughly degassed and 250 μL was cast onto the center of the film. After waiting about 30 seconds to allow the mixture to fill the inside of the nanofilament, spin-coating was performed. Spin-coating was performed in three stages: ① 500 rpm, 10 s (to spread the PDMS evenly), ② 5,000 rpm, 30 s (to adjust the thickness), and ③ 7,500 rpm, 3 s (to blow off the PDMS that had gathered at the edge due to centrifugal force). The prepared film was thermally crosslinked by heating it in an oven at 70 $^{\circ}\text{C}$ for 12 h.

The FE-SEM image of the obtained sample is shown in Fig. 2-6. At this time, the thickness of PDMS protruding from the PSt nanofilament was 12 μm , and it was confirmed that the mimetic structure had been created because PDMS was sufficiently

filled into the nanofilament. In addition, a flat PSt/PDMS structure was created by spin-coating PDMS on the flat PSt obtained after heat pressing (Fig. 2-6). At this time, the PDMS thickness was $44.71 \pm 1.67 \mu\text{m}$ (± 1 S. D, $n = 27$) at the spin-coating speed (②) of 500 rpm and $13.34 \pm 1.39 \mu\text{m}$ (± 1 S. D, $n = 27$, Fig. 2 1) at 5,000 rpm. Since the protruding thickness of the mimetic structure was about $12 \mu\text{m}$, the spin-coating speed was set to 5,000 rpm in the future.

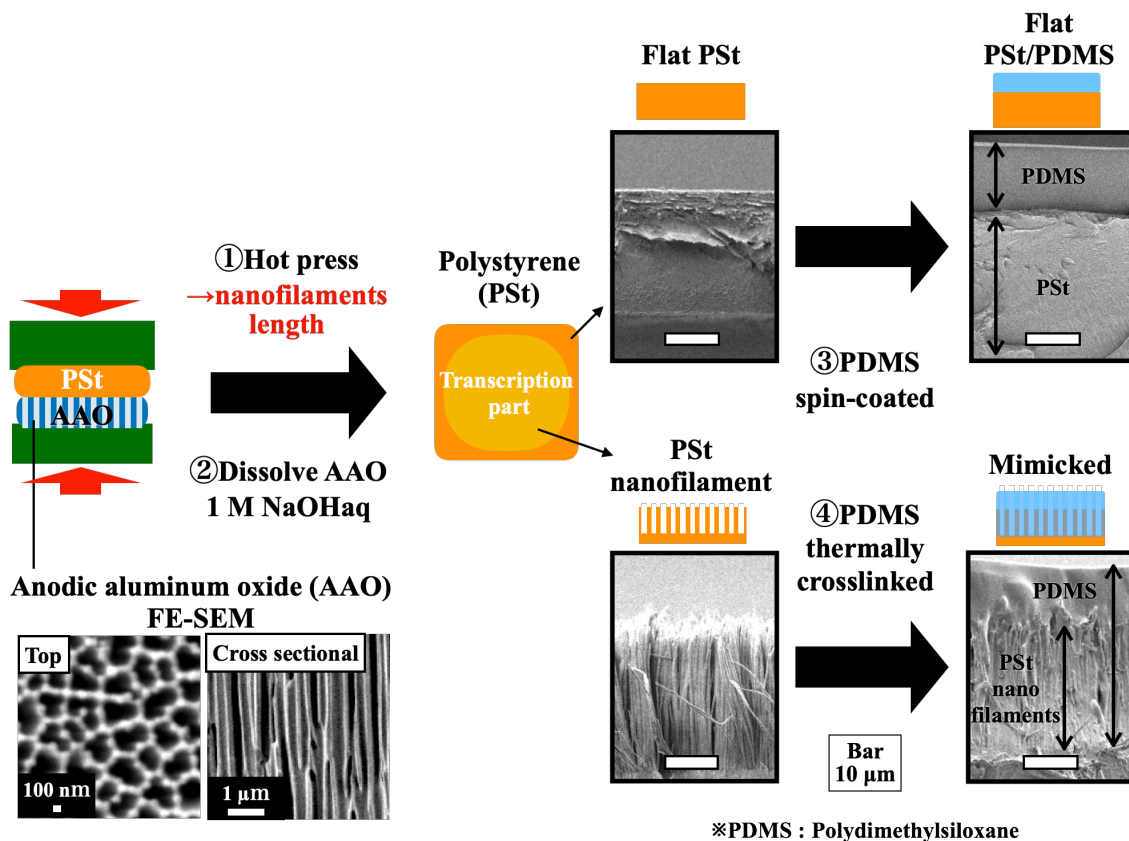


Fig. 2-6 Sample preparation procedure

<Additional Note: Determining Press Time>

The annealing and heat pressing times mentioned above were determined in the following way. Samples that were annealed (~ 1 MPa) for 2, 5, 7, 9, and 11 minutes, and samples that were annealed for 2 minutes and then heat pressed at 2.5 MPa for 3, 5, 7, and 9 minutes were observed with FE-SEM and the hair lengths were compared.

The results are shown in Fig. 2-7. It was confirmed that hair was formed even with an annealing time of 2 minutes and that the hair was sufficiently softened. In addition, although the hair did not reach 15 μm or more after 5 minutes, it was clear that even longer hair could be produced by applying pressure with a heat press. Based on these results, the annealing time was fixed at 2 minutes and the heat pressing time at 3 minutes (5 minutes in total) for the future.

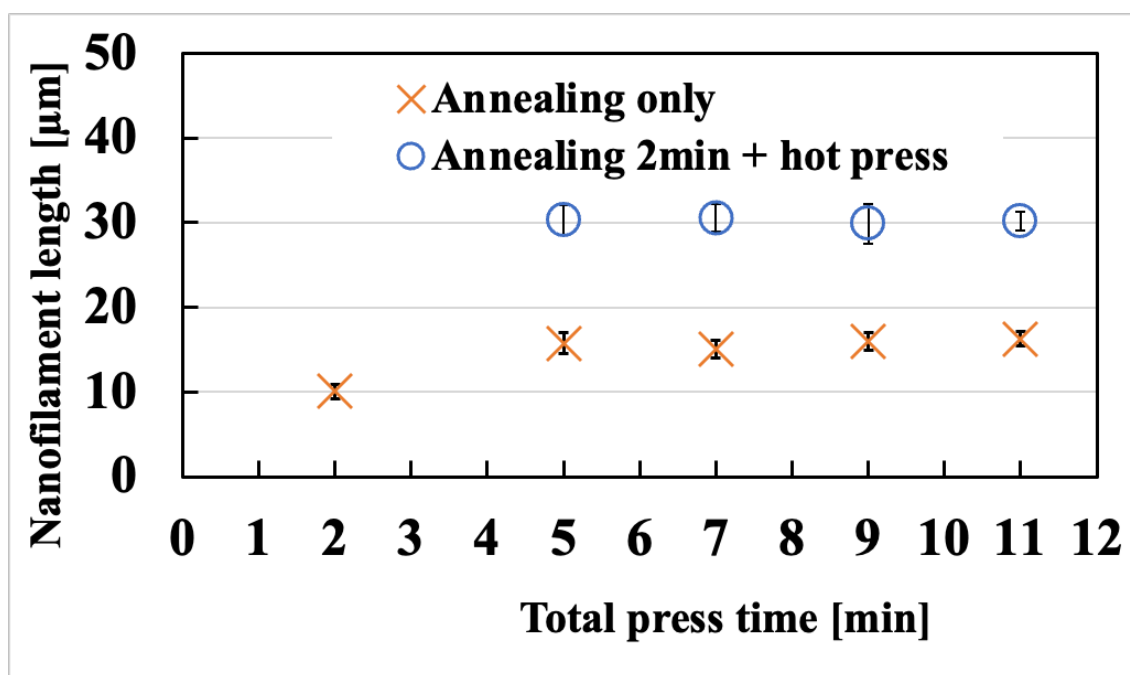


Fig. 2-7 Nanofilament length after annealing and hot pressing

2-2-2 Investigating the effect of mimicked structures on adhesive strength

• Adhesion test method

An adhesion test was conducted using the samples prepared in 2-1 ($n = 4$). The adhesion test was conducted according to the method shown in Fig. 2-8. The samples were fixed to a jig with sponge-like double-sided tape to prevent uneven contact. The samples were then pressed vertically against glass fixed with bolts and then pulled up to measure the adhesive strength between the glass and the PDMS surface. As shown in Fig. 2-8, the maximum force measured when pulled up was taken as the adhesive strength (adhesive strength [kPa]) of each sample.

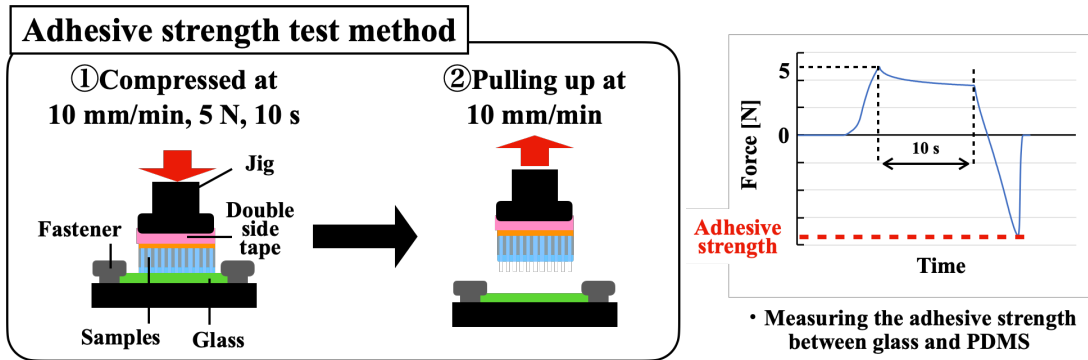


Fig. 2-8 Adhesion test schematic and force curve

<Supplement: Determining the pressing force>

In addition, the adhesive strength of the mimicked structure was measured by changing the pressing force from 1 to 6 N in 1 N increments using the method described above. The results are shown in Fig. 2-9. The adhesive strength of the mimetic structure was maximum when the pressing force was 5 N, so the pressing force was fixed at 5 N.

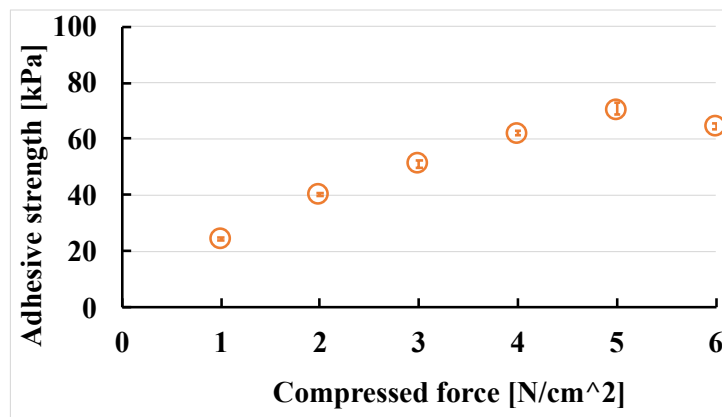


Fig. 2-9 Adhesive strength of mimicked structures when pressing force is changed

To investigate the effect of nanofilament on adhesion, adhesion measurements were performed using the three types of samples prepared in 3-1 (nanofilament only, flat PSt/PDMS, and mimicked structure, Fig. 2-6).

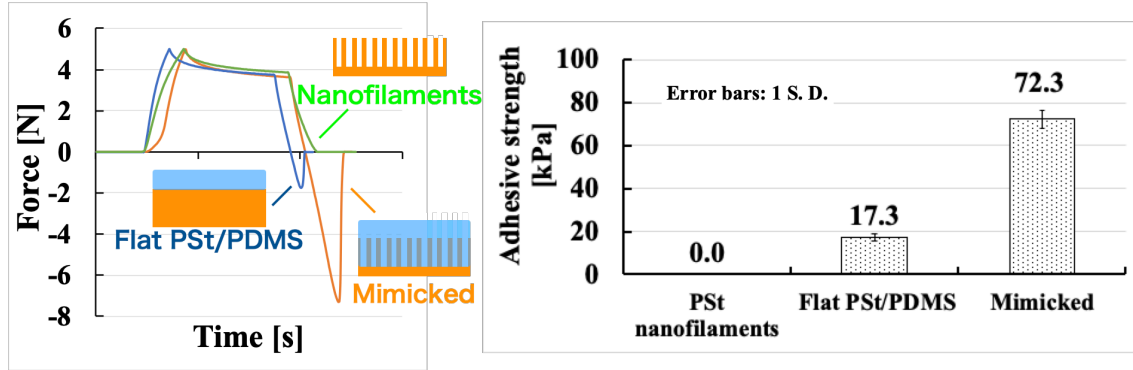


Fig. 2-10 Force curves and adhesive strength test results for each sample

The results of the adhesive strength test are shown in Fig. 2-10. Nanofilament did not produce adhesive strength. This is thought to be because nanofilament did not produce softness (increase in contact area) in a hard material such as PSt. On the other hand, flat PSt/PDMS produced adhesive strength due to the softness of PDMS, and the mimetic material showed an adhesive strength about four times stronger than that of flat PSt/PDMS. This revealed that the adhesive strength was not produced by nanofilament as in the gecko-mimetic tape, but by PDMS. In addition, nanofilament increased the adhesive strength of PDMS. The reason why nanofilament did not produce adhesive strength is thought to be that simple nanofilaments without a hierarchical structure, such as spatulas made from hard PSt, did not provide sufficient softness (increase in contact area) among the softness and hardness required for strong adhesion. In fact, many gecko-mimetic tapes are made from flexible PDMS, and many of them have hierarchical shapes. Our results show that the function of nanofilament in clingfish-mimetic adhesive materials is significantly different from that of gecko-mimetic tapes, which was previously commonly believed.

2-2-3 Adhesion of greased Nanofilament

Previously, PDMS, which can change its softness and thickness, was used as a mucus-mimicking material, but compared to the actual clingfish mucus, which is highly viscous but liquid, PDMS is solid and has different physical properties. Although PDMS is sufficient in terms of achieving both softness and hardness, in order to get closer to the actual clingfish, a mimicked structure was created using grease, a liquid lubricant that is semi-solid and semi-liquid, instead of PDMS, as a mucus-mimicking material, and adhesive strength was measured.

A sufficient amount of grease was placed on the nanofilament with a hair length of 30 μm to cover the surface, and the adhesive force was measured in the same manner as shown in 3-2. The same experiment was also performed with grease placed on a PSt flat film.

The results are shown in Fig. 2-11. At this time, unlike the experimental method shown in 3-2, the adhesive force was $44.65 \pm 11.85 \text{ kPa}$ ($\pm 1 \text{ S. D.}$, $n = 6$) when the pressing force was 5 N, whereas it was $58.28 \pm 7.25 \text{ kPa}$ ($\pm 1 \text{ S. D.}$, $n = 4$) when the pressing force was 19.5 N (the limit value of the measuring device), so the pressing force was set to 19.5 N. The adhesive force of the sample with nanofilament was higher than that of the sample without nanofilament. This is thought to be because, like PDMS, the grease and nanofilament achieve both softness and hardness. The above results demonstrate that the clingfish adhesive mechanism is valid even with semi-fluid grease. It is likely that clingfish, which have actual mucus, also use nanofilaments to enhance the adhesive strength of their mucus.

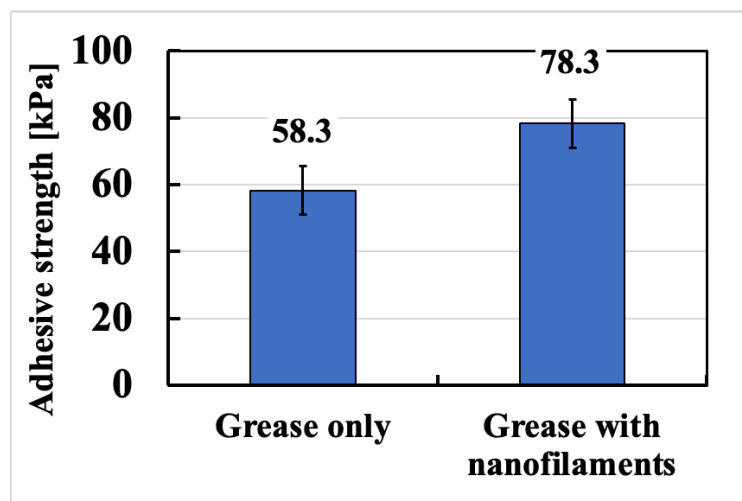


Fig. 2-11 Adhesion strength of samples made with grease

2-2-4 Underwater adhesion study

As mentioned in 2-1, adhesion is generally difficult underwater because water penetrates the contact interface. However, the PDMS used for the mucus-mimicking part in this study has a hydrophobic surface, so water molecules are completely pushed out when pressed underwater^{18,49}. Therefore, we conducted an experiment to see if it would be possible to create a clingfish-mimicking tape that can be adhered underwater by using PDMS.

A glass substrate was attached to the inside of a polystyrene case with adhesive, fixed with bolts, and then filled with water. The adhesive strength was then measured using the same method as in 2-2-1.

The measurement results are shown in Fig. 2-12. It became clear that the clingfish imitation tape is capable of exerting strong adhesive strength without impairing the underwater adhesiveness of PDMS. As the clingfish imitation tape has strong adhesive properties even underwater, it was possible to adhere and lift a 160 g beaker filled with water (clingfish imitation tape size: 1 x 1 cm).

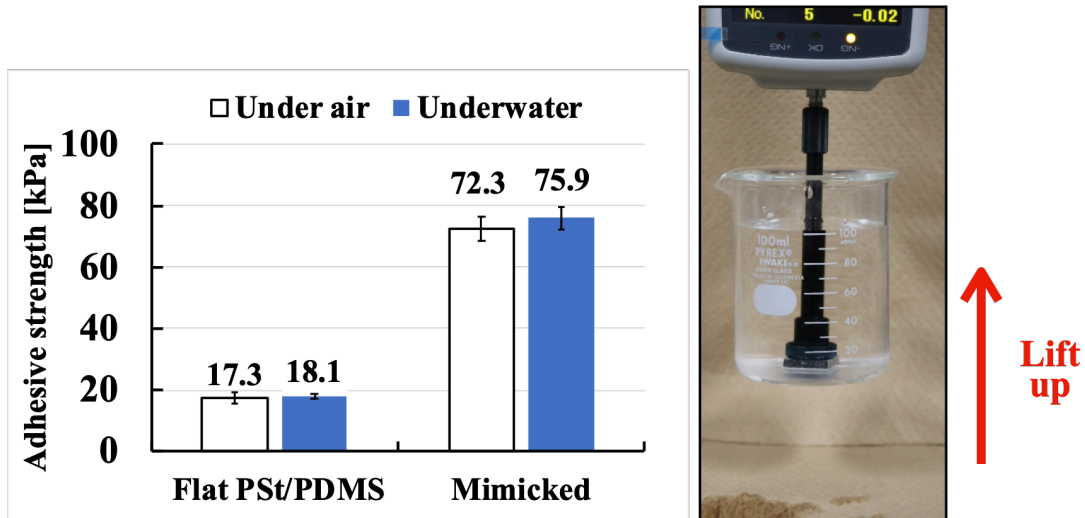


Fig. 2-12 Underwater adhesion of clingfish mimicking tape and lifting of beaker filled with water

2-2-5 Investigating adhesion to rough surfaces

Next, we verified whether adhesion to rough surfaces was possible. As mentioned in 2-1-3, adhesive materials using microstructures generally have a reduced adhesive strength when they are applied to rough surfaces. This is said to be due to a decrease in the contact area. Although the clingfish-mimicking tape we created uses a microstructure, we thought that it might be possible to adhere it to rough surfaces because the surface is covered with flexible PDMS.

Adhesion strength was measured using the method described in 2-2-1 for sandpaper and cloth of different grits (roughness) fixed in the experimental system. The arithmetic mean surface roughness (S_a) of the sandpaper was measured using a laser microscope. Sandpaper cloth was used because sandpaper was not available for grits #400 and above. The bristles were 30 μm long, the degree of cross-linking of the PDMS was 10:1, and the overhang thickness was 12 μm .

The laser microscope and adhesion test results obtained are shown in Fig. 2-13. The mimicked structure maintained high adhesion strength up to a surface roughness S_a of 5.5 μm , but the adhesion strength decreased thereafter. This is presumably due to the effect of a decrease in the contact area, so it may be possible to achieve strong adhesion to even rougher surfaces by using softer PDMS or materials. In addition, while no adhesion strength was observed at all when the surface became rough with flat PSt/PDMS, the adhesion strength of the mimicked structure decreased but did not reach 0, and adhesion itself occurred. In fact, we were able to glue and lift up even #40 sandpaper, which is so coarse that the grains of the sandpaper are clearly visible to the naked eye (Fig. 2-14). In addition, we were able to glue and lift up #400 sandpaper even underwater, which shows that it is possible to glue even rough surfaces underwater, such as clingfish. In the case of cloth sandpaper, the glue contained only in cloth sandpaper began to dissolve underwater, making it impossible to glue, so we were unable to test even rougher surfaces. From the above, it was clear that the imitation material can be glued to rough surfaces.

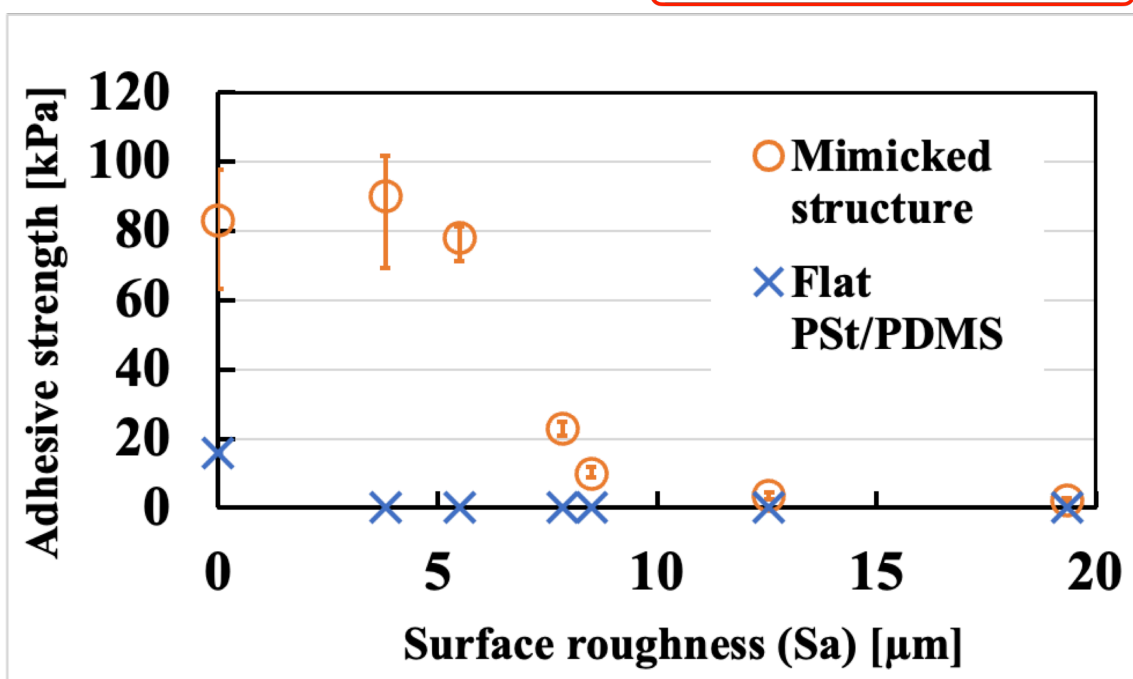
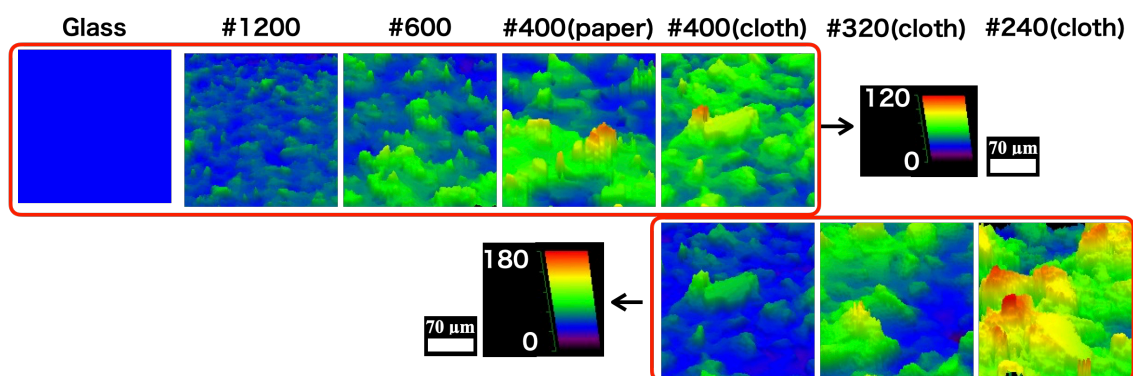


Fig. 2-13 Laser microscope images of file surfaces and their adhesive strength

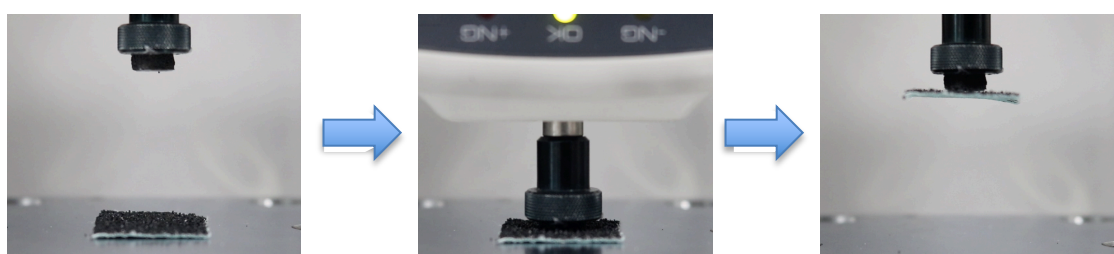


Fig. 2-14 Adhesion image of #40 grit sandpaper

2-2-6 Repeated adhesion of clingfish mimicked tape

It is said that adhesive materials inspired by the microstructure of general living organisms are difficult to adhere repeatedly. This is because the microstructure can be contaminated, peeled off, or destroyed, making it impossible to achieve both softness and hardness, which are the basic principles of adhesion. Therefore, in this section, we investigated the repeated adhesion of clingfish-mimicking tape.

The change in adhesive strength was investigated when the sample was bonded to glass 20 times in succession using the method described in 2-2-1. The sample with the mimicked structure was observed by FE-SEM before and after the measurement to check for any damage to the structure. The PSt hair was 30 μm , the PDMS crosslinking degree was 10:1, and the overhang thickness was 12 μm .

The results are shown in Fig. 2-15. The adhesive strength decreased slightly after the first three times, but there was no change thereafter, and a high adhesive strength was maintained even after 20 repetitions. There was no change in the structure of either the nanofilament or the PDMS, and it was confirmed that there was no residue or damage to the structure. It is thought that the nanofilament did not break because the PDMS surrounding it protected the structure. In addition, general adhesives are soft, but their hardness (adhesion strength between molecules) is low, so they are easy to tear and leave residue. The adhesive strength of the mimicked structure created this time is strengthened by the nanofilament, so there is no need to use unnecessarily soft materials to obtain strong adhesive strength. Therefore, it is thought that the PDMS, which has a certain degree of hardness, did not break. From the above results, it was revealed that the clingfish mimicked structure has repeated adhesive properties.

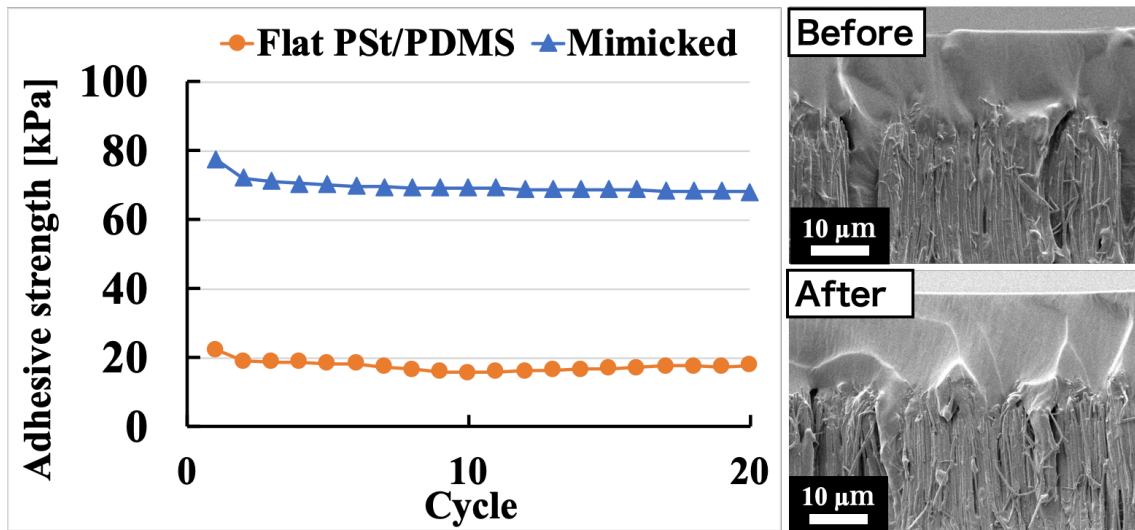


Fig. 2-15 Repeated adhesion of mimicked structure and FE-SEM images before and after measurement

2-2-7 Shortening underwater bonding time using microlens arrays (MLAs) structure

As mentioned in 2-1, adhesion is inhibited by water molecules underwater. However, there are materials that can adhere underwater, and clingfish-mimicking tape has been demonstrated to be possible. However, even materials that can adhere underwater often require time to completely expel water molecules. Therefore, we attempted to shorten the adhesion time by introducing MLAs to the mimicked PDMS surface. This is to enable water to be expelled from the interface quickly by introducing water passages on the surface, as seen in tires⁶⁴. Hexagonal microstructures found in the fingers of living organisms such as frogs play a similar role (Fig. 2-17⁶⁵), and if the grooves between the microstructures divide the surface into n contact areas (r (radius of contact area) $< R$ (radius of substrate)/ $\sqrt{n}$), the drainage time is said to be n times shorter⁶⁵. In addition, the protrusions called papillae that the clingfish possesses are arranged in a hexagonal lattice pattern to allow water to pass easily along the edge of the sucker, and their size also satisfies the above equation (sucker radius: approximately 1000–2000 μm , contact area (papilla) radius: approximately 50–200 μm , number of papillae (n): approximately 50–100), so they may play a similar role (Fig. 2-2 (c)).

First, PSt pillar structures were fabricated using the breath figure method as previously reported [62]. PDMS was then poured into the PSt pillar structures to fabricate PDMSMLAs. Ni pillar structures were fabricated by electroforming⁶⁶ using the PDMSMLAs as a mold. The electroformation was performed by Nosaka, who was a master's student in the Shimomura Hirai Laboratory at the Chitose Institute of Science and Technology at the time. The Ni pillar structures were treated with ozone for 15 minutes to improve the releasability from PDMS, and then silane-coupled with FAS17 in the same manner as in 2-1-2. FE-SEM images of the fabricated Ni pillar structures are shown in Fig. 2-16. It was confirmed that pillar structures were formed by electroforming. After that, PDMS was spin-coated onto the PSt nanofilament, and the PDMS surface was placed on the Ni pillar structures and thermally crosslinked to transfer the structure. The mimicked structure was fabricated under the conditions shown in 3-1 (nanofilament 30 μm , PDMS cross-linking degree 10:1, overhang thickness 12 μm). The mimicked structure with the fabricated MLAs structure and the mimicked structure with a normal flat surface fabricated by the method shown in 3-2 were subjected to adhesion tests in water by changing the pressing time.

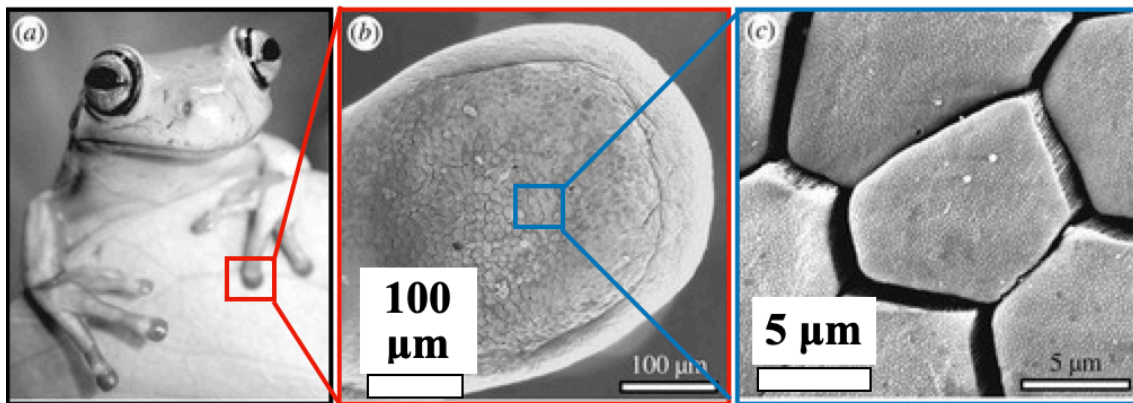


Fig. 2-17 A photo of a frog and the microstructures present in its fingers. ⁶⁵

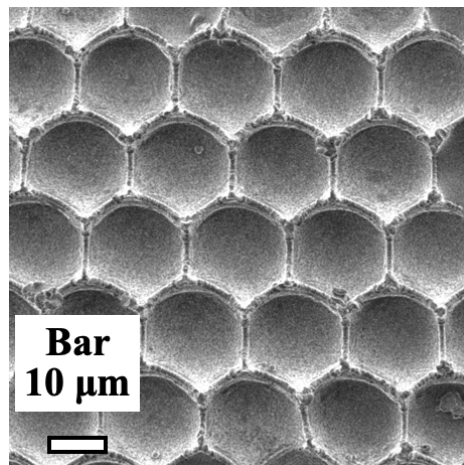


Fig. 2-16 Ni surface SEM image of Ni pillar structure

The mimicked structure that was created is shown in Fig. 2-18. An MLAs structure was successfully created on the surface. The results of an adhesion test are shown in Fig. Fig. 2-18 (b). With the mimicked structure, which has a flat surface, the adhesive strength decreased if the structure was not pressed for more than 10 seconds, but with the MLAs structure, quick adhesion was possible even with a pressing time of 0 seconds. This is thought to be because the MLAs structure quickly expelled water. However, the adhesive strength itself decreased. This is thought to be because the contact area of the MLAs structure was reduced by providing a path for water to pass through. Adjusting the shape of the microstructure may enable quick adhesion while maintaining adhesive strength.

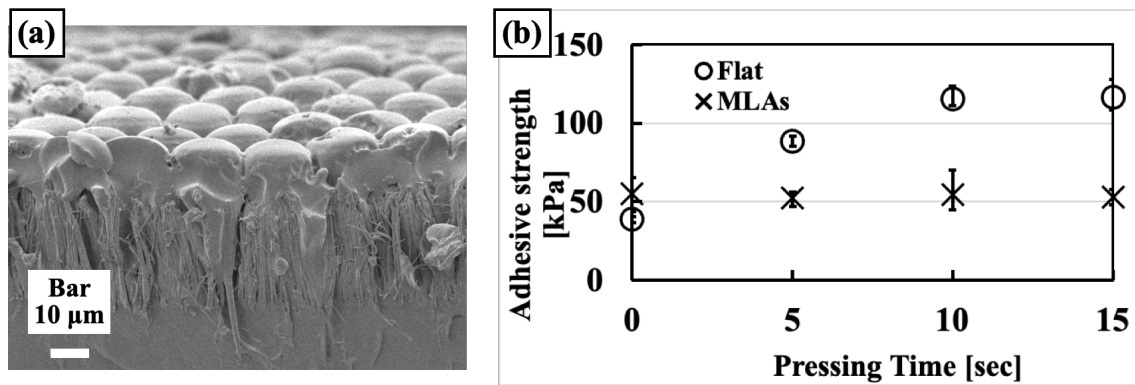


Fig. 2-18 (a) FE-SEM image of a mimicked structure with MLAs and (b) adhesion force when pressed for different times in water

2-3 Elucidation of the principle

2-3-1 Creating a simulation model

In addition to the adhesive strength test, a finite element method simulation was performed to verify the results through computational science. In a finite element method simulation, a model with any shape and material properties is created within the software, and it is possible to analyze the stress distribution, amount, and degree of deformation when any force is applied, which is difficult to visualize and quantify through actual measurement. In this case, the structure is divided into multiple finite elements (meshes) and a numerical analysis is performed, i.e., the shape is divided so that it can be analyzed, hence the name finite element method.

The finite element method simulation method is described below. The simulation was performed using a simulation software called COMSOL Multiphysics 5.6. First, a model was created within the geometry of the model builder. The model was made to be the same as the sample created in 3-1, with flat PSt/PDMS (PSt: $X \ Y \ Z = 10 \times 10 \times 40 \ \mu\text{m}$, PDMS: $10 \times 10 \times 10 \ \mu\text{m}$) and mimicked structure (PSt: $10 \times 10 \times 10 \ \mu\text{m}$, Nanofilament: hair thickness 200 nm, hair length 30 μm , center distance between hairs 1 μm , PDMS: $10 \times 10 \times 40 \ \mu\text{m}$). Glass ($20 \times 20 \times 5 \ \mu\text{m}$) was attached to each PDMS surface (Fig. 2 19(a, b)). At this time, each geometry was integrated. The material properties corresponding to each material were set from the material library for the created model. The density of PSt was 1.06 kg/m³, Young's modulus was 1 GPa, and Young's modulus of PDMS was 3.996 kPa. The Young's modulus of PDMS was determined by measuring the elastic modulus with AFM (described in 3-2-5). The Poisson's ratio of PDMS is 0.5, but since this would cause an error (denominator would be 0), it was calculated as 0.49. The mesh size of the model was set to the COMSOL-specified sizes (very coarse, even coarser, coarser, coarse, normal, fine, finer, even finer, and extremely fine). In this case, a warning about low-quality elements occurred at "coarse" for the mimicked structure, and memory shortage occurred at "even finer." When memory shortage occurs in the simulation, the calculation may not converge and erroneous results may be obtained. The same is true if the mesh size is too large. Taking these factors into consideration, the mesh was cut at "normal" (maximum element size 5.5 μm , minimum element size 0.99 μm , maximum element growth rate 1.5, curvature factor 0.6, narrow area resolution 0.5). In addition, for flat PSt/PDMS, the mesh size was set to be "even finer" (maximum element size 16.5 μm , minimum element size 2.97 μm , maximum element growth rate 1.85, curvature factor 0.9,

narrow area resolution 0.2) to match the mimicked structure (Fig. 2-19(c, d)). The measurements were performed in a steady state, and the deformation stress was calculated when the top of the PSt was pulled with a force of 100 kPa while the bottom of the glass was fixed and restrained.

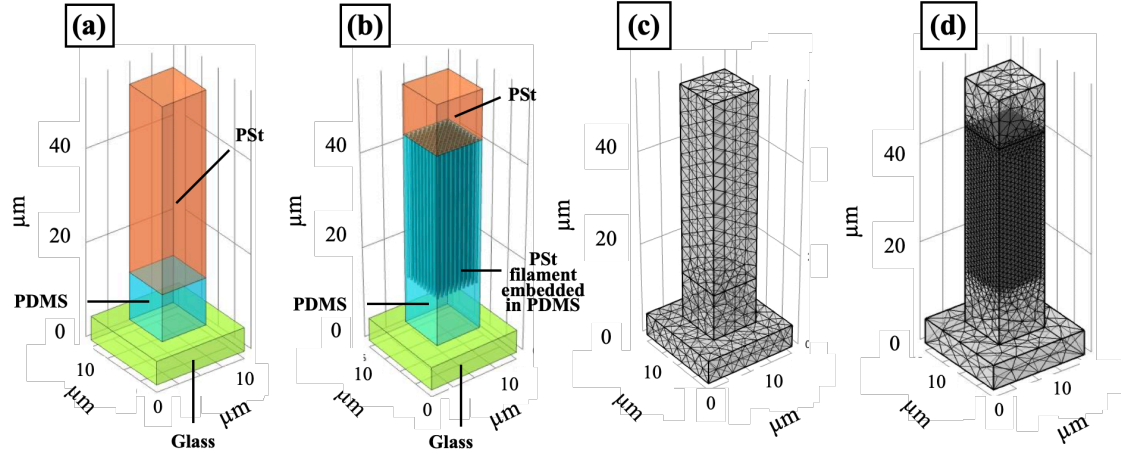


Fig. 2-19 (a, b) Model images and (c, d) mesh images of (a, c) flat PSt/PDMS and (b, d) mimicked structures.

The simulation results are shown in Fig. 2-20 (a, b). In the case of flat PSt/PDMS, a large deformation stress was concentrated at the adhesive interface, but no stress concentration was observed in the mimicked structure (red arrow in the figure). Generally, peeling occurs from the edge, so this indicates that flat PSt/PDMS is in a state where it is easier to peel than the mimicked structure. It is thought that no stress concentration was observed in the mimicked structure because the deformation stress was dispersed to the internal nanofilament. This result was not only observed at the interface between the glass and PDMS, but also at the interface between the upper PSt and PDMS. In addition, the calculation results for each time when the bottom surface of the glass was fixed and the upper part of the PSt was pulled at a speed of 1×10^{-6} m/s from 0 to 0.1 seconds are shown in Fig. 2-20 (c, d). For ease of calculation, the shape of the nanofilament hair was square, with each side measuring $1 \mu\text{m}$, and the mesh was set to a maximum element size of $1 \mu\text{m}$, a minimum element size of $0.5 \mu\text{m}$, a maximum element growth rate of 1.6, a curvature factor of 0.7, and a narrow area resolution of 0.4. Similarly, the more the nanofilament was pulled, the more the stress was dispersed to the internal nanofilaments, and the stress on the adhesive interface was less in the mimicked structure (the force applied to the corner of the PDMS/glass interface at 100 ms was 114.15 kPa for flat PSt/PDMS, and 96.498 kPa for mimicked structure). At this time, the force was applied to the tip of the nanofilament first, and then the stress was applied to the remaining part. This is thought to be because the PDMS stretches and deforms, applying a force that deforms the nanofilament in the lateral direction, and then the force in the tensile direction was applied to the entire structure. From the above results, it became clear that the mimicked structure has a high adhesive strength because the internal nanofilaments resist the deformation stress of PDMS (stress dispersion). In other words, the nanofilament increases the adhesive strength of PDMS by dispersing stress.

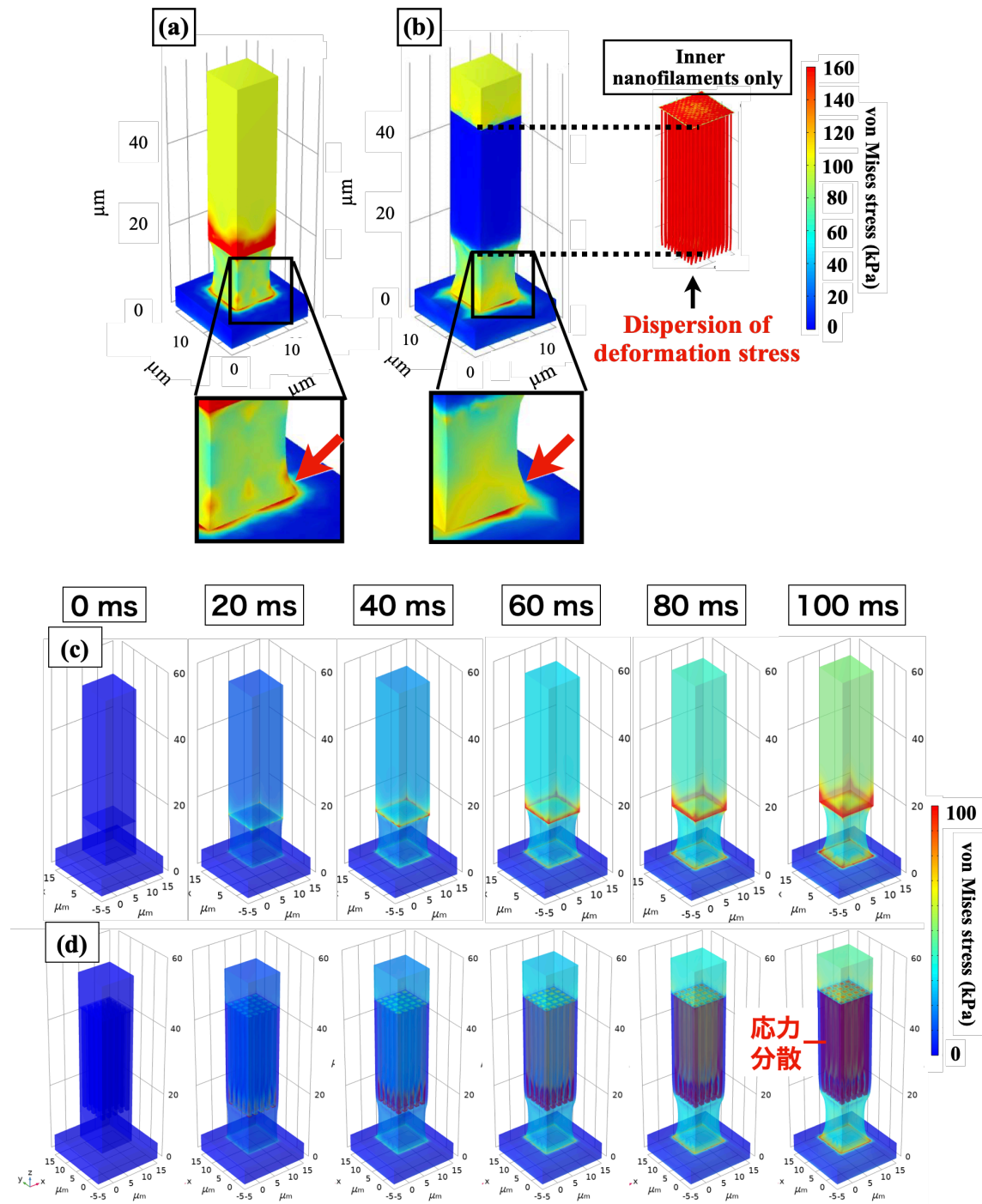


Fig. 2-20(a, c) Flat PS/PDMS and (b, d) mimicked structures. (a, b) Steady-state measurements and (c, d) time-dependent simulation results. Redder colors indicate stronger deformation stress.

2-3-2 Investigating the effect of nanofilament length on adhesion

Next, we investigated the effect of each parameter of the nanofilament and the mucus-mimicking part on the adhesive strength. In this section, we investigated the effect of the length of the nanofilament on adhesion. The length of the hair in the sample was adjusted in the following way. The PDMS overhang thickness of the mimicked structure was about 12 μm (all spin-coating rotation speeds were 5,000 rpm). In addition, we performed finite element simulations by changing the length of the hair.

• Nanofilament の長さ調整

We tried to adjust the length of the hair by changing the temperature of the heat press. We changed the temperature of the heat press from 90°C to 240°C in 30°C increments, and created nanofilaments at each temperature.

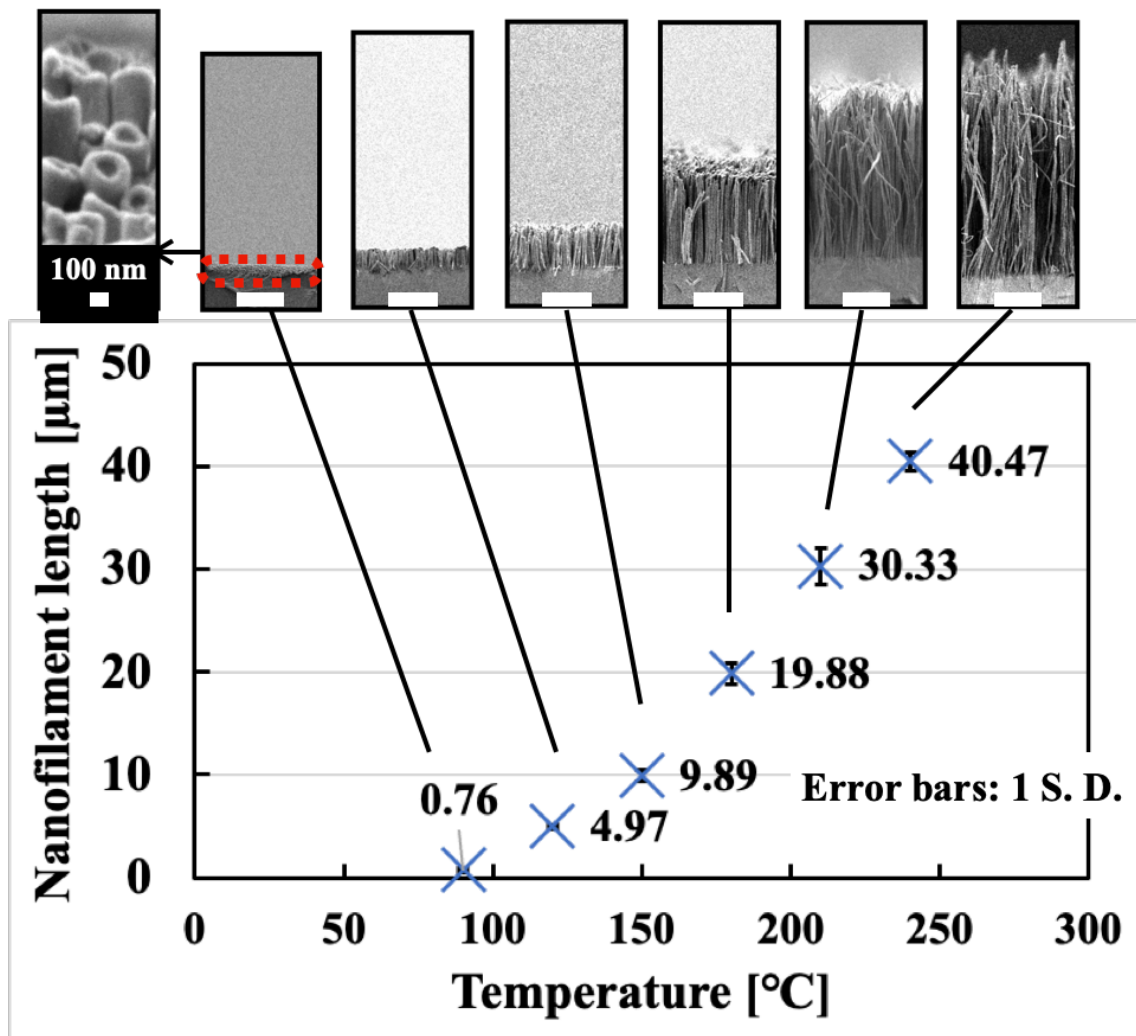


Fig. 2-21 Nanofilaments length at each temperature (n = 9) and cross-sectional FE-SEM images of each. Scale bar: 10 μm

The results are shown in Fig. 2-21. At 90°C, only a few hundred nm in length was formed, but as the temperature was increased, the length of the hair increased proportionally. The length of the hair varied by a few microns. In addition, since the thickness of the AAO used as the mold was about 50 μm , when the hair was made thicker than 50 μm , the hairs bonded at the tip. These results demonstrated that the length of the hair can be freely adjusted by changing the temperature of the heat press.

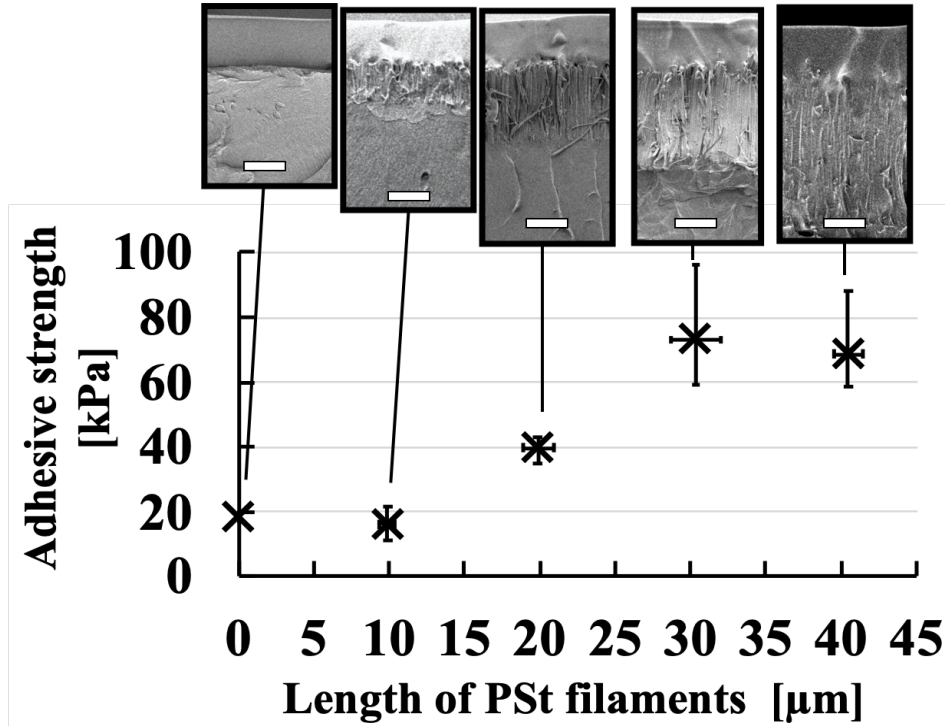


Fig. 2-22 Adhesion test results and cross-sectional FE-SEM images when varying the length of nanofilaments, Scale bar: 10 μm

The fabricated sample and FE-SEM images are shown in Fig. 2-22. The adhesive strength increased in proportion to the length of the hair, and did not change significantly beyond the hair length of 30 μm . The increase in adhesive strength is thought to be due to the fact that the contact interface between PDMS and the nanofilament increases with increasing hair length, allowing more stress to be dispersed.

The simulation results are shown in Fig. 2-23. The deformation stress was distributed to the nanofilament in the same way regardless of the hair length. This indicates that the amount of stress distributed to the nanofilament increased as the hair became longer, and it is thought that this increase in stress dispersion increased the adhesive strength. This effect of increasing the amount of stress dispersion was also seen in the PSt part, where the stress concentration decreased as the hair became longer, and

was hardly observed after 30 μm , where the adhesive strength was at its maximum (red arrow). In addition, the deformation stress was strongly concentrated at the tip of the hair. It is thought that this is related to the fact that the increase in adhesive strength plateaued at 30 μm . In general, the longer the nanofilament, the softer the tip behaves, so if the hair is too long, the amount of deformation stress dispersion decreases, and the adhesive strength does not increase. These results support the idea that the presence of nanofilament is responsible for the increase in adhesive strength.

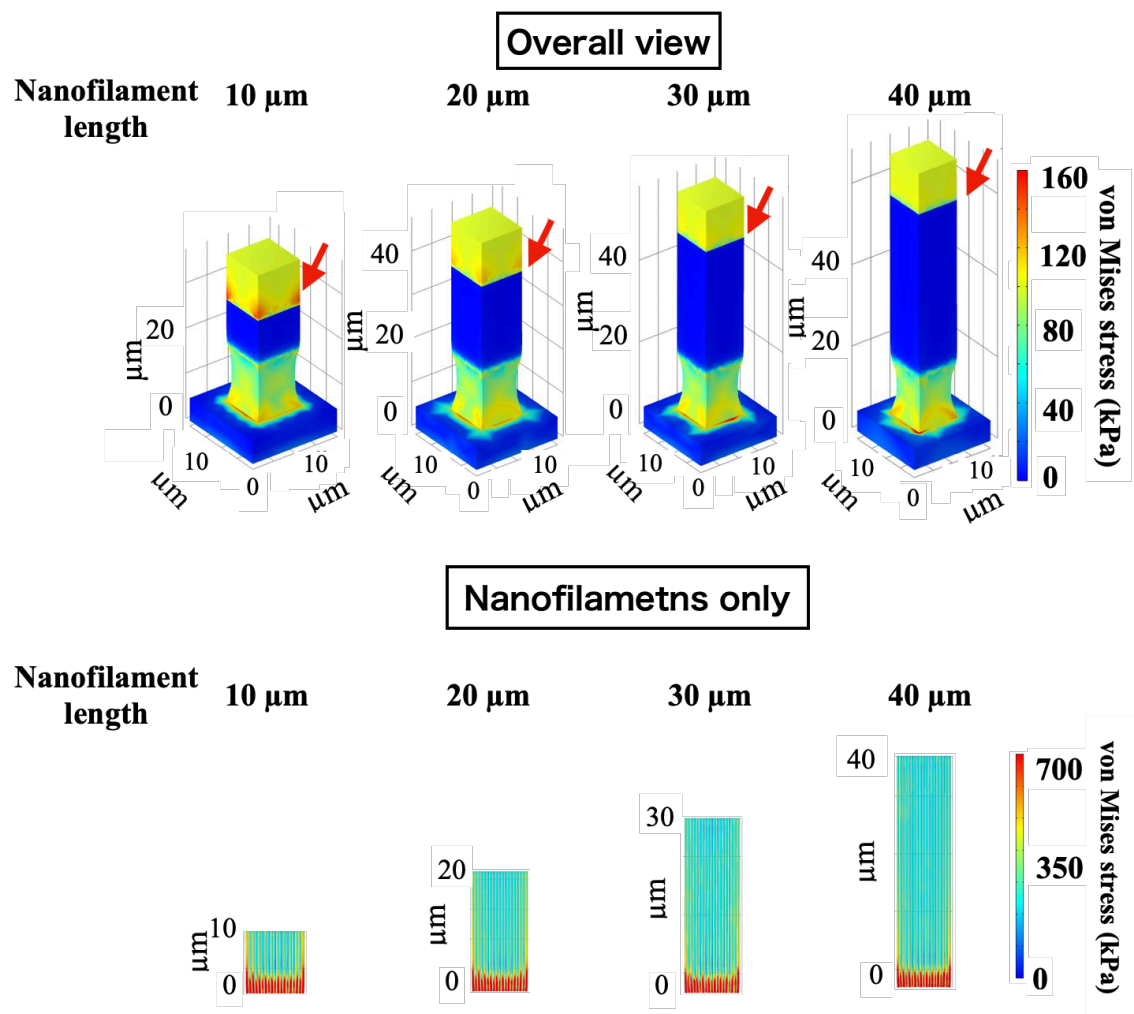


Fig. 2-23 Simulation results of mimicked structure when nanofilaments length is changed

2-3-3 Investigating the effect of nanofilament hardness on adhesive strength

The above results suggest that the stress dispersion to the nanofilament is responsible for the increase in adhesive strength. Since the effect of stress dispersion also affects material strength, in this section we investigated the effect of nanofilament hardness on adhesion using polyethylene (PE) of different densities. It is generally believed that the higher the density of PE, the higher the hardness (material strength such as Young's modulus). By using this, we were able to verify the effect of hardness without changing other properties.

First, nanofilaments were made from PE with different densities (PE, LDPE, MDPE, HDPE, Sigma-Aldrich Co. LLC., USA., in ascending order of density). PE was pressed at 60, 70, 80, 90, 120°C, LDPE at 120, 150, 180, 210°C, MDPE at 80, 90, 95, 100, 105, 110, 120°C, and HDPE at 105, 120, 135, 145, 150, 180, 210, 240°C. The length of the fibers was adjusted by changing the heat press temperature (Fig. 2-24). As with PSt, the length of the fibers could be adjusted by adjusting the heat press temperature. After nanofilament fabrication, mimicked structures were fabricated at temperatures (PE: 80, LDPE: 210, MDPE: 100, HDPE: 120°C) at which the hairs were 30 μm thick. The PDMS overhang was adjusted to approximately 12 μm (spin coat speed 5,000 rpm). The FE-SEM image after fabrication is shown in Fig. 2-24. It was confirmed that the inside of the nanofilament was filled with PDMS, creating a mimicked structure. These samples were used for the adhesion test.

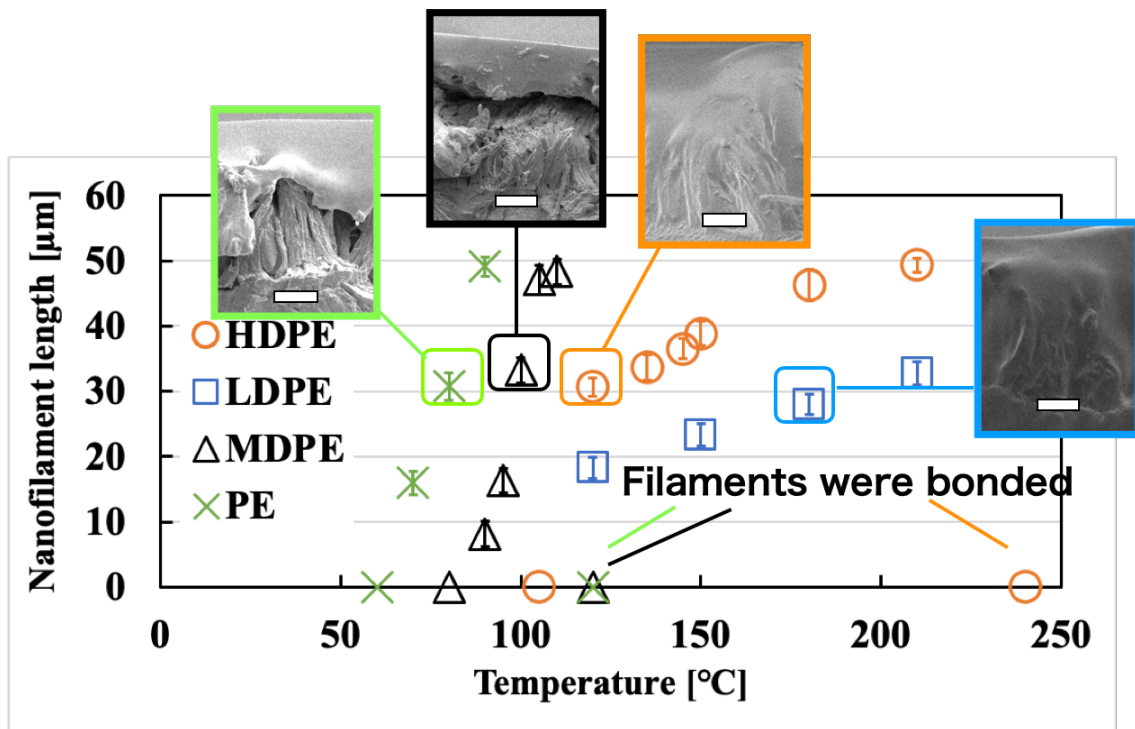


Fig. 2-24 Nanofilament length at different temperatures for PE with different densities and cross-sectional FE-SEM images of the mimicked structure (nanofilament length 30 μm), scale bar: 10 μm

The results of an adhesion test using the mimicked structure we created are shown in Fig. 2-25. The adhesion strength increased in proportion to the density of the hairs, i.e., their hardness. The results of a simulation are shown in Fig. 2-26. In this simulation, the material of the hairs was changed from PSt to PE, and the hardness of the hairs was changed by changing the Young's modulus to 0.1, 0.5, and 1 GPa. As a result, it was revealed that the range of deformation stress dispersion in the nanofilament increased in proportion to the Young's modulus, and the amount of stress dispersion increased by making the nanofilaments harder, thereby increasing the adhesion strength.

These results support the idea that the stiffness imparted by nanofilaments increases adhesive strength. They also show that adhesive strength can be controlled by changing the degree of stress dispersion through the stiffness of the nanofilaments.

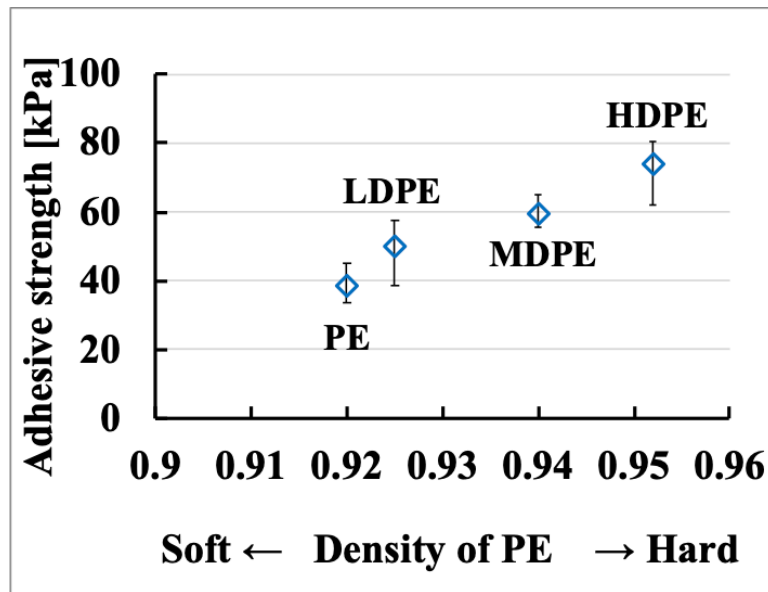


Fig. 2-25 Change in adhesive strength when changing the density (hardness) of nanofilaments

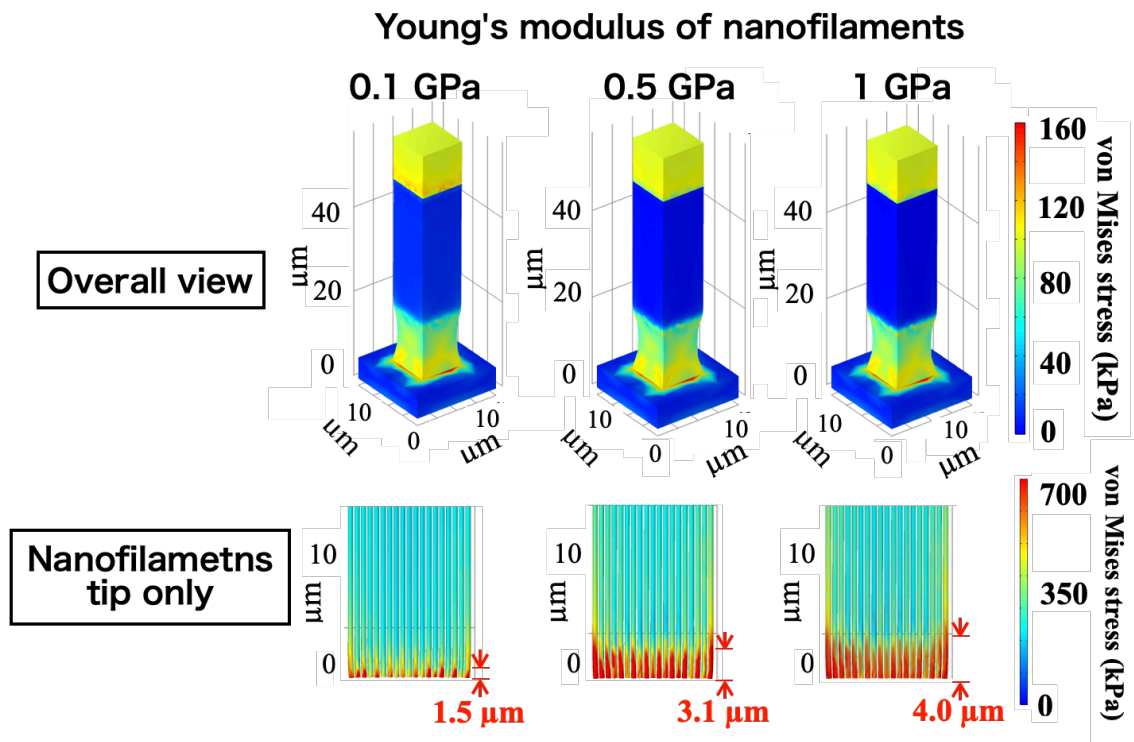


Fig. 2-26 Simulation results when changing the stiffness of the nanofilament

2-3-4 Investigating the effect of PDMS overhang thickness on adhesive strength

The above study clarified the effect of nanofilament hardness on adhesive strength, so we next investigated the effect of the mucus-mimicking part. According to the clingfish adhesion mechanism, the role of the mucus-mimicking part is to generate adhesive strength (increase in contact area). The PDMS used in this study is harder than grease or mucus, so its thickness does not change during adhesion and remains constant. Therefore, if the thickness is too thin, the variation in the length of the nanofilament will not generate a sufficient contact area, and conversely, if it is too thick, the distance to the nanofilament will be too far, and stress will not be sufficiently distributed, so the adhesive strength may not increase. Therefore, in this section, we investigated the change in adhesive strength when the protruding thickness of the PDMS, which is the mucus-mimicking part, is changed. The protruding thickness of PDMS was adjusted by the method shown below. The nanofilament was made of PSt and its length was 30 μm . We also performed a simulation when the protruding thickness was changed. In the simulation, the protruding thickness was 1, 5, 10, and 20 μm , and the length of the PSt nanofilament was all 30 μm .

• PDMS thickness adjustment

In order to adjust the PDMS thickness of the mimicked structure, the spin coat rotation speed was changed to fabricate the mimicked structure. The spin coat rotation speed was changed in increments of 1,000 rpm in the range of 1,000 to 6,000 rpm, and the PDMS thickness was measured using FE-SEM. The thickness of the PDMS protruding from the nanofilament was taken as the PDMS thickness. The length of the nanofilament was 30 μm (heat press: 210°C).

The results are shown in Fig. 2-27. It was confirmed that the nanofilament was sufficiently filled with PDMS. The overflow thickness decreased in proportion to the rotation speed, but no difference was observed after the rotation speed of 5,000 rpm, and the thickness could not be made less than 10 μm . This is thought to be due to the interaction between PDMS and nanofilament.

Next, we attempted to adjust the thickness by mixing hexane into PDMS to reduce the viscosity. After mixing hexane into the PDMS precursor while taking care not to introduce bubbles, we cast it onto a nanofilament film. After spin-coating, the sample was thoroughly evacuated to volatilize the hexane, and thermally crosslinked. The spin-

coating rotation speeds were 3,000 rpm (hexane mixture ratio: 40 wt% only) and 5,000 rpm (hexane mixture ratios: 0, 20, 40, 60, 80 wt%).

The results are shown in Fig. 2-28. At both 3,000 rpm and 5,000 rpm, the thickness of the overhang could be reduced by mixing hexane. This demonstrated that the thickness can be adjusted by the spin-coating speed and the hexane mixing ratio.

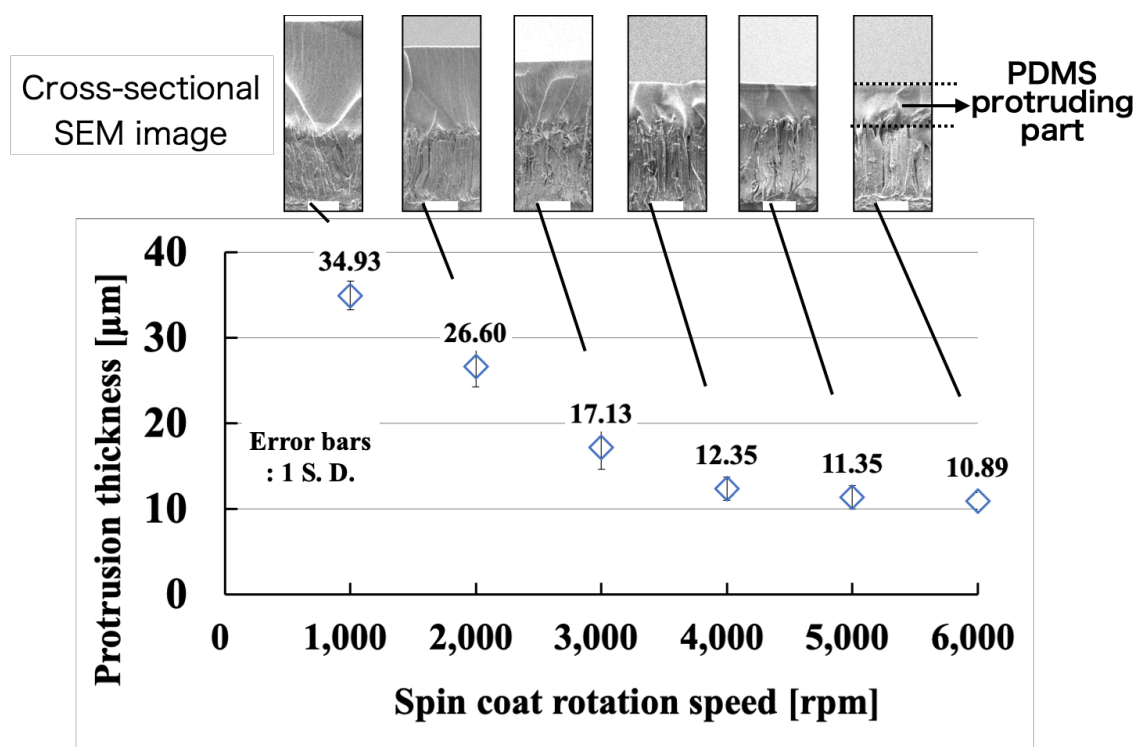


Fig. 2-27 PDMS protrusion thickness at each spin-coat rotation speed ($n = 27$) and cross-sectional FE-SEM images, Scale bar: 10 μm

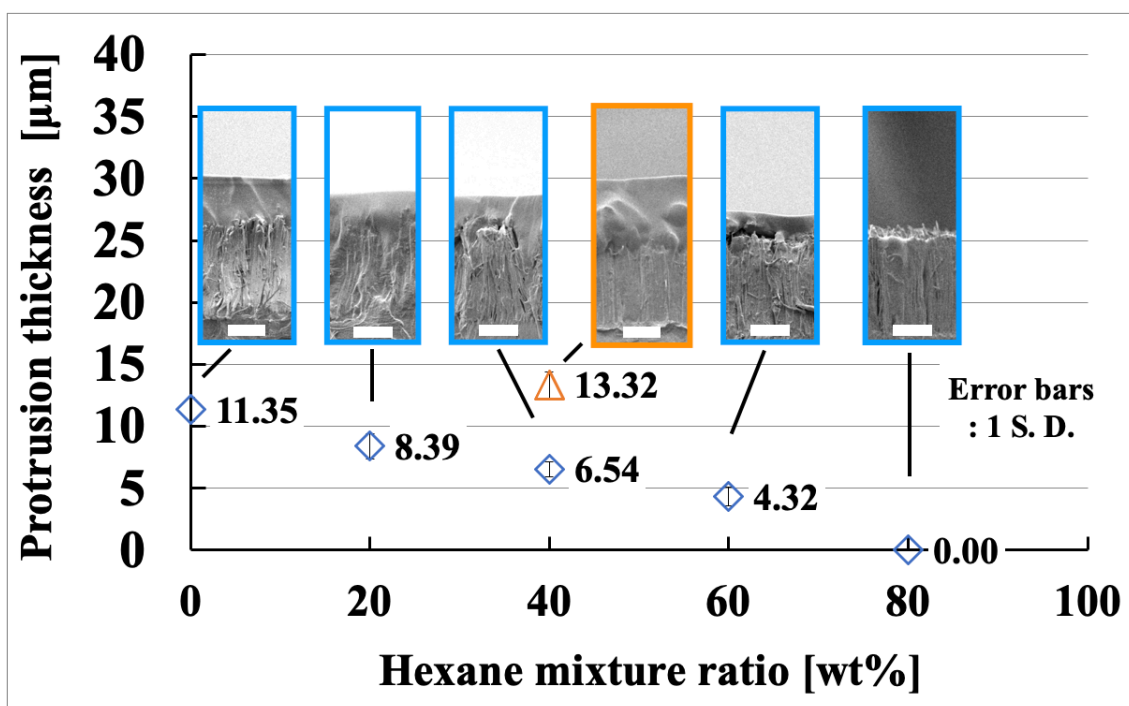


Fig. 2-28 PDMS overflow thickness for each hexane mixing ratio (n = 27) and cross-sectional FE-SEM images, blue: 5,000 rpm, orange: 3,000 rpm, scale bar: 10 μm

The results of the adhesion test are shown in Fig. 2-29. The adhesion increased to a peak at about 10-15 μm regardless of whether hexane was mixed or not. The simulation results are shown in Fig. 2-30. The deformation stress dispersion was maximum when the overhang thickness was 10 μm . From these results, it is considered that when the PDMS is too thick, the nanofilament and the adhesive interface are too far away, and the adhesion force does not increase because the stress dispersion effect is not sufficient. In addition, the tips of the nanofilaments prepared in this study are irregular and have an error of several microns (Fig. 2-21). Therefore, it is considered that when the PDMS overhang thickness is too thin, the contact area is not sufficient due to the influence of the nanofilament. To support this argument, no adhesion was generated when the overhang thickness was 0 μm . In addition, the simulation results confirmed that when the PDMS is thin, the amount of deformation is large and it is in a state where it is easy to peel off. From the above results, it became clear that there is an appropriate PDMS overhang thickness for the mimicked structure.

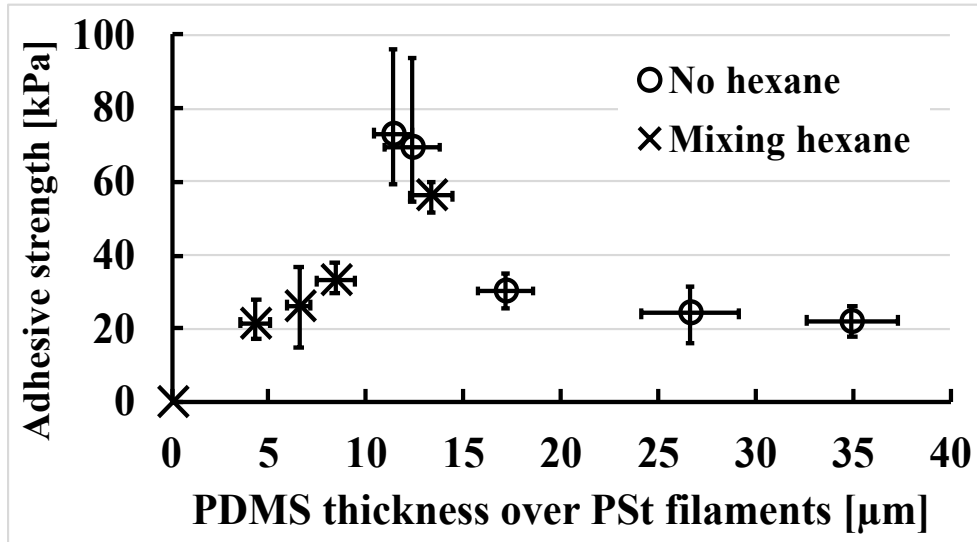


Fig. 2-29 Adhesion strength of mimicked structures for each PDMS overhang thickness

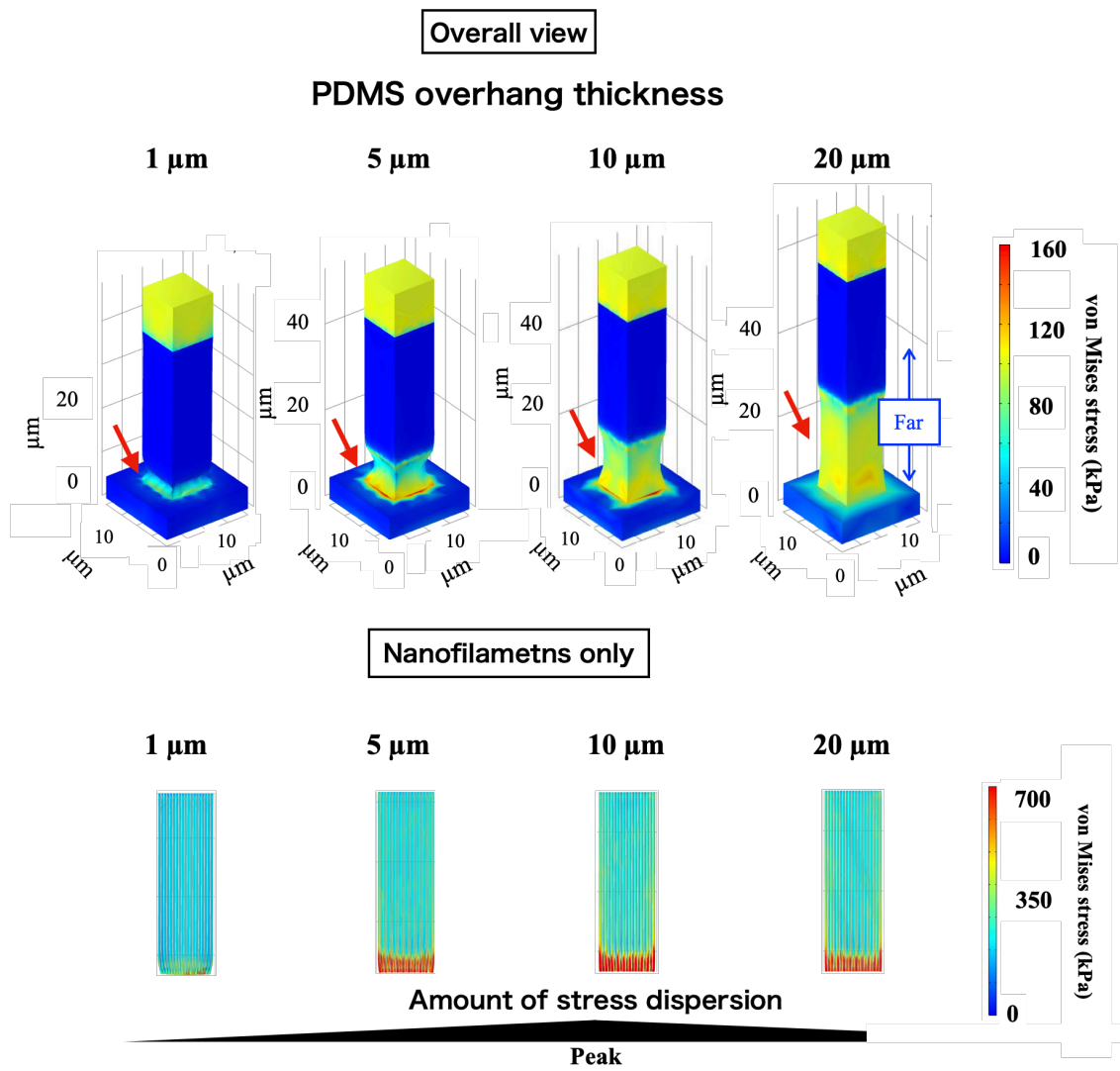


Fig. 2-30 Simulation results for each PDMS thickness

2-3-5 Investigating the effect of PDMS softness on adhesion

It is known that the role of the mucus-mimicking part is that the contact area of an object greatly affects the softness of the material. Therefore, in this section, we investigated the change in adhesive force when the softness of PDMS was changed. The softness of PDMS was adjusted by changing the mixture ratio of the base agent and the hardener. In this study, the base agent:hardener ratio was 10:1, 20:1, 30:1, and 40:1. The length of the nanofilament was fixed at 30 μm . The softness (Young's modulus) of PDMS was measured using an atomic force microscope (AFM), and simulations were performed as before.

• Measurement of PDMS softness using AFM

AFM is a microscope that can observe and analyze the surface by bringing a nanometer-sized sharp needle close to the sample and detecting the atomic force between the needle tip and the sample. A laser is applied to the back of a cantilever with a probe at the tip, called a cantilever, and the sample is scanned by moving it. At this time, the surface condition can be measured by detecting the deflection of the cantilever caused by the unevenness of the surface or attractive and repulsive forces with a photodiode.

In addition to the usual surface measurements described above, AFM can measure mechanical properties because the force acting on the tip and sample is distance-dependent. In surface topography measurements, the sample is moved horizontally relative to the cantilever, but a force curve is obtained by pressing the tip vertically into the sample. A force curve with the tip movement amount plotted on the horizontal axis is called a force-distance curve. Fig. 2-31 shows a schematic diagram of the cantilever at each pressing distance. First, as the tip approaches the sample, it receives a slight attractive force from the sample, causing the cantilever to bend downward. This is called jump-in (①). Then, as the tip is pressed into the sample, the cantilever receives a repulsive force, causing it to bend upward (②). At this time, the spring constant of the cantilever is harder than the sample, so most samples are deformed even with a small force. Considering a simple cylindrical tip with a radius of 10 nm, a force of 30 nN, which is a typical size for AFM measurements, is said to correspond to a stress of approximately 100 MPa⁶⁷. This stress exceeds the yield stress of many plastics, and even elastic samples such as rubber undergo deformations of the order of 10–100 nm⁶⁷. When the sample is then pulled away, a strong attractive force is exerted between the tip and the sample due to the adhesion forces that occur between them (③).

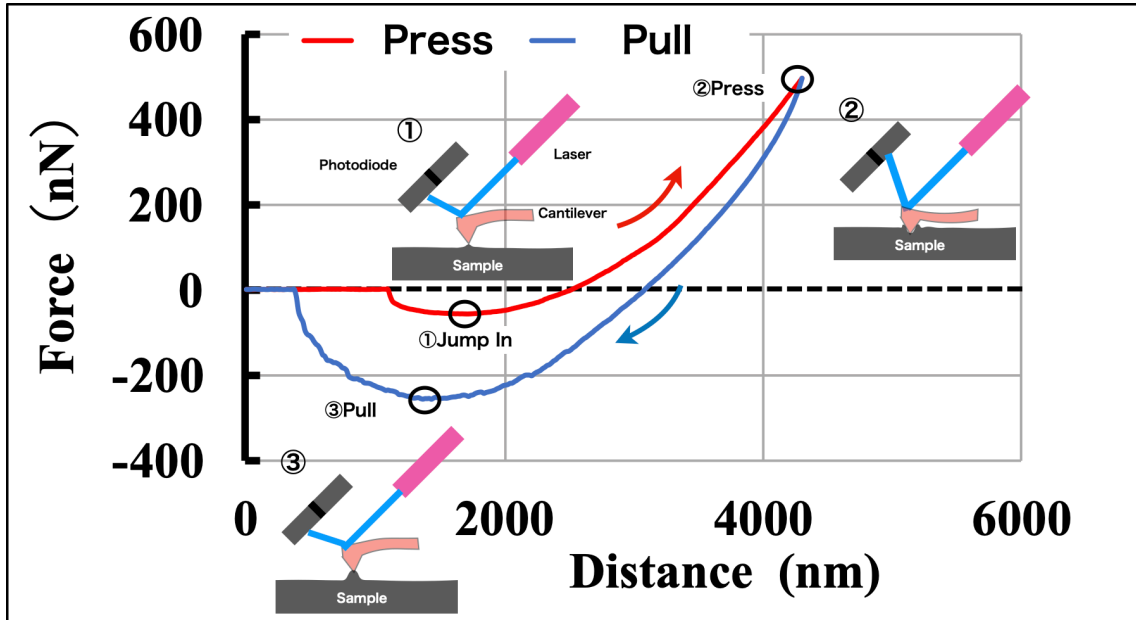


Fig. 2-31 Force-distance curve and schematic diagram of a cantilever

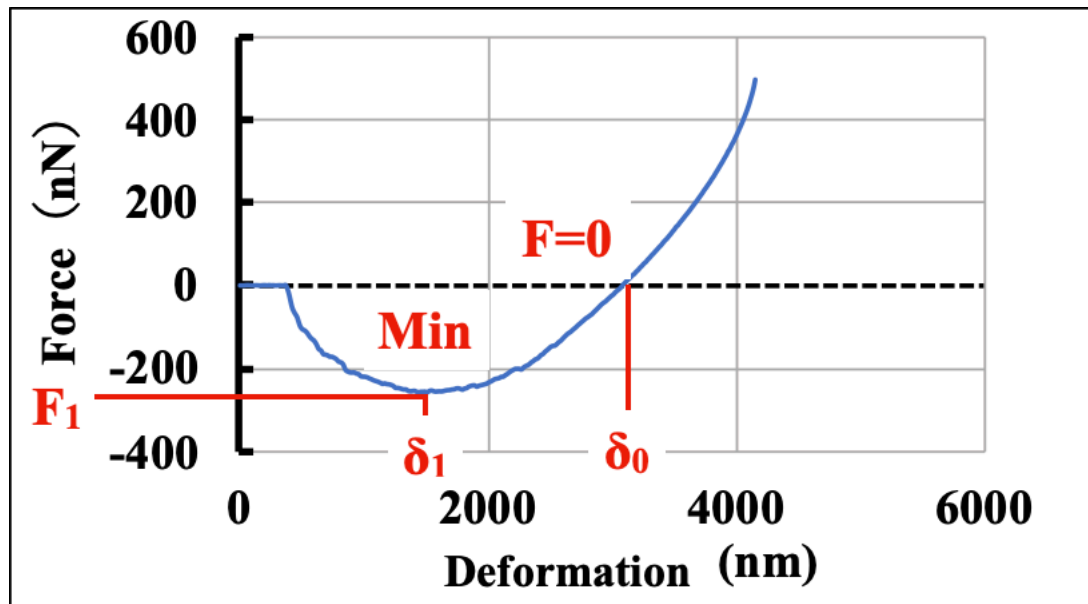


Fig. 2-32 Force-Deformation Curve, only pull-away, Fig. 2-31 transformed

The vertical axis of the force-distance curve is the force detected from the cantilever deflection (the cantilever deflection multiplied by the spring constant k), and

the distance plotted on the horizontal axis is the amount of cantilever movement. Therefore, this distance includes both the deformation of the sample and the displacement of the cantilever. Therefore, the cantilever deflection is removed using Eqs. (2, 3), and the deformation of the sample is plotted on the horizontal axis, which is called the Force-Deformation Curve (F- δ Curve) (Fig. 2-32).

$$\Lambda = F/k \quad \cdot \cdot \cdot \cdot (2)$$

$$\delta = z - \Lambda \quad \cdot \cdot \cdot \cdot (3)$$

Here, F is the force felt by the probe, k is the spring constant of the cantilever, Λ is the displacement of the cantilever, z is the distance, and δ is the deformation of the sample.

Young's modulus can be calculated as stress/strain. With AFM, the force applied to the sample can be determined from the force curve, but to determine the strain, it is necessary to know the contact area between the probe and the sample surface. However, in reality, it is nearly impossible to actually measure this. Therefore, Young's modulus is usually calculated using a theoretical calculation formula for a contact model that assumes the shape of the probe tip is a sphere. A representative contact model is the Hertz model, which only considers the repulsive force acting on a sphere and is expressed by the following formula:

$$F = K * R^{1/2} * \delta^{3/2} \quad \cdot \cdot \cdot \cdot (4)$$

where K is the elastic constant, R is the radius of the tip, and δ is the maximum deformation of the sample.

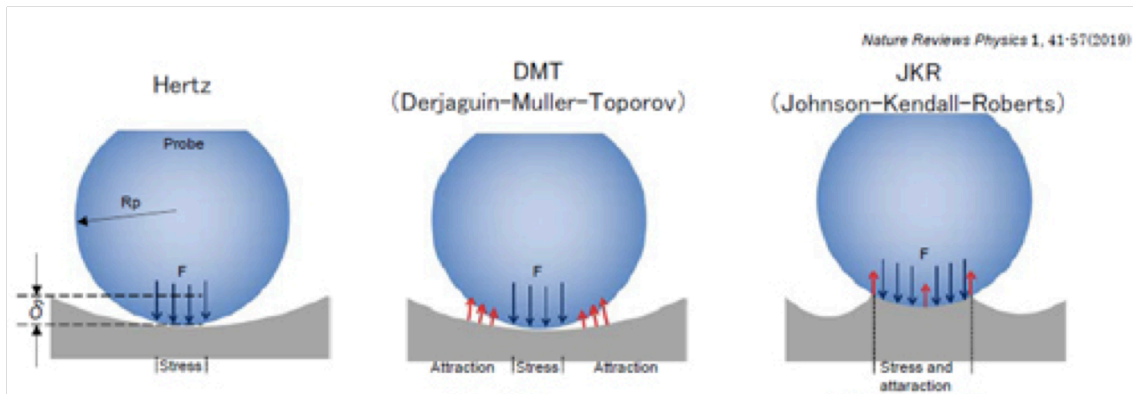


Fig. 2-33 Schematic diagram of the Hertz model, DMT model, and JKR model when the tip is indented

However, this model does not take adhesion into account, as shown in Fig. 2-33, and is therefore not applicable to samples such as PDMS, where adhesion occurs. The DMT model and JKR model are well-known contact models that incorporate the adhesion effect. The DMT model is expressed by the following equation.

$$\delta = \frac{a^2}{R}, \quad a^3 = \frac{R(F + 2\pi wR)}{K} \quad \cdot \cdot \cdot (5)$$

where a is the radius of the contact line and w is the adhesion energy.

In this equation, $2\pi wR$ represents the adhesion force. As can be seen from this equation, the DMT model does not take into account the increase in adhesion force caused by changes in the contact area when the probe is pressed in. In many cases, the DMT theory can be used for hard materials with an elastic modulus exceeding GPa, but it cannot be applied to soft materials in general that have an elastic modulus below that. Therefore, this time we applied the JKR model by Johnson, Kendall, Roberts et al., which evaluates the correct adhesion force at each stage of different contact areas. The JKR model is expressed by the following equation.

$$\delta = \frac{a^2}{R} - \frac{2}{3} \sqrt{\frac{6\pi w a}{K}}, \quad a^3 = \frac{R}{K} (F + 3\pi wR + \sqrt{6\pi wRF + (3\pi wR)^2}) \quad \cdot \cdot (6)$$

Although it is not impossible to eliminate a from equation (6) and express the relationship between F and d as an implicit function, it is not easy to fit the AFM experimental results with this function. Therefore, the Young's modulus of PDMS was calculated using the JKR 2-point method, which can be a reliable means of quantitatively evaluating the elastic modulus of soft materials with adhesion. The JKR 2-point method can calculate the elastic modulus using the following equation:◦

$$K = \left(\frac{1 + 16^{1/3}}{3}\right)^{3/2} * \frac{F_1}{\sqrt{R(\delta_0 - \delta_0)^3}} \approx 1.27 * \frac{F_1}{\sqrt{R(\delta_0 - \delta_0)^3}} \quad \cdot \cdot (7)$$

Here, the elastic modulus K is expressed by the following formula:

$$K = \left(\frac{4E}{3(1 - \nu^2)}\right) \quad \cdot \cdot (8)$$

E : Young's modulus, ν : Poisson's ratio, R : tip radius, and finally, the Young's modulus of PDMS is calculated by the following equation [69].

$$E = \left(\frac{3(1 - \nu^2)}{4} \right) * \left(\frac{1 + 16^{1/3}}{3} \right)^{3/2} * \frac{F_1}{\sqrt{R(\delta_0 - \delta_0)^3}} \quad \cdot \cdot (9)$$

As shown in Fig. 2-32, F_1 , δ_0 , and δ_1 are the force and strain at the maximum adhesion point and the equilibrium point ($F = 0$) of the F - δ curve, so the elastic modulus can be calculated from only two points. This is the JKR two-point method⁶⁸. In this case, the force is shown as a negative value on the graph, but this is irrational and physically meaningless in the calculation, so absolute values are taken when substituting.

Fig. 2-34 shows the F - δ curves of PDMS made with $R = 8 \text{ nm} = 8 \times 10^{-9} \text{ m}$, $\nu = 0.5$ and crosslinking degrees of 10:1, 20:1, 30:1, and 40:1. The adhesion force increased as PDMS became softer. The Young's modulus was calculated from this and the results are shown in Fig. 2-35. The Young's modulus decreased as the crosslinking degree decreased. This shows that the softness (Young's modulus) of PMDS can be changed by changing the crosslinking degree of PDMS, and the adhesive force can also be adjusted. Mimetic materials were fabricated using PDMS with this degree of crosslinking, and the adhesive strength was measured. As shown in Fig. 2-32, F_1 , δ_0 , and δ_1 are the force and strain at the maximum adhesion point and the equilibrium point ($F = 0$) of the F - δ curve, so the elastic modulus can be calculated from only two points. In this case, the force is shown as a negative value on the graph, but this is irrational and physically meaningless in the calculation, so absolute values are taken when substituting .

Fig. 2-34 shows the F - δ curves of PDMS made with $R = 8 \text{ nm} = 8 \times 10^{-9} \text{ m}$, $\nu = 0.5$ and crosslinking degrees of 10:1, 20:1, 30:1, and 40:1. The adhesion force increased as PDMS became softer. The Young's modulus was calculated from this and the results are shown in Fig. 2-35. The Young's modulus decreased as the crosslinking degree decreased. This shows that the softness (Young's modulus) of PMDS can be changed by changing the crosslinking degree of PDMS, and the adhesive force can also be adjusted. Mimetic materials were fabricated using PDMS with this degree of crosslinking, and the adhesive strength was measured.

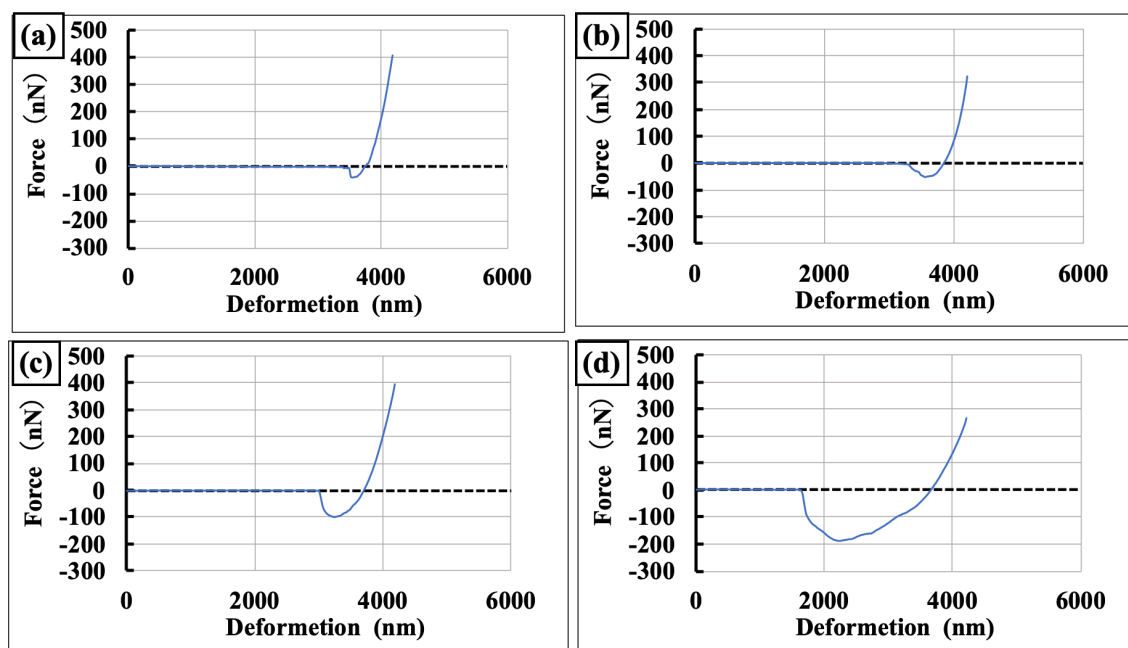


Fig. 2-34 F- δ curves of PDMS prepared with crosslinking ratios of (a) 10:1, (b) 20:1, (c) 30:1, and (d) 40:1

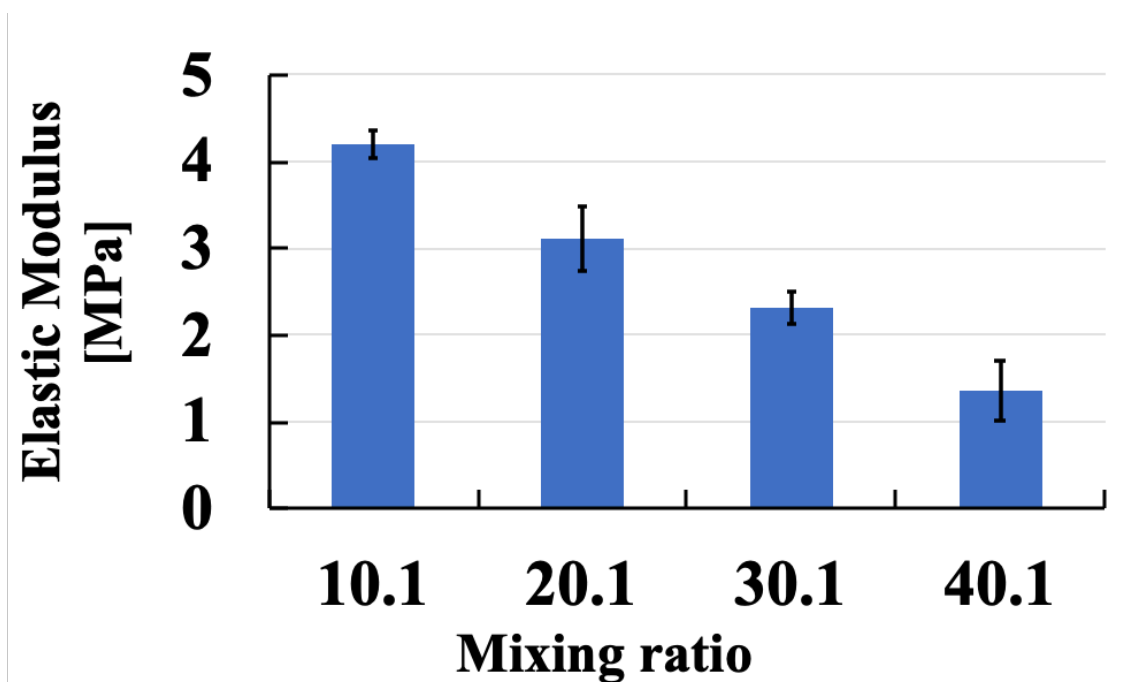


Fig. 2-35 Young's modulus for each degree of crosslinking ($n=10$), error bars are

• Adhesion test and simulation results

The results of the adhesion test are shown in Fig. 2-36. The adhesion strength increased in proportion to the softness of the PDMS. In addition, the flat PSt/PDMS and the mimicked structure showed almost the same increasing trend, but the mimicked structure had a slightly higher rate of increase. The simulation results are shown in Fig. 2-37. The deformation stress was distributed to the nanofilaments, but the lower the Young's modulus (the softer it became), the slightly higher the amount of distribution to the nanofilaments. In general, the softer the material, the lower the yield stress (the stress at which the material begins to deform; after this point, the force is used to deform the material and it is difficult to apply any more force to the material). Therefore, it is thought that the lower the Young's modulus of PDMS, the easier it is for stress to be distributed to the nanofilaments, and the slightly higher the adhesion strength. From the above results, it became clear that the adhesion strength and the amount of stress dispersion change depending on the softness of the PDMS. In addition, the PDMS overhang thickness was previously fixed at about 12 μm , but the softer the PDMS, the thicker the overhang thickness at which the adhesive strength increased (Fig. 2-38).

This is thought to be because softer PDMS has a smaller yield stress and a larger amount of deformation even with the same force, so a larger thickness is required.

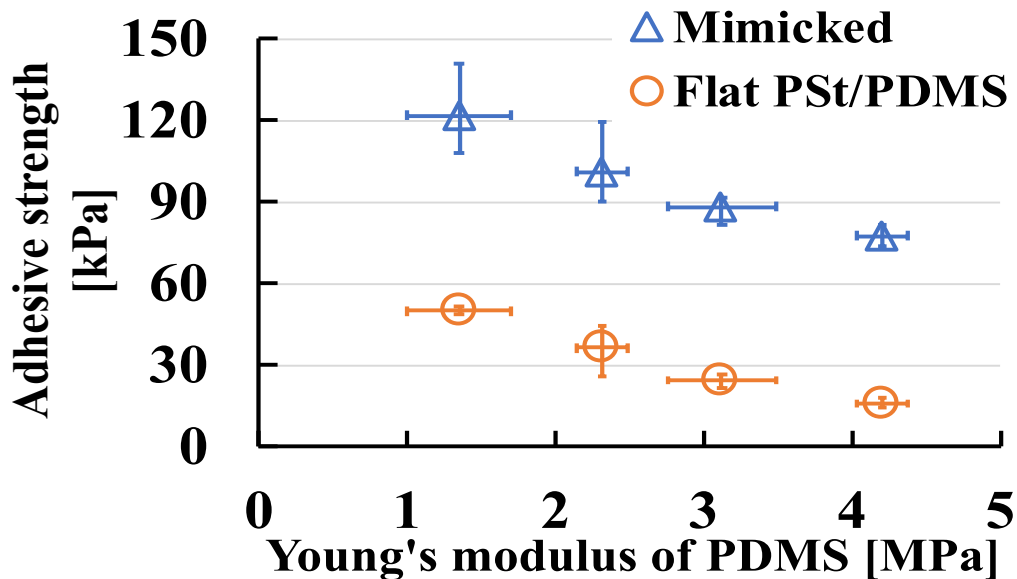


Fig. 2-36 Adhesive strength when changing the softness (Young's modulus) of PDMS

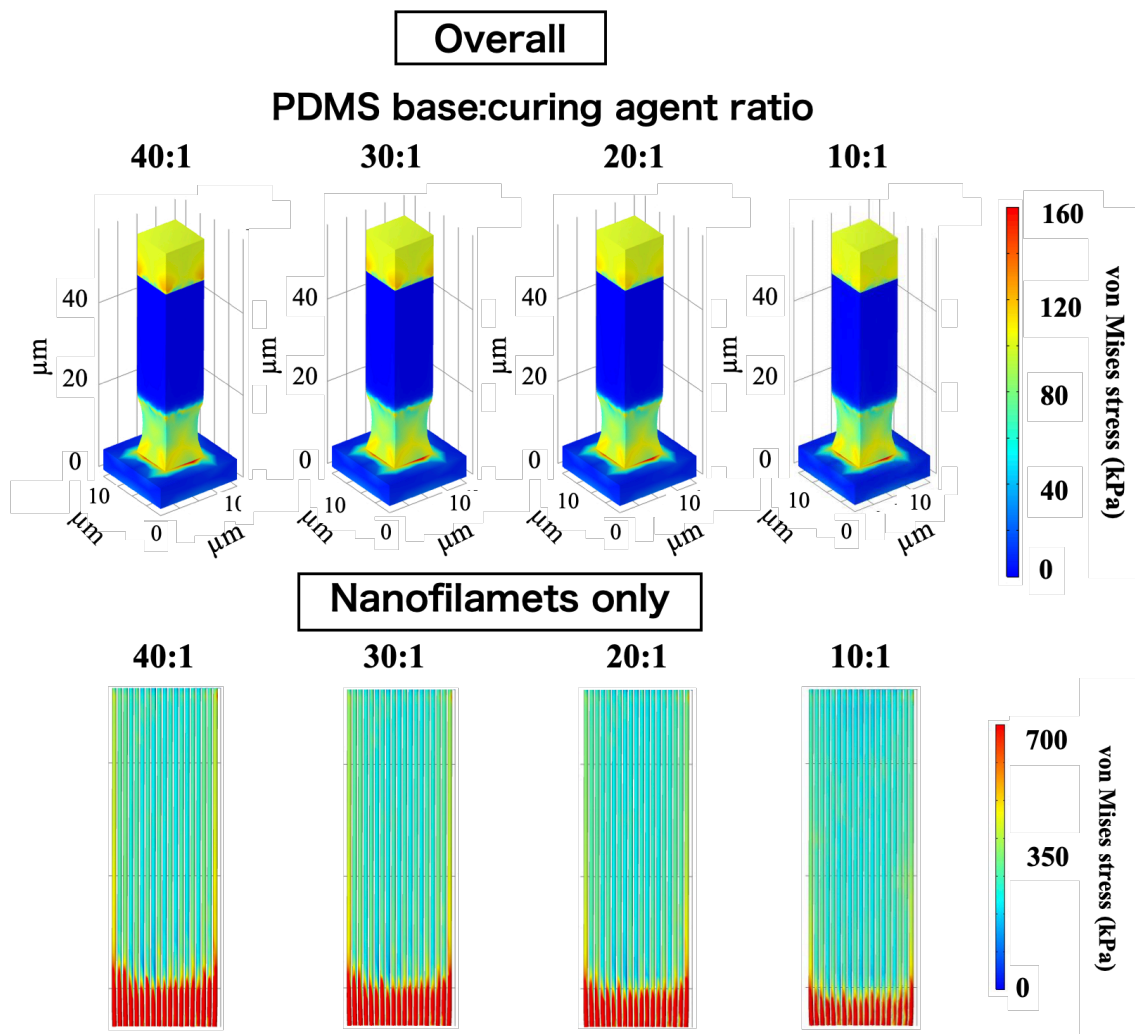


Fig. 2-37 Simulation results for each PDMS softness (Young's modulus):

Young's modulus: 40:1=1.35, 30:1=2.32, 20:1=3.12, 10:1=4.20 [MPa]

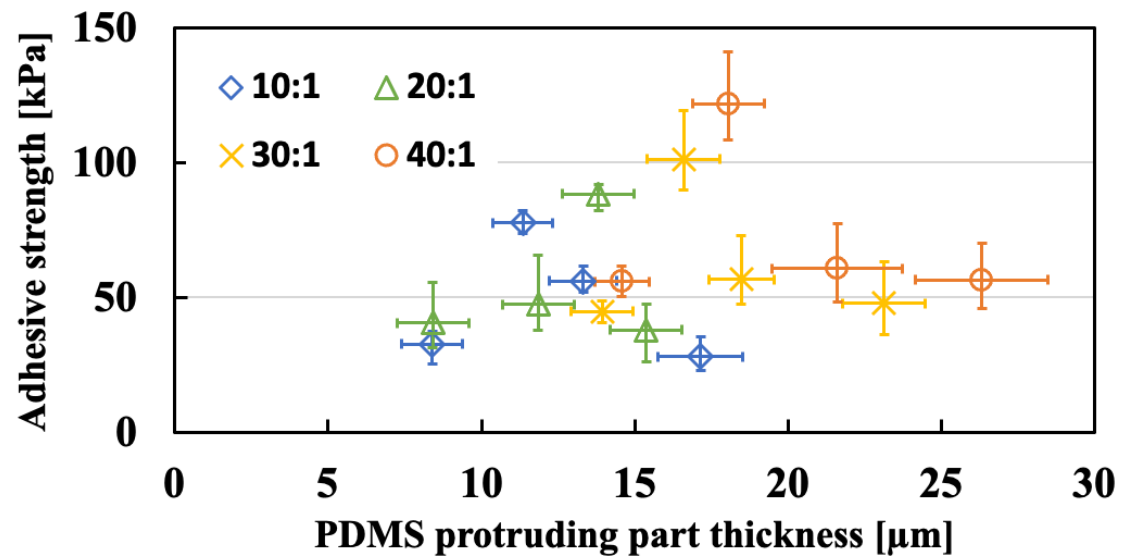


Fig. 2-38 Adhesion strength for each degree of crosslinking when PDMS overhang thickness is changed

2-4 Application (Nano-particle composite adhesive material)

2-4-1 Introduction

In this study, we used AAO as a mold to imitate nanofilaments. AAO is made by self-organization, but it is difficult to make nanofilaments with materials other than thermoplastic resins because it necessarily requires heat pressing. When making nanofilaments by electroforming, the holes are filled in a short time, making it difficult to make nanofilaments with a large aspect ratio. In addition, making nanofilaments with ceramics requires etching, which requires cost, time, and equipment, making it difficult to put into practical use^{2,36-40}. In addition, even with thermoplastic resins, it is difficult to peel the mold from the nanofilament, so the mold is melted each time and used as a disposable item. Therefore, although some functions were imitated, the manufacturing cost is inferior to that of living organisms. Therefore, in this section, we aimed to create an adhesive material inspired by clingfish using fine particles. It has been found that the functions of clingfish mucus and nanofilaments are to generate and strengthen adhesive forces, respectively, and that this requires an increase in contact area due to softness and avoidance of stress concentration on the peeling surface by stress dispersion. Therefore, if stress dispersion occurs, the structure is not limited to nanofilaments and can be free. This time, we used fine particles as a substitute for nanofilaments. Fine particles can be self-organized to create nm-scale materials in a bottom-up manner. Many companies have successfully commercialized fine particles through research to date, and the materials used range from polymers to silica and metals^{69,70}. In addition, as shown in 1-4-2 and the results of the research I conducted from my fourth year of undergraduate studies through my master's degree⁷¹, materials that contain an appropriate concentration of fine particles are known to have improved material properties, such as a significant increase in Young's modulus, toughness, and material strength⁶⁹⁻⁷¹. This is thought to be due to stress dispersion, so we thought it would also be effective in adhesive materials.

2-4-2 Sample preparation

First, silica particles (Hipressica) with a diameter of 500 nm were mixed with PDMS. First, silica particles (Hipressica) with a diameter of 500 nm were mixed with PDMS (base material) at a concentration of 25 wt% by stirring overnight with a magnetic stirrer. At this time, 60 wt% hexane was mixed with PDMS because the viscosity of PDMS was too high to disperse the particles evenly if it was used as is. After stirring, the

mixed solution in which the PDMS curing agent was mixed in a ratio of 10:1 was spin-coated onto a polystyrene film (①500 rpm, 3 s, ②1000 rpm, 7 s), the film was degassed to remove the hexane, and the PDMS was thermally crosslinked at 70°C for 12 hours(Fig. 2-39). A sample without particles was also prepared for comparison, and 60 wt% PDMS/wall reference solution was spin-coated, degassed, and thermally crosslinked under the same conditions. The prepared sample was pressed against glass with a force of 10 N using a tensile tester for 10 seconds, and the maximum adhesive strength was measured when it was pulled back at a speed of 10 mm/min. Although the material we created incorporates the principle, it can no longer be called an imitation of the structure, so we decided to call it ClingS (Clingfish learning adhesive material using soft material and hard nano-micro structure).

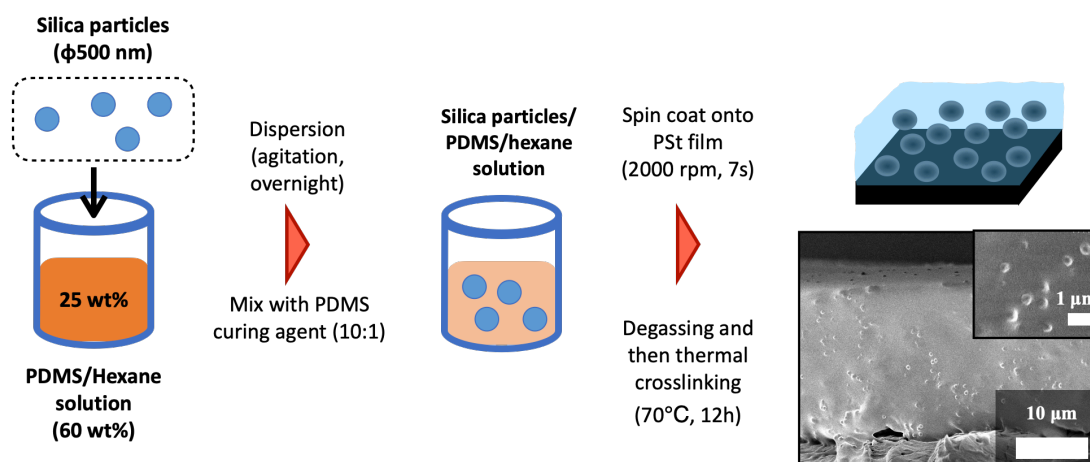


Fig. 2-39 The procedure for producing ClingS, Clingfish learning adhesive material using soft material and hard nano-micro structure.

2-4-3 Adhesion test results

The FE-SEM images and adhesion test results of the prepared samples are shown in Fig. 2-40. It was confirmed that the microparticles were uniformly mixed in the PDMS, and that the particles were not protruding from the surface, and were entirely covered with PDMS. In this case, the PDMS thickness of the sample without microparticles was $20.17 \pm 1.26 \mu\text{m}$, and that of ClingS was $20.47 \pm 1.29 \mu\text{m}$. The adhesion test showed that the sample containing particles showed higher adhesion strength both in water and in air than the sample without particles (Fig. 2-40 (c)). This is thought to be because, like clingfish, the thin PDMS layer near the adhesion surface became an adhesive layer, and the adhesion strength was enhanced by the filler effect of the hard microparticles. In addition, when the thickness was changed by changing the spin coating speed (②) to 500 rpm, 1000 rpm, and 3000 rpm, the adhesion strength only changed for ClingS, suggesting that there is an appropriate thickness for the filler effect (Fig. 2-41).

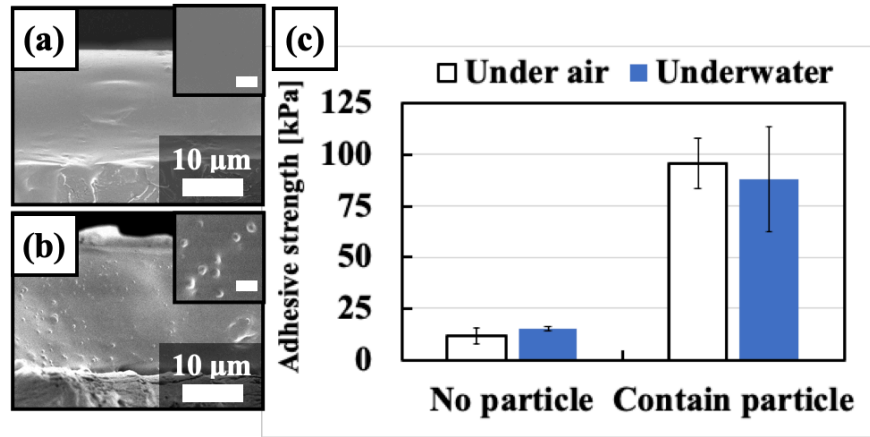


Fig. 2-40 Cross-sectional FE-SEM images of (a) the no particle and (b) the contain particle. Insert images of magnified images. Scale bar : 1 μm. (c)

Graph of the adhesive strength of the sample.

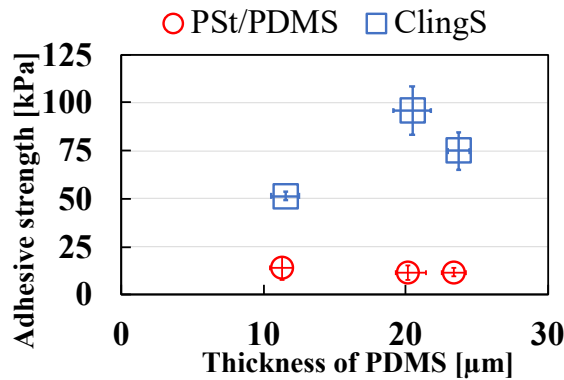


Fig. 2-41 Adhesive strength (n=4) for each sample thickness (n=9)

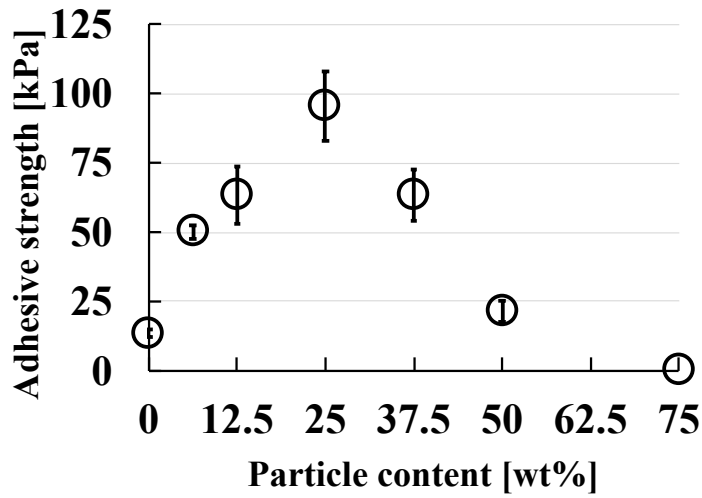


Fig. 2-42 Adhesion strength when changing the concentration of fine particles

Next, since many papers report that the material strength changes depending on the microparticle concentration, Fig. 2-42 shows the results of the adhesion test when the concentration of the mixed microparticles is changed. For the sample, silica microparticles were mixed into a PDMS solution mixed with 60 wt% hexane as in the previous work. The adhesion strength changed depending on the microparticle concentration, with 25 wt% having the highest adhesion. In this study, the spin coating speed was 1000 rpm and the thickness was approximately 20 μm for all samples, but only 50 wt% had a thickness of about 27 μm . However, the adhesion strength did not increase significantly even when the thickness was changed. This is thought to be because the microparticle concentration was too high to exhibit the filler effect, and there was no change due to thickness like in PSt/PDMS. In addition, there were many microparticles in 75 wt %, which caused roughness on the surface, so no adhesion strength was observed. From the above results, it became clear that the adhesion strength increased due to the microparticles, and the adhesion strength changed depending on the concentration. In addition, when checking the force curve during the adhesive strength test, the mimicked structure using nanofilament shown in the previous section showed the same slope as the sample without nanofilament or without microparticles, but ClingS with microparticles had a steeper slope than the other samples (Fig. 2-43). In tests like this, a steeper slope indicates an increase in hardness. Therefore, while the sample using nanofilament only changes G_c without changing the hardness in the formula, it is possible that the adhesive strength of ClingS increases due to the change in hardness. This suggests that the

parameters for increasing adhesive strength can be individually changed depending on the shape.

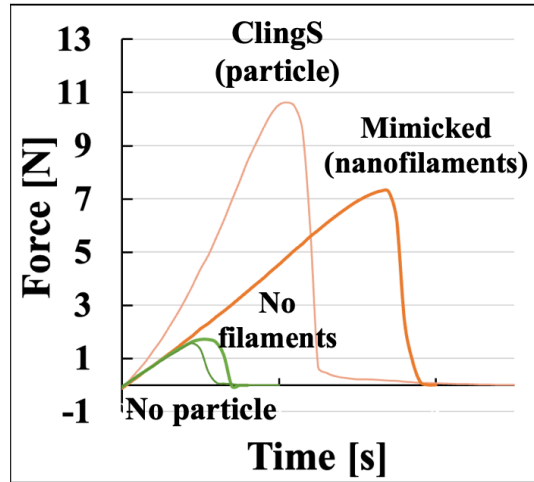


Fig. 2-43 Adhesion test force curves for each sample

In addition, to investigate the functionality of ClingS, we measured the adhesive strength of ClingS to sandpaper underwater, and found that it showed high adhesive strength even on rough surfaces underwater (Fig. 2-44 (a)). However, as with the sample using nanofilament, the adhesive strength decreased as the roughness increased. In addition, when repeated adhesion tests were performed on rough surfaces underwater ($S_a = 4.56 \mu\text{m}$, #1000 sandpaper), the adhesive strength decreased but remained constant at just under 60 kPa, and maintained a higher value than samples without particles (Fig. 2-44 (b)). From these results, we have succeeded in producing a material that has high adhesive strength even on rough surfaces underwater, can be repeatedly bonded, and can be easily produced, like clingfish. In addition, although this time it was produced by spin coating, properties such as increased adhesive strength, underwater adhesiveness, and repeated adhesiveness were obtained by submerging a PSt solution in a PDMS solution containing particles, taking it out, degassing, and thermal crosslinking (Fig. 2-45). The adhesive strength is lower than that of the sample produced by spin coating because the thickness could not be adjusted. Therefore, if the thickness can be adjusted appropriately by changing the hexane concentration, it may be possible to produce a large area using methods such as roll to roll, which makes it easy to produce large areas.

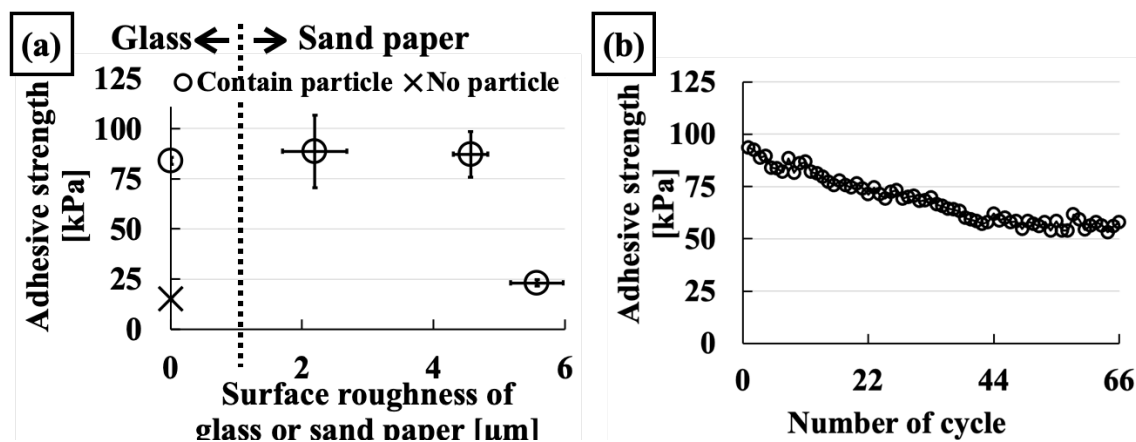


Fig. 2-44 Adhesion test results on sandpaper with different roughness. Repeated test results on sandpaper ($S_a = 4.56 \mu\text{m}$). All measurements in water.

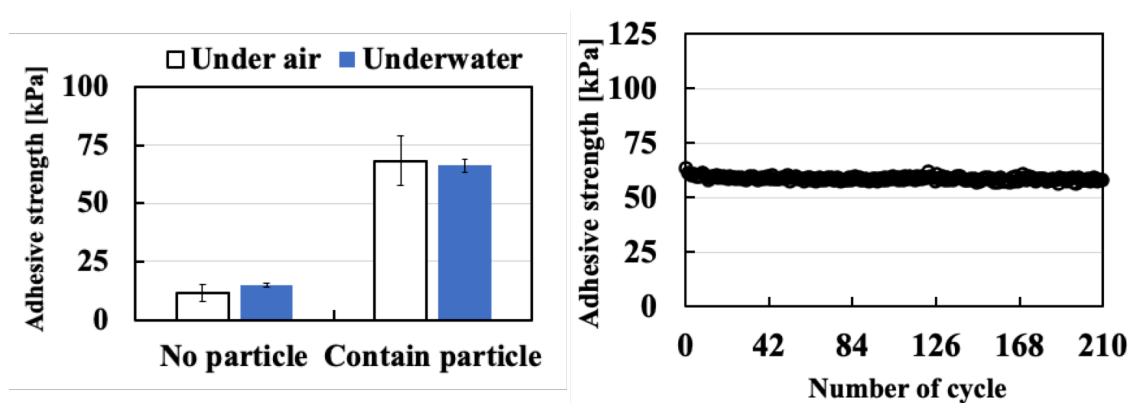


Fig. 2-45 The results of adhesion tests in water and air for a fine particle sample prepared by dipping a PSt film into a mixed solution, and the results of a repeated test in air. A sample without fine particles was prepared by spin coating. Both tests were for adhesion to glass, and the repeated test was performed in air.

2-4-4 Simulation results

Next, a simulation was performed to confirm the stress dispersion. The model and results are shown in Fig. 2-46. The model was created in 2D, and the force when pulled upwards at the same thickness was calculated. As a result, as with the nanofilament, the force on the edge was reduced by dispersing the stress to the microparticles, which is thought to lead to an increase in adhesive strength by avoiding stress concentration through stress dispersion. In addition, as shown in Fig. 2-47, when the deformation amount against the pulling force was examined, the nanofilament model had the same slope as the model without nanofilament, while the model with microparticles had a steeper slope. As with Fig. 2-43, this result supports the idea that the hardness of the nanofilament does not change, but that of the microparticles does change.

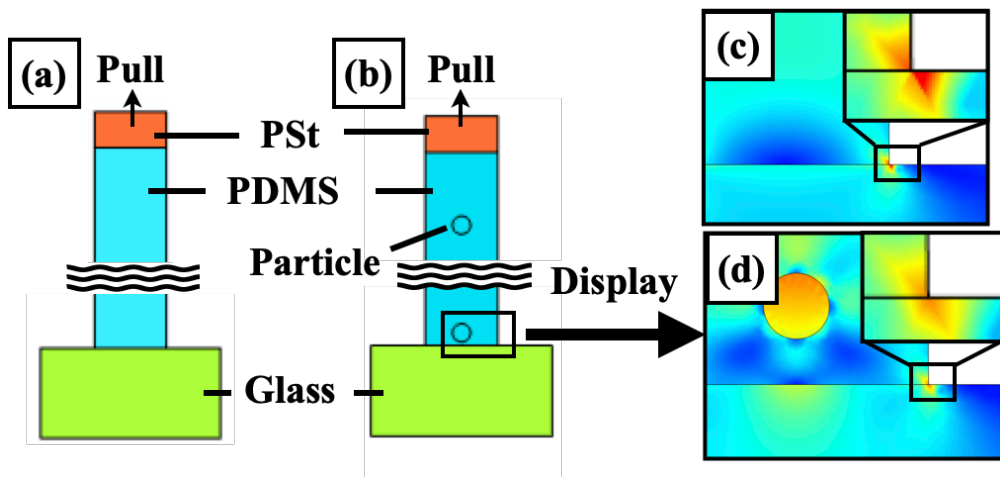


Fig. 2-46 FEM simulation (a, b) model and (c, d) result of (a, c) the no particle and (b, d) the contain particle. Some parts are long, so omitted. Insert images of magnified images.

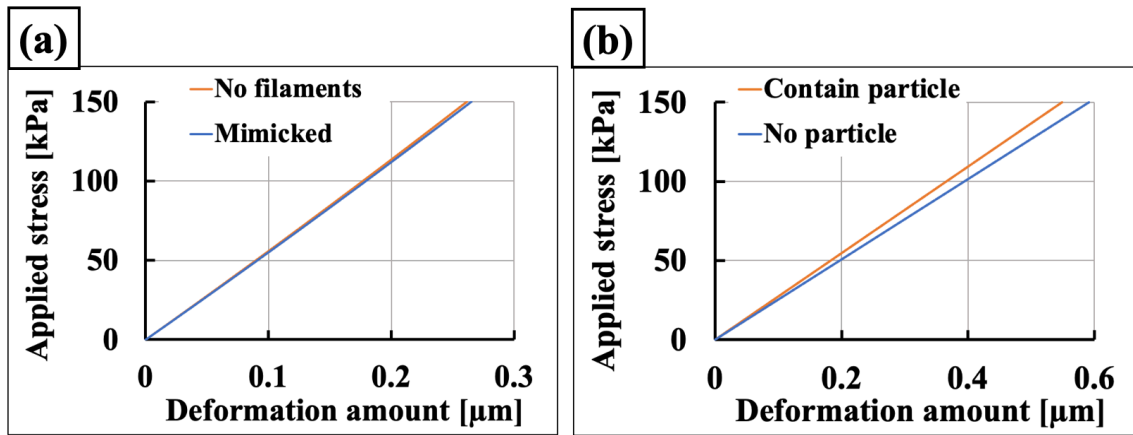


Fig. 2-47 Calculation results of deformation amount against pulling force of (a) nanofilaments
(b) particle structural model

2-5 Comparison with other organisms and studies

In this section, we compare nanofilaments with other organisms as a feedback to biology. The functions of nanofilaments have been described in many organisms, including geckos, beetles, and spiders^{4,14,18,72,73}. Mimicked structures have also been created for these structures, but their function is to generate adhesive forces, which is different from clingfish. There are also differences in shape, and unlike the single clingfish nanofilaments, their nanofilaments have structures such as spatulas at the tip. The softness required for adhesion (increased contact area) is achieved by contact with the spatula structure and contact division by the nanofilaments, and the hardness role of resistance to peeling is exerted by being made of a hard material. Therefore, simple nanofilaments alone do not exert sufficient effects, and sometimes adhesive forces are not generated, as in the results of our research. The mimicked structure also approaches the adhesive forces of geckos for the first time by using spatulas. In addition, unlike the hard nanofilaments of geckos, artificial materials are often made of PDMS, which is easy to adhere to. This is because it is difficult to create a highly functional adhesive material (microstructure) that is a hard material. Tape made from highly oriented carbon nanotubes (CNTs) exhibits adhesive strength similar to that of geckos without the need for structures such as spatulas, but after one application, the CNTs are crushed and the adhesive strength decreases. In addition, most organisms that have structures such as spatulas are basically terrestrial. Given the presence of water and mucus, which makes it difficult to produce adhesive strength, and the results of our research, it is expected that the functions of nanofilaments in terrestrial organisms such as geckos are significantly different from those of nanofilaments in clingfish.

Other organisms with nanofilaments like clingfish have been reported (Fig. 2-48). These include octopuses^{74,75}, sea anemones⁷⁶, and planarians⁷⁷, but all of these organisms live in water and many of their bodies are covered with mucus. The functions of sea anemones and planarians have not been investigated. As for octopuses, it was hypothesized that (i) they use the presence of mucus and filaments to increase the viscosity coefficient at the interface and resist shear forces; (ii) they use the high bulk modulus of the water between the sucker and the substrate to resist tensile stress (detachment forces), which is close to our ideas⁷⁴. This paper also had doubts about the generation of van der Waals forces in water or in the presence of water, but it was not demonstrated. Another well-known organism with nanofilaments and mucus is the

abalone (Fig. 2-49)^{78,79}. Detailed adhesion tests of these organisms to plates with holes and grooves have been conducted⁷⁸. It was subsequently revealed that vacuum adhesion accounts for more than half of the total adhesive force of the abalone's gastropods, accounting for more than 60%, van der Waals force for more than 20%, and nanofilament tubule force for only about 1%. However, in this study, the total adhesive force actually measured was smaller than the adhesive force calculated based on the assumption of a 100% vacuum, so van der Waals force was calculated first, but the calculation was made under the assumption that the substrate and the fiber were in contact (Fig. 2-49 (a)). The nanofilament tubule force was calculated from adhesive force tests and theoretical calculations of abalone that had died and could no longer generate adhesive forces (vacuum adhesion force and van der Waals force). Since the nanofilament tubule force is negligibly small, the vacuum adhesion force was calculated by subtracting the van der Waals force from the total adhesive force. In addition, the breakdown of the vacuum adhesion force was calculated as the adhesion of the entire gastropod, the local adhesion of the gastropod, and the friction equivalent vacuum adhesion. However, the generation of van der Waals forces in the presence of water or mucus was not discussed. On the other hand, another researcher who studied adhesion in abalone has shown that mucus is essential for successful adhesion of abalone based on nanofilament tubule and viscous forces, and that the mucus in the non-adhesive zone, which has a low adhesive force and friction coefficient, may function as a lubricant for locomotion⁷⁹. This is a completely different claim from the above researcher. Theoretical calculations also show that the microstructures may help the abalone to form multiple liquid bridges with the mucus secreted by the abalone, significantly increasing the wet adhesive force of the abalone. This researcher, like us, assumes that the mechanical interlock, van der Waals forces are limited by a thin hydration film in water, and although the one in Fig. 2-49 (b) has had most of the mucus removed by ultrasonic treatment, a nanofilament completely covered by mucus is also shown in the text. Adhesion forces have also been measured in clingfish, and the function of the nanofilament has been estimated, but it was claimed that it is the generation of frictional forces. In addition, there is an adhesive material that has a layer with embedded nanofilaments and an adhesive layer, inspired by snail mucus and structure, and it was confirmed that the nanofilaments also significantly increase the adhesive strength (Fig. 2-50)⁸⁰. However, the size of the structure, the direction in which they grow, and the behavior during peeling are different from ours. Ours is 200 nm in

size, while the snail's is 10-100 μm . Above all, in this study, the nanofilament is made of soft PDMS, and the material that covers it is made of hard PUA. A soft PDMS is attached to it to form an adhesive layer. Although the design is different from ours, the idea of an adhesive layer and a stress dispersion layer is the same. From the above, the current function of a single nanofilament in water is divided into the generation of interlock, van der Waals forces, frictional forces, etc., and the strengthening of adhesive forces of the mucus that covers the nanofilament by dispersing stress. These two claims are difficult to reconcile in terms of whether the nanofilament itself generates forces that contribute to adhesion. Interestingly, the opposite idea has existed even in the same species, such as abalone and clingfish. The key to resolving the above contradiction lies in the generation of mechanical interlock and van der Waals forces in the presence of water and mucus. Normally, it has been thought that these are difficult to generate in the presence of water and mucus, and we have also conducted research based on this idea. Therefore, the function of nanofilament, such as the generation of interlock and van der Waals forces, is a hypothesis that does not take into account the existence in the actual environment of water and mucus. However, there are few studies that have actually investigated in detail the effects of water and mucus on nanofilaments made by living organisms. We have also revealed that greased nanofilaments have the function of strengthening adhesive forces, but of course the chemical properties of the materials are very different. Some living organisms, such as barnacles, can generate substances that can adhere to the surface of the material without forming a hydration layer even in water^{62,63}. It is difficult for humans to completely reproduce these substances, and it is possible that the nanofilaments and mucus of clingfish and abalone can generate mechanical interlock and van der Waals forces by nanofilaments even in the presence of water or mucus. If it is a simple frictional force, it may be possible to generate it even in water. In addition, in the case of clingfish, the species is different and the shape of the microstructure is different. The nanofilaments of clingfish, which claim frictional force, have thin tips and are divided into nanofilaments of several hundred nanometers, so they may be easier to generate frictional force. The hierarchical structure with fine nanofilaments at the tips of the nanofilaments has an adhesive effect in air. At least, the function of the nanofilaments in the mimic material in the versatile material we make is not mechanical interlock, van der Waals forces, or frictional force generation, but rather strengthening the adhesive force of the mucus by dispersing stress. This is because the nanofilaments are in the bulk PDMS and

cannot touch the surface. An increase in adhesive strength was observed when grease was applied, and many previous studies have noted that the above forces are less likely to be generated in water, so it is likely that living organisms have a similar function; however, the functions of living materials have not been fully elucidated, so further research is needed to unravel these functions.

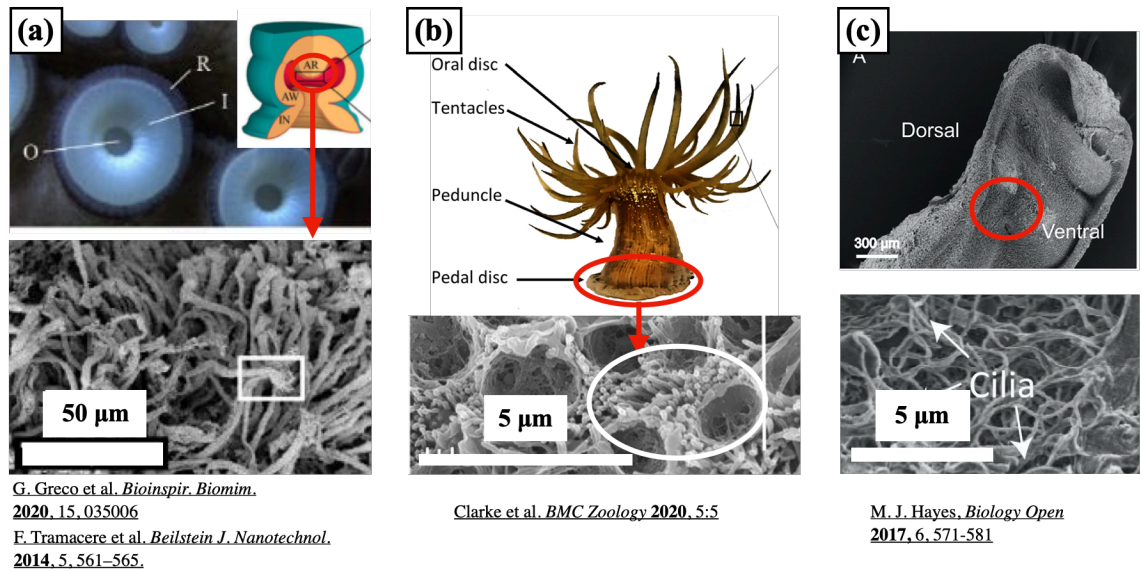


Fig. 2-48 Nanofilaments in (a) octopus, (b) sea anemone, and (c) planarian

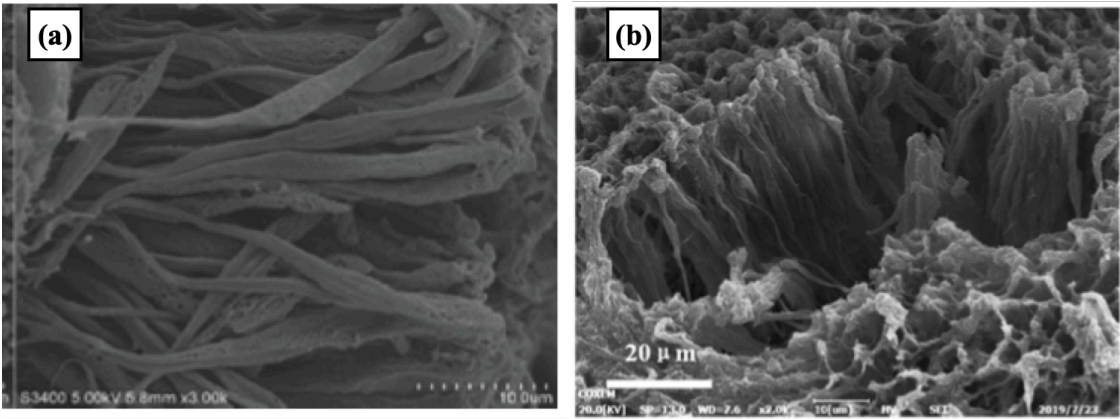


Fig. 2-49 SEM images of abalone nanofilaments. (a) Jing Li et al.⁷⁹, (b) Peng Xi⁷⁸ et al.

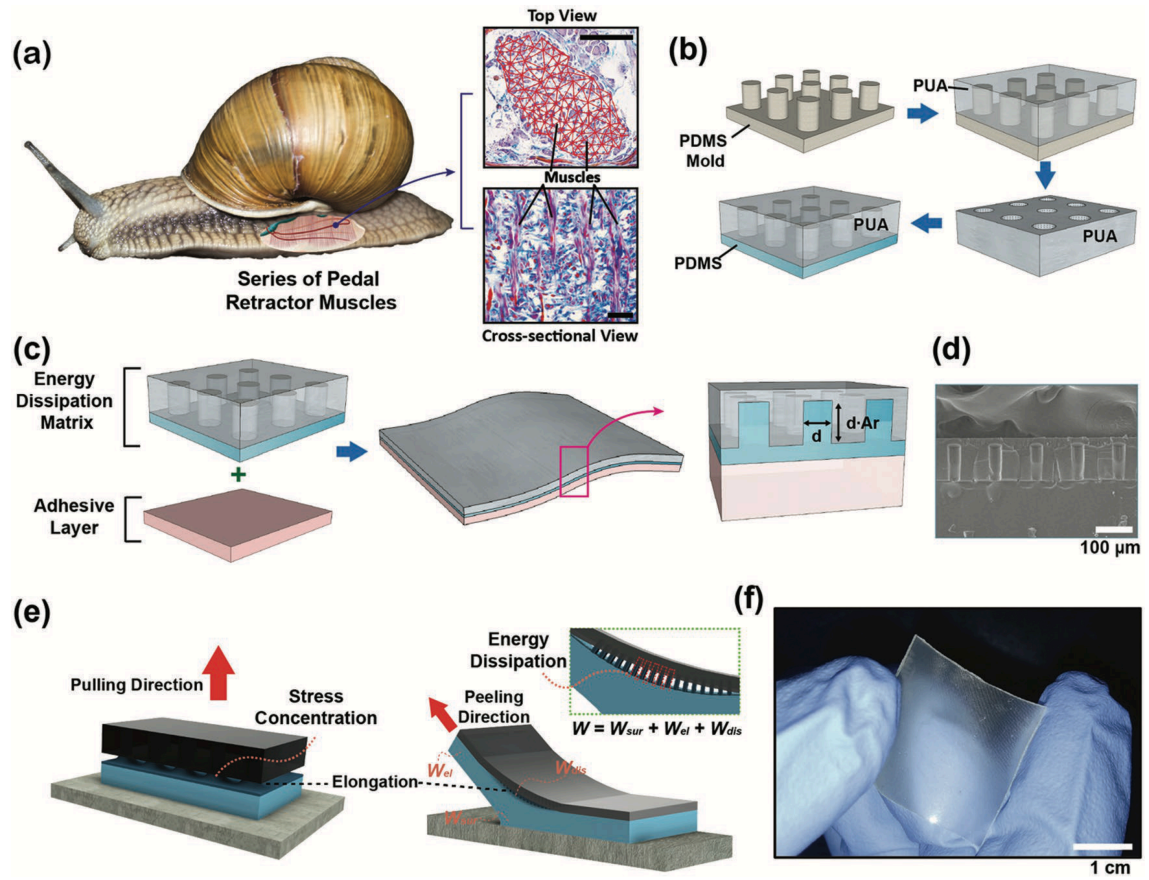


Fig. 2-50 SIA with interlocked microstructures. a) A schematic snail pedal muscle structure. Right optical microscopic images of a section of a snail pedal. Reproduced with permission. b) A schematic of fabrication of snail-inspired microstructures. c) Detailed illustrations and d) a cross-sectional SEM image of the SIA with embedded microstructures. e) Mechanism of the SIA for pulling and peeling direction. f) An optical image of the SIA.⁸⁰

2-6 Summary

In this chapter, we investigated the effect of nanofilaments covered with soft materials on adhesion, focusing on the surface microstructure and mucus of clingfish suckers. Experiments were conducted by changing the length and hardness of the nanofilament, the softness of the PDMS, and the overhanging thickness. The results of adhesion tests and simulations suggested that nanofilaments increase the adhesive strength of soft materials (PDMS), and that this is caused by stress dispersion to the nanofilament. We also found that there are appropriate thicknesses and lengths of PDMS and nanofilaments that increase adhesive strength.

These results suggest that the function of clingfish nanofilaments is not to generate adhesive strength or friction, but to increase the adhesive strength of the soft material that surrounds them. It has become clear that artificial materials, too, can simultaneously achieve both softness and hardness, which are the basis of adhesion, with hard nanofilaments covered with soft materials. If this principle can be achieved, it will be possible to create adhesive materials for various purposes using various materials and structures. In fact, it has become possible to achieve superior functionality using the same principle even in fine particles, which have greater advantages in process and after use than nanofilaments. In this study, hydrophobic, flexible PDMS was used, which enabled adhesion even to uneven surfaces in water. In addition, the structure of covering the nanofilament with a soft material has made it possible to use it repeatedly without leaving any residue or causing damage. In addition, by attaching the MLAs structure to the top of the PDMS, water can be quickly expelled even underwater, enabling quick adhesion.

In addition, although we focused only on clingfish this time, clingfish are not the only organisms that have this combination of mucus and nanofilament. It has been reported that it exists inside the sucker of octopus^{74,75}, sea anemones⁷⁶, planaria⁷⁷, abalone^{78,79}, etc. (Fig. 2-48), but its function was unclear. Therefore, the adhesion mechanism revealed in this study may not be special but may be general in nature. In this context, the clingfish adhesion mechanism may contribute not only to material development but also to elucidating the adhesion mechanisms of other organisms. Since other clingfish have a different microstructure from the clingfish investigated in this study,

Based on the principles learned from clingfish, we have succeeded in creating high-functioning and multifunctional clingfish-like organisms using human technology.

Although it was possible to achieve multifunctionality using nanofilaments, the manufacturing process could be reproduced using low-cost microparticles. This is not a simple imitation of functions, but rather contributes to the creation of products that are in harmony with the environment, such as clingfish.

Chapter 3

Discovery, principle elucidation, and application of carion beetle surface microstructure

3-1 Introduction

3-1-1 Discovery of surface microstructure of carrion beetle

We newly discovered that the *Necrophila japonica*, a type of carrion beetle, exhibits a hierarchical surface microstructure in which a submicron-scale wrinkle structure exists on a microscale microstructure dome (Fig. 3-1). This structure was mainly confirmed on the elytra but not on the back of the elytra (Fig. 3-2). The *Necrophila japonica* mainly feeds on dominant macroinvertebrates of soil ecosystems and often lives on forest surfaces (beneath fallen leaves, etc.)⁸¹⁻⁸⁵ (Fig. 3-1 (a)); however, the functions of its hierarchical surface microstructure are unknown.

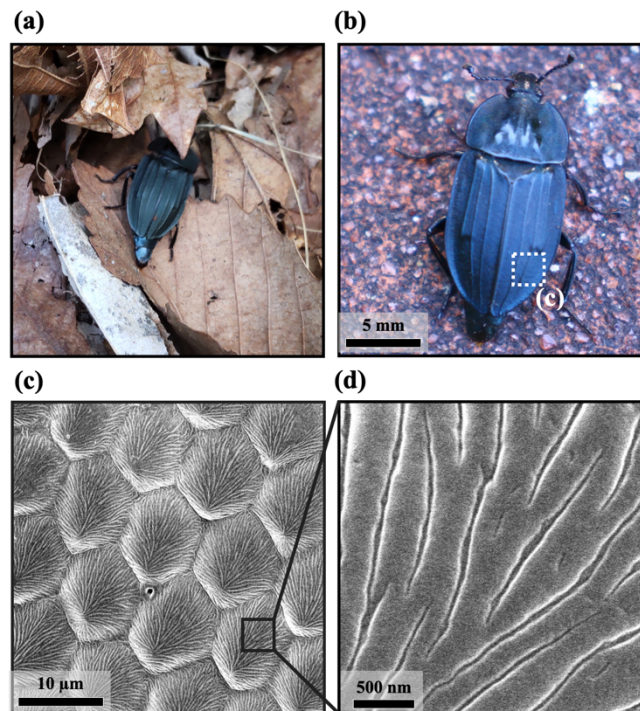


Fig. 3-1 (a, b) Photographs of carrion beetle (*Necrophila japonica*) and (c, d) field emission scanning electron microscope (FE-SEM) images of elytra. In elytra, (c) microstructures of micron order were arranged almost in a hexagonal lattice, and (d) wrinkle structures of submicron order were on the microstructures. Samples were collected in Bibi, Chitose City, Hokkaido, Japan.

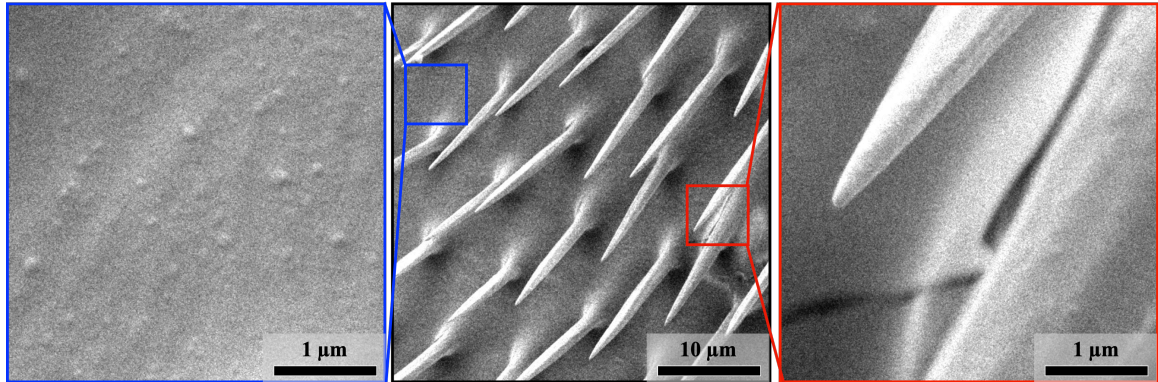


Fig. 3-2 FE-SEM image of the back of the elytra of the carrion beetle. The images surrounded by blue (left), black (middle), and red (right) borders represent the magnified view of the microtrichia-free surface, the overall image, and the magnified view of the microtrichia, respectively.

3-1-2 Friction reduction by surface microstructure

Previous studies revealed that the hierarchical surface microstructure mostly exhibits superhydrophobicity functions (self-cleaning of dirt-containing bacteria, ensuring breathing, etc. ^{9,86–89}), and famous examples include lotus leaves ^{3,9,90} and springtails ^{86,91} that live around forest surfaces, similar to the carrion beetle. However, superhydrophobicity was not observed when we evaluated the wettability of the hierarchical structure of *Necrophila japonica* (contact angle: $\sim 90^\circ$, For details, see Fig. 3-40). The wettability of the hierarchical surface microstructure of the slithering gecko (a gecko crawls like snakes as it does not have legs), which exhibits a wrinkled structure on top of a microstructure similar to the *Necrophila japonica*, has been evaluated, although it also failed to exhibit superhydrophobicity ⁹². Conversely, the group that studied slithering geckos suggested, without examining the details, that the hierarchical structure on their dorsal side may have evolved from their contact with leaves and other objects on their backs while crawling and may have exerted some frictional effects ⁹². Notably, numerous living organisms deploy surface microstructures to control friction without lubricants ⁹³. The shapes and sizes of these microstructures differ, such as firebrats (submicron-sized wrinkle structures) ⁹⁴ and crickets (hexagonal micropatterns) ⁹⁵. However, the details of their hierarchical structure remain unclear. *Necrophila japonica* also lives in an environment where its back contacts various objects, and its hierarchical

structure is similar to that obtained by combining a firebrat and cricket; therefore, the surface microstructure of its back may be involved in friction.

3-1-3 Knowledge of friction and structural mechanics and their fusion

Reducing friction is a challenge faced not only by living organisms but also by humans. The reduction of friction is a critical issue in many industries and everyday life. While friction is necessary for activities such as walking and the operation of machinery, excessive friction can sometimes cause problems. Friction consumes approximately 20% of energy consumption worldwide, with total economic losses due to frictional contact in all sectors of society of an estimated €2,536 billion per year, of which 73% is due to friction and 27% to wear ⁹⁶. In the case of automobiles, direct friction losses, excluding those from braking, account for 28% of fuel energy consumption. In 2009, 208 billion liters of fuel were used worldwide in 2009 to overcome this friction ⁹⁷. Therefore, friction reduction is not only economically advantageous but also important for addressing energy issues such as carbon neutrality and related medical concerns ⁹⁸. A common method to reduce frictional forces is the application of lubricants; however, lubricants must be replenished periodically and can introduce new issues, such as the risk of combustion ⁹⁹. In addition, the , the leakage of harmful lubricants poses significant environmental risks ^{99,100}. Lubricant-free friction reduction methods are therefore increasingly sought after in various fields due to work environment constraints and concerns over environmental pollution. According to the Amontons-Coulomb law, the nanoscale real contact area that generates friction is much smaller than the apparent contact area and increases linearly with the load ¹⁰¹. Therefore, for many plastically deforming materials, the coefficient of friction (friction force/load) remains constant. In other words, the deformation of the roughness (increase in the real contact area) is proportional to the load. However, while this principle holds true at the macroscale, where the contact area of the indenter in hard plastically deforming materials is sufficiently large, other scenarios remain to be explored. The aforementioned principle is difficult to apply to nanoscale regions such as atomic force microscope measurements ¹⁰², low load regions where plastic deformation is unlikely to occur ¹⁰³, and elastically deforming material surfaces where the apparent and real contact areas are almost indistinguishable due to their flexibility ^{98,104,105}. Furthermore, advances in electron microscopy and microfabrication have made it possible to create complex surface microstructures at the submicron scale ^{13,106,107}, and research on the friction of such microstructured surfaces has been actively conducted ^{93,94,108}. Many studies have demonstrated that surface microstructures effectively reduce friction forces compared to flat surfaces, even in hard plastically deforming materials

^{93,108–113}. These studies emphasize that the reduction in friction is due to the reduction in contact area promoted by the microstructure, which has been supported by the Hertz equation, among others. However, many principles of friction different from the Amonton-Coulomb law have been studied, but all of them have different scales, loads, and material properties to explain the reduction in friction under macroscale indentation conditions by microstructures fabricated from plastically deforming materials. In particular, although the apparent contact area may be reduced by surface microstructures, the mechanism of why the pressure concentrated on the reduced apparent contact area does not increase the effective contact area proportionally to the load remains unclear.

On the other hand, some insects, such as ants, show a remarkable ability to traverse vertical surfaces due to the increased effective contact area caused by the microstructure of their legs ^{14,65,73,114}, but struggle to maintain traction even at slight inclinations on surfaces with moth-eye structures ¹¹⁵. This has been attributed to a reduction in the contact area. Correspondingly, artificial and natural rough surfaces with dimensions comparable to those of moth-eye structures significantly reduce the frictional forces of insects ^{116,117}. In fact, the trends observed in the calculated and measured values are consistently consistent, revealing a large variation in frictional forces depending on the size of the structure ¹¹⁷. Based on these findings, contact on the surface of the moth-eye structure is mainly concentrated at the tip, where the apparent contact area and the true contact area are almost the same. Even in a microcontact state, when a high load is applied, such as that of an ant, the moth-eye structure itself deforms in proportion to the load, increasing the true contact area. However, it has been suggested that the shape and size of the structure affect its deformation characteristics in various fields. As shown by arched bridges and submicron-scale metamaterials, certain structural shapes can suppress deformation by distributing the load to the sides and bottom rather than the surface, avoiding the stress concentration required for deformation ^{118–120}. In addition, Holmberg et al. implied that increasing the surface roughness on the submicron scale reduces the dry friction coefficient of plastically deforming materials, and they reasoned that this is because the roughness supports the load over a larger area, reducing the contact area and adhesion. Therefore, in a surface with a microstructure, the true contact area does not increase proportionally with the load, and the friction force is expected to decrease compared to a surface without a structure that obeys the Amonton-Coulomb law. In addition, while reducing the number of contact points to reduce friction in a single-layer

structure causes pressure concentration, a hierarchical structure like that of a carrion beetle can reduce the number of contact points while widely distributing the load to the wrinkles and the dome layer below, which may improve the deformation suppression effect and reduce friction more than a single-layer structure.

3-1-4 Evaluation of functions through the microstructure of carrion beetle, elucidation of the principles, and application

First, we measured the friction of the body surface of *Necrophila japonica*. We then removed the microstructure of the body surface by sanding, and compared the frictional force. We also created a hierarchical structure by combining the breath figure method¹⁰⁷ and wrinkle fabrication techniques^{121,122}, and transferred this to create a hierarchical mimicked structure of a single material using a plastic deformation material. We compared the different structural shapes of the mimicked structures: hierarchical structure (MLAs + wrinkle), MLAs only, and wrinkles only. We performed friction force tests on the mimicked structures to investigate the effect of the hierarchical surface microstructure on friction. We then analyzed the principle of friction reduction by finite element method (FEM) simulations.

3-2 Functional evaluation

3-2-1 Friction measurements on the surface of carrion beetle

First, to investigate the friction-reducing effect of the surface microstructure of *Necrophila japonica*, friction measurements were performed on the elytra of *Necrophila japonica*. Friction measurements on the sample surfaces were performed with a constant load in one direction using a TRIBOGEA (HHS2000, SINTO scientific Co., Ltd., Japan). A stainless steel ball (SUS304, AS ONE Corporation, Japan) with a diameter of 3 mm was used as the indenter, and the sliding speed and load for the friction measurements were 1 mm/s and 9.8 mN (≈ 1 g), respectively. The measurement direction was parallel to the line connecting the head and the buttocks. Friction measurements were performed five times for each sample, and the average friction force was calculated for measurement times ranging from 1000 to 3000 ms. All error bars in the graphs represent standard deviations. Samples were prepared from elytra of *Necrophila japonica* killed in a freezer and from elytra whose surfaces were polished with sandpaper. The sandpaper used was #2000, $S_a = 2.19 \mu\text{m}$ (Fig. 3-3). SEM images of the polished elytra are shown in Fig. 3-4. It was shown that the polished sample had no surface microstructure. The measurement results are shown in Fig. 3-5. The unpolished sample with the microstructure had approximately 35% less friction than the polished sample without the microstructure, and the friction coefficient was 0.072, less than 0.1, showing low friction comparable to that of lubricated surfaces^{100,123–126}. These results clearly demonstrated that the surface microstructure of *Necrophila japonica* has a friction-reducing effect.

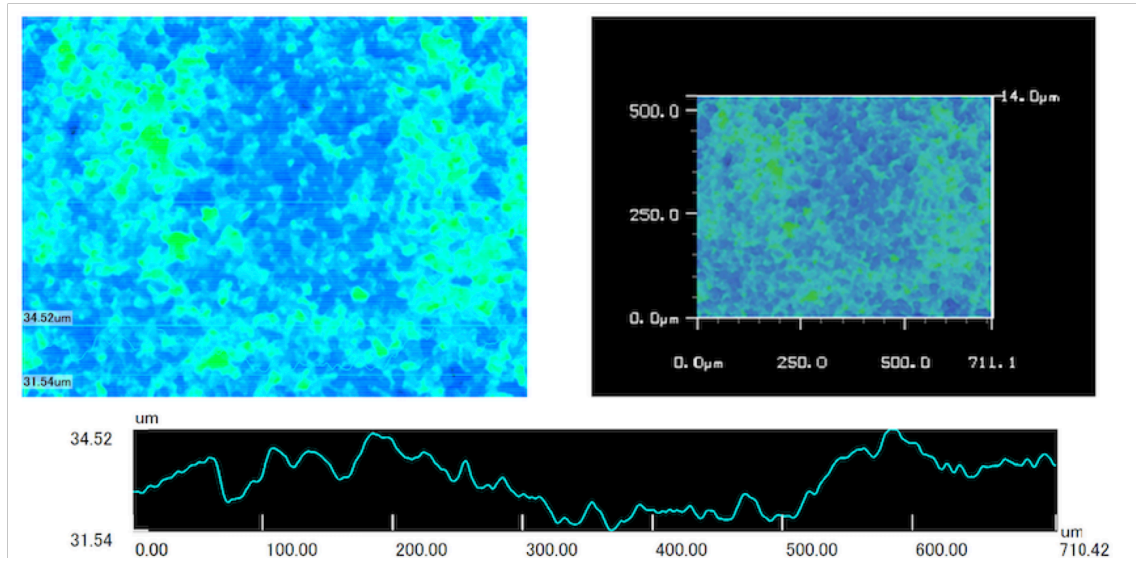


Fig. 3-3 Laser microscope image of sandpaper (#2000, $S_a = 2.19 \mu\text{m}$). The top two are surface images with height indicated by color, and the bottom is a cross-sectional image.

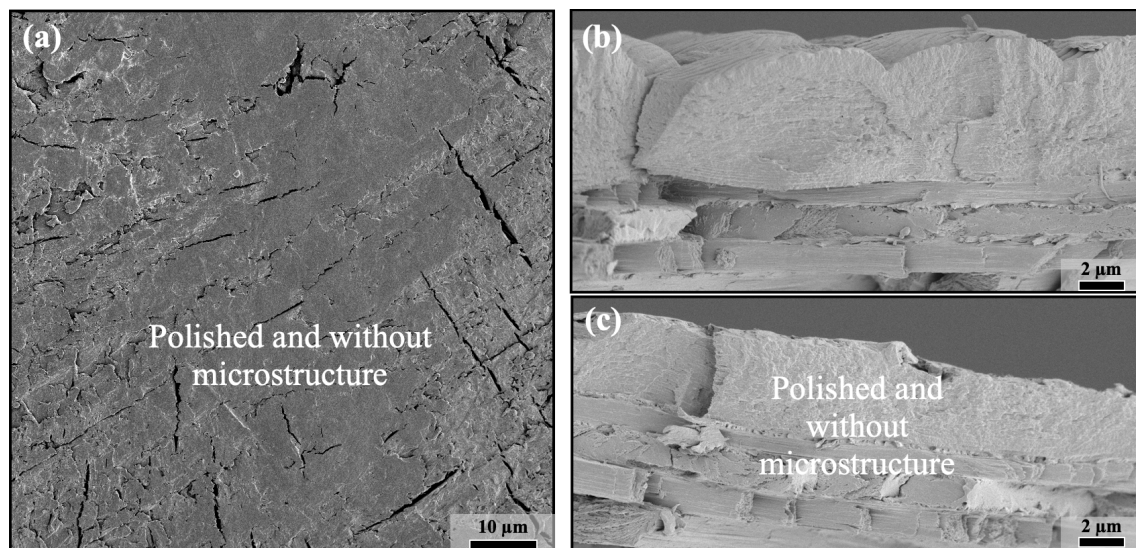


Fig. 3-4 (a) Surface and (b, c) cross-sectional SEM images of (a, c) an elytra polished with sandpaper and (b) an elytra that was not polished.

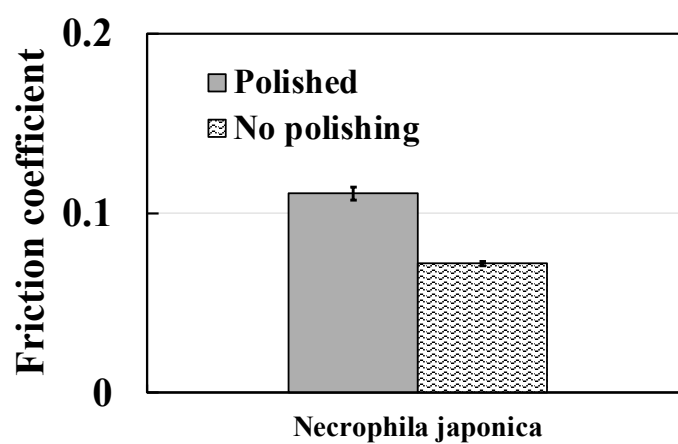


Fig. 3-5 Friction measurements of polished and no polishing samples

3-2-2 Mimicked structure fabrication

In 3-2-1, it was revealed for the first time that the microstructure of *Necrophila japonica* has a friction reducing effect. However, since it is difficult to investigate the effect of shape by measuring the body surface of an organism, friction measurements were carried out using a mimicked structure.

• Preparation of mold

This section describes the method for preparing the mold used for the measurements (Fig. 3-6, Fig. 3-7, Fig. 3-8). The polyvinyl alcohol (PVA, FUJIFILM Wako Pure Chemical Corporation, Japan) cross-linked with an ORGATIX (TC315, Matsumoto Fine Chemical Co. Ltd, Japan) solution was prepared by mixing both solutions with a magnetic stirrer overnight so that the mixing ratio of PVA (hydroxyl group) to ORGATIX (carboxyl group) was 1:1. To prepare the wrinkle structure, a 1 wt% PVA aqueous solution was dropped on flat polydimethylsiloxane (PDMS, crosslinking ratio = base agent:curing agent = 10:1, DOWSILTM, SILPOT 184 W/C, DuPont Toray Specialty Materials K.K., Japan) without a structure and spin-coated. First, the solution was spread by spin coating at 500 rpm for 5 s, after which the thickness was adjusted by spin coating at 3000 rpm for 40 s. To prepare the mimicked structure, a 2 wt% PVA aqueous solution was dropped onto the PDMS-MLAs and spin-coated at 500 rpm for 5 s, followed by 2000 rpm for 5 s. The solution concentrations and spin-coating conditions for the wrinkled and mimicked structures differed because, in the case of MLAs, the liquid easily escapes owing to the microstructure, which is similar to a frog adhesive toe pad 57. The spin-coated samples were dried in an oven at 70°C for 12 h to improve adhesion. Next, the PVA/PDMS sample was compressed by 20% using a hand-operated device (X-Axis, Left/Right Screw, Open/Close Width Adjusting Units, XANON60, MISUMI Corporation, Japan) for the precise control of PDMS compression. Finally, an epoxy resin (Epok 812, Okenshoji Co., Ltd, Japan) was dropped onto the 20% compressed sample, covered with a cleaned glass substrate, and heated at 60°C for 12 h to transfer the structure to the epoxy resin. The MLAs structure employed for the friction measurements was prepared by compressing the symmetric MLAs structure in a mold with a jig without spin coating, followed by transferring it to the epoxy resin.

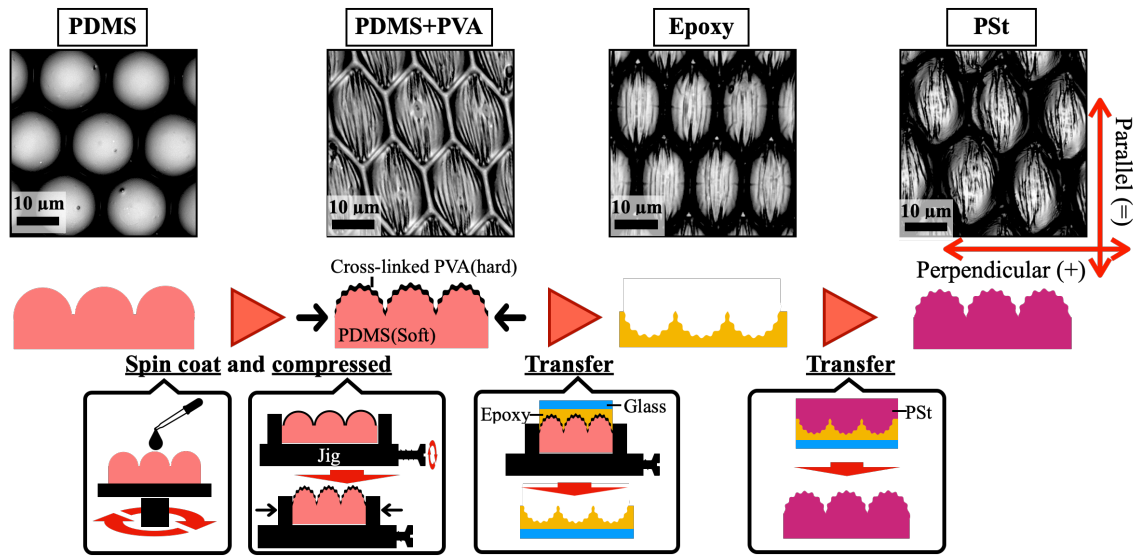


Fig. 3-6 Preparation procedure of the polystyrene (PSt) carrion beetle mimicked structures. Wrinkles were formed by preparing a hard PVA layer on soft PDMS MLAs and compressing it. Although the PVA layer has a large effect on friction, preparing a mimicked structure in a single material using two transfers allows us to investigate the effect of microstructure on friction. Insert images were the laser microscope images of each samples.

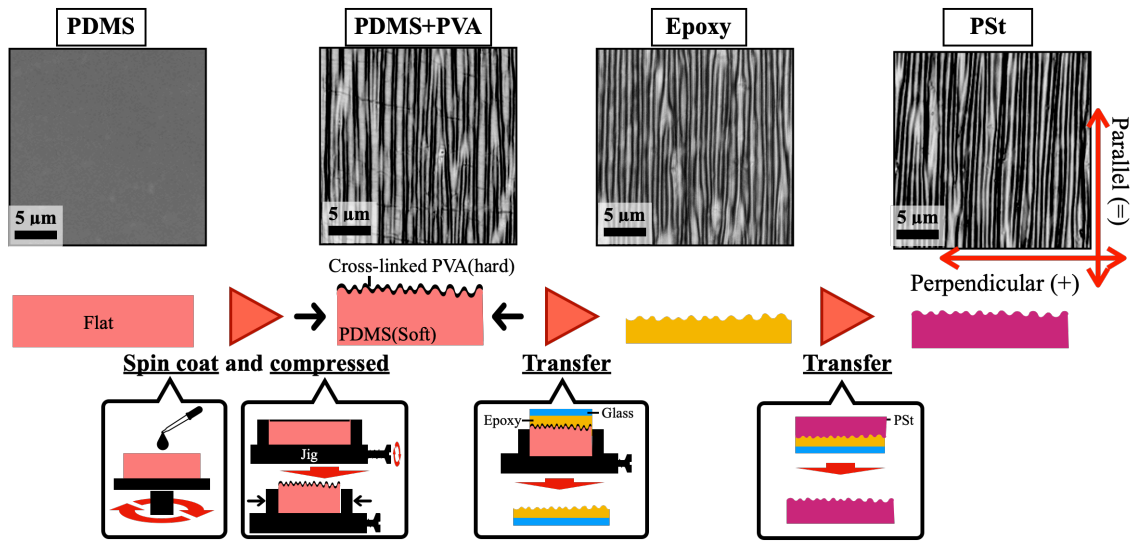


Fig. 3-7 Preparation of the PSt wrinkle structures. The wrinkles were formed by preparing a hard polyvinyl alcohol (PVA) layer on soft polydimethylsiloxane (PDMS), followed by compression. The inset images are the laser microscopy images of each sample. The red arrow indicates the sliding direction.

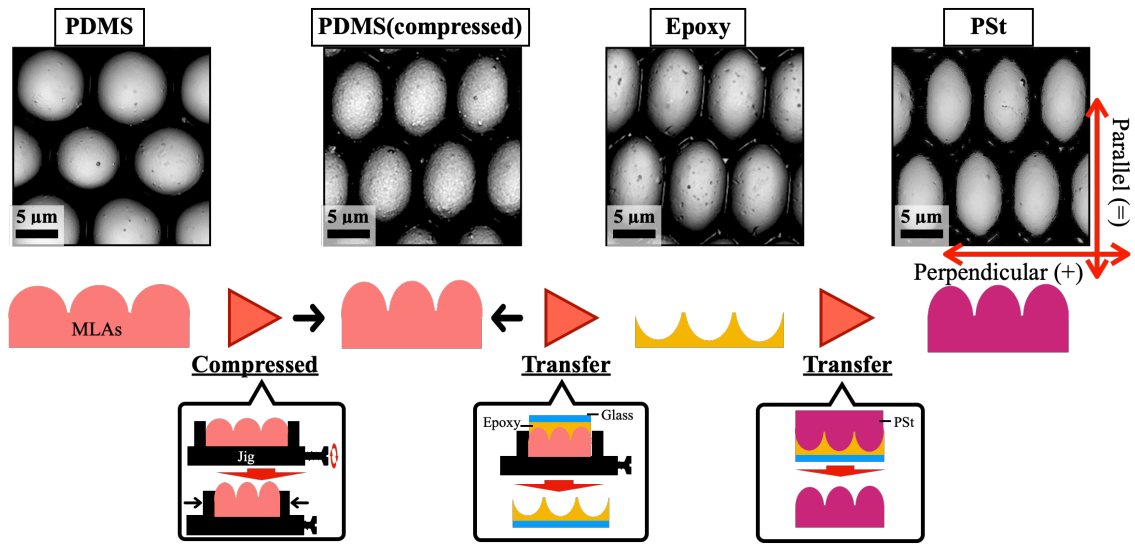


Fig. 3-8 Preparation of the PSt microlens array (MLA) structures. The MLAs used for the measurements were formed by compressing PDMS-MLAs with circular structures. PVA was not spin-coated. The inset images are the laser microscopy images of each sample. The red arrow indicates the sliding direction.

• Sample preparation

The PSt sample was prepared by dropping a PSt chloroform solution (100 g/L) onto an epoxy resin mold with the structure prepared in the same manner as that employed for the preparation of the reverse microstructures: letting it air dry and peeling it off (Fig. 3-6, Fig. 3-7, Fig. 3-8). The flat samples were prepared by dropping the solution onto a glass substrate and peeling it off after natural drying.

The structures of the prepared samples were observed and measured ($n = 20$) by a laser microscope (VK-X200, Keyence Corp., Japan). The prepared structures exhibited the following size: wrinkle ($1.39 \pm 0.31 \mu\text{m}$ diameter, $0.32 \pm 0.13 \mu\text{m}$ depth) and MLAs ($12.10 \pm 0.68 \times 9.80 \pm 0.29 \mu\text{m}$ diameter, $1.73 \mu\text{m} \pm 0.31$ depth); the mimic structure exhibited a combined size (wrinkle: $1.38 \pm 0.33 \mu\text{m}$, MLAs: $12.23 \pm 0.47 \times 9.77 \pm 0.33 \mu\text{m}$). The size was the mean value ($n=20$) for any structure and the number after $\pm$ was the standard deviation.

3-2-3 Friction Test

The frictional force of the sample was measured by sliding the sample in one direction with a constant load using TRIBOGEA (HHS2000, SINTO Scientific Co., Ltd., Japan). The employed indenter was a stainless steel ball (SUS304, AS ONE Corporation, Japan) with a 10 mm diameter ($R_a = 32.7 \text{ nm}$). The sliding speed for the friction measurement was 1 mm/s, and the load was 9.8 mN ($\approx 1 \text{ g}$). The friction measurements were performed four times per sample, and the average frictional force was calculated for measurement times ranging from 1000 to 5000 ms (μs). The measurement directions were divided into two types: those measured in the direction across the wrinkles or MLAs (perpendicular, +), and those measured in the direction parallel to the structure (parallel, =). This difference in the sliding direction did not apply to flat surfaces, and unless otherwise specified, the sliding direction was perpendicular. All error bars in the graph represented the standard deviation.

• Effect of structural shape on friction

The frictional force of the samples was measured by sliding the samples with a 9.8 mN load in one direction using a friction tester. To investigate the effect of an anisotropic structure on friction, the measurement direction was distinguished between the perpendicular (+) direction, which crossed the structure, and the parallel (=) direction, which was parallel to the structure (Figure 3). The measured frictional force and friction coefficient are shown in Figures 3(e, f). The friction force varied greatly with the shape of the microstructure, decreasing in the flat > wrinkle > MLAs > mimicked structure order regardless of the sliding direction, and the friction force on the mimicked structure surface was approximately ~50% lower than that on the flat surface with no microstructure. These results indicated that the mimicked structure exerted a friction-reducing effect that is greater than that exerted by a single-layer structure. Additionally, the frictional force of samples other than the flat structure was investigated by changing the sliding direction, revealing that the frictional forces of the wrinkle, MLAs, and mimicked structure were smaller in the perpendicular direction than in the parallel direction. The anisotropies of the frictional force in different sliding directions could be caused due to stiction length and stiffness, as revealed in previous studies¹²⁷. The paper shows that when the structure size is less than 10^{-3} of the indenter size, the anisotropy due to the stiction length becomes more pronounced, and our paper has the same scale ratio, since the b/R in the

original paper is around 10^{-4} to 10^{-3} . The microstructures in this study that exhibited anisotropy in frictional forces also had shorter stiction lengths because contacts perpendicular to the microstructure are shorter than contacts parallel to the microstructure. The stiffness, which is inversely proportional to the surface deformation, may be explained by the load dispersion that will be discussed later in the text. That is, different shapes have different deformation resistances and therefore different frictional forces. Of the two, the stiction length was similar to the difference in contact continuity that explains the anisotropy of friction forces in metal surface microstructures¹⁰⁸.

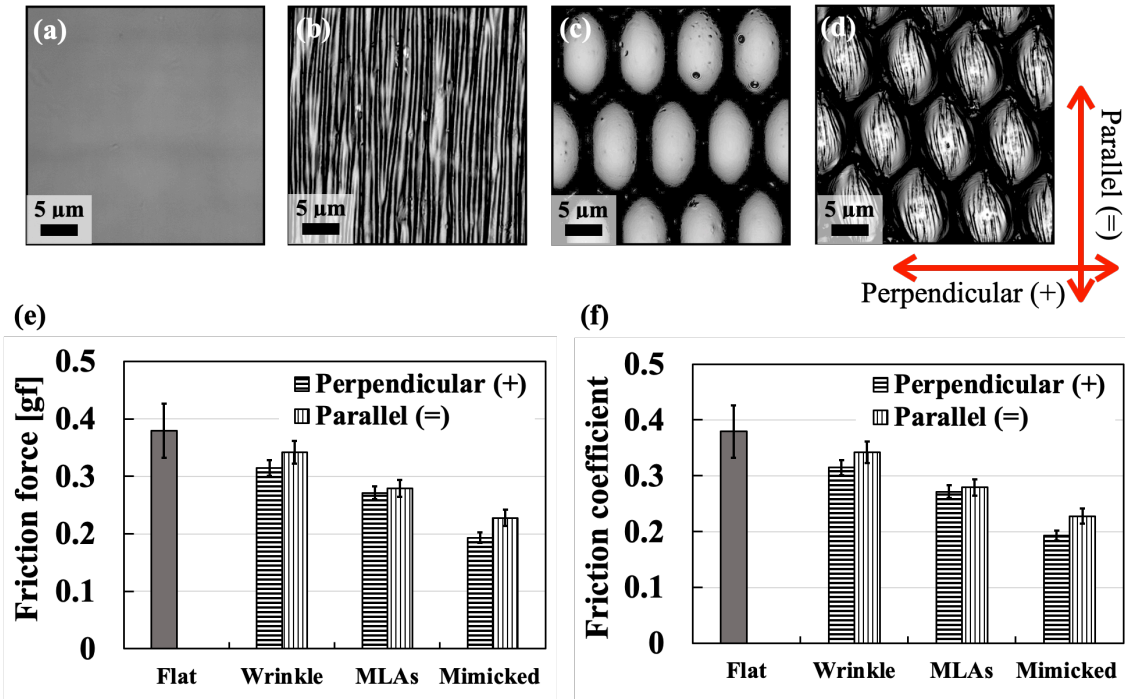


Fig. 3-9 (a-d) The laser microscope images, (e) the frictional force and (f) friction coefficient of (a) flat, (b) wrinkle, (c) MLAs, and (d) carion beetle mimicked structure. Load was 9.8 mN.

The friction coefficient was calculated from each dynamic friction force and load, and the dynamic friction force was the average of the friction forces at 1000-5000 ms. Red arrows show sliding direction (+ ; perpendicular, = ; parallel). All error bar were standard deviation.

3-3 Elucidation of the principle of friction

3-3-1 Simulation Model

A simplified friction model was considered to investigate the friction reduction of the hierarchical structure (Fig. 3-10). In this model, the frictional force was obtained by the product of the contact pressure generated by the virtual spring inserted at the interface and the friction constant. Put differently, the calculation was based on Coulomb's law, which considers the real contact area, which varies proportionally to the load, rather than the apparent contact area¹²⁸. The indenter of the same size as in the actual measurement was placed on top of the modeled microstructure sample used for the actual measurement ((Fig. 3-10 (i, j))). Fig. 3-10 (j) shows a magnified image of one part of the indenter and the PSt model. The frictional force, stress, and contact pressure applied to the PSt model when the indenter was slid laterally with a 9.8 mN load were calculated. The calculation results are shown in 3-3-2 onwards, and the detailed methods and principles are given below.

• Details of the simulation model and calculation method

Finite element method (FEM) simulations were performed using COMSOL Multiphysics with the structural mechanics module (COMSOL Multiphysics 6.1, COMSOL Inc., USA) running on Windows 10 Pro (Ver. 1909). The mimicked-structure model was prepared by arranging hemispheres (diameter: 11 μm) in a row horizontally on a rectangle as an MLAs structure, and with hemispheres (diameter: 1.4 μm) arranged as a wrinkled structure on the circumference of the MLAs (Fig. 3-10 (d)). The MLAs structure was prepared by arranging only 11 μm diameter hemispheres in a rectangular pattern (Fig. 3-10 (c)), and the wrinkle structure was prepared by arranging only 1.4 μm diameter hemispheres in a rectangular pattern (Fig. 3-10 (b)). The rectangle and hemisphere corresponding to the PSt parts were all integrated, leaving no interface. The rectangle and hemisphere that corresponded to the PSt parts were completely combined (union). The flat structure does not have hemispheres lined up but consists only of rectangles (Fig. 3-10 (a)). The thickness and width of the rectangle were 30 and 66 μm , respectively, but the wrinkle structure was set to 65.8 μm to avoid differences in calculations owing to gaps and edges without structures (47 hemispheres with a diameter of 1.4 μm are arranged without any gaps, and it has been confirmed that this size difference exerted almost no effect). Considering the influence of curvature, the indenter

was set to 10 mm, the same as the actual indenter, and only a portion was cut out or hidden when displaying the results and model (Fig. 3-10 (j)). The indenter was placed 10 μm to the left from the center of the PSt structural model and adjusted vertically so that the lower apex of the indenter and upper apex of the structural model were at the same height. The material properties of the stainless steel indenter and structural model were obtained from the COMSOL material library, and the calculation mesh size was adjusted to suit the calculation. The mesh model is shown in Fig. 3-10 (e-h). The contact model in COMSOL was used as a model for calculating the frictional force generated at the indenter and structure interface. This model defined the contact by inserting a very thin spring at the interface. In this method, the frictional force was calculated in a simplified manner using the product of the contact pressure generated by the interface spring and the friction constant (constant). Put differently, the focus was not on the apparent contact area; rather, it was on the real contact area, which varied depending on the load and adhesive force generated at the interface. Therefore, the calculation was based on the Coulomb law ¹²⁸. The parameters in the contact model were entered with values that were adjusted to ensure accurate calculations (Contact pressure penalty factor (P_n): $5\text{e}3$ [N/mm³], Friction force penalty factor (f_t): 0.01 (Automatic, Soft), Spring foundation (k_{tot}): $7\text{e}5$ [N/m]). The penalty method was chosen for the contact model. The frictional force was calculated by fixing one part of the model to ensure correct calculations, applying a load from the top of the indenter, and sliding the indenter 20 μm laterally (Fig. 3-10 (j)). A spring foundation was used for fixing the indenter to stabilize the calculations^{129,130}. The frictional force displayed in the graph is the average value of the obtained frictional force. The calculated friction was the steady state friction. The contact force in the graph was calculated by summing all contact pressures applied to the structure surface during sliding. The contact force and reaction force (y component) are normalized to the flat value. (so the ratio of flat is 1). In other words, the contact force is the product of the contact pressure and the contact area. The contact pressure and stress during sliding, which were not total values, were shown at a sliding distance of 14 μm .

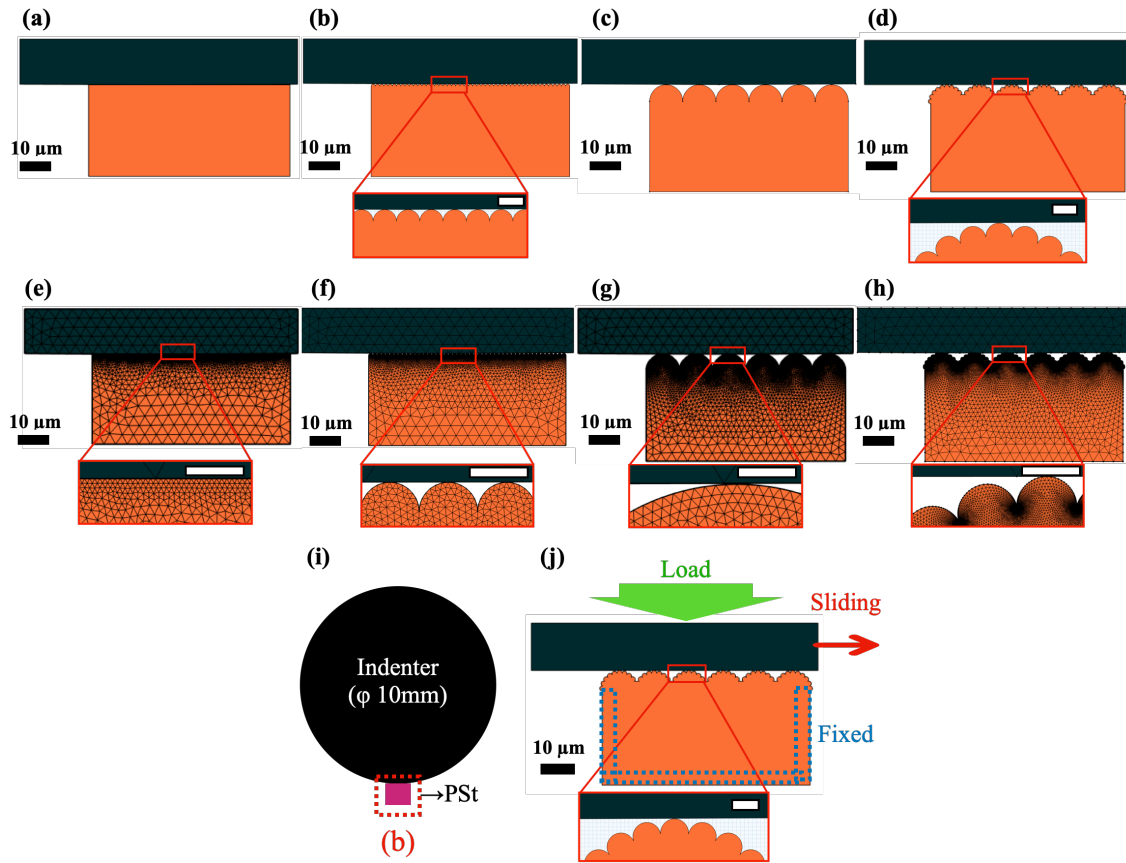


Fig. 3-10 (a, b, c, d) Simulation and (e, f, g, h) meth models of (a, e) flat, (b, f) wrinkle, (c, g) MLA, (d, h) mimicked structures. The images with red borders represent the magnified views, and the scale bar is 1.4 μm. Only a portion of the indenter is shown. 4 Simulation model and calculation results. (i) Schematic diagram of the simulation. PSt to indenter ratio was not accurate. (j) Simulation model of mimicked structure. Insert image was magnified image. Only a portion of the indenter was shown. White scale bar was 1.4 μm.

The definition of contact and friction in COMSOL, such as contact pressure, is described in the following URL.

COMSOL Documentation, Contact Analysis Theory
https://doc.comsol.com/6.0/docserver/#!/com.comsol.help.sme/sme_ug_theory.06.73.html

From the above, the contact pressure (T_n) is calculated in COMSOL using the following formula:

$$T_n = \begin{cases} -p_n g_n + p_0 & \text{if } g_n < p_0 / p_n \\ 0 & \text{else} \end{cases} \quad \dots (10)$$

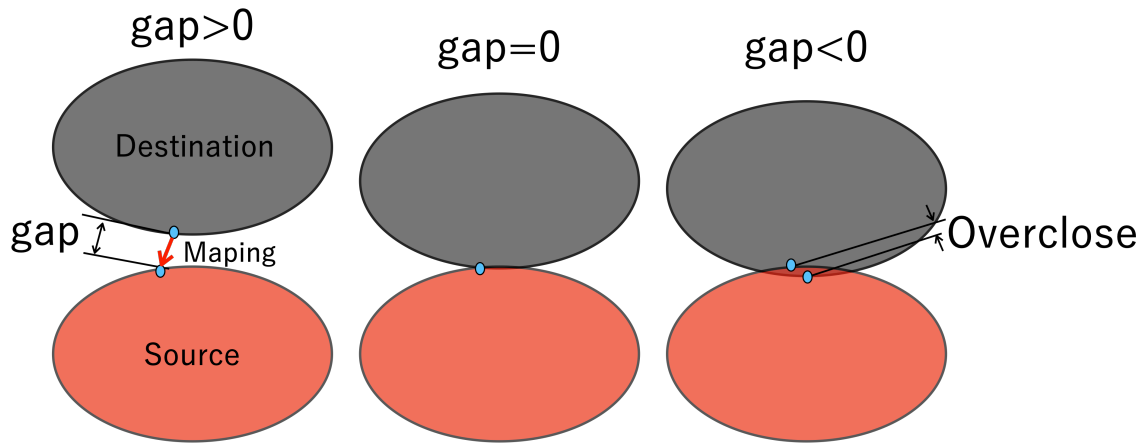


Fig. 3-11 Schematic diagram of contact in comsol.

p_n is the penalty coefficient (default is Young's modulus of the material, if it is high it is more accurate but difficult to calculate, if it is low it is less accurate but easy to calculate¹³¹), p_0 is the pressure when the gap between two points of the two models is zero as shown in Fig. 3-11. In the default and our case of $p_0 = 0$, there must always be some overclose for contact pressure to occur. In other words, the indenter model penetrates the structural model, albeit slightly, because the appropriate parameters are inserted. The above equation is an approximate solution to the non-penetration condition. g_n is the physical gap function, which is represented by the geometric distance between the source and destination boundaries as shown in the figure, and the offset at the source and destination interfaces. The offset varies along the shape. This distance is the shortest distance that a point can reach on the opposite model by mapping it to a certain direction in the space frame (red arrow emanating from the light blue point in the figure), not necessarily the shortest distance between the interfaces of the two models. Therefore, the shape of the geometry may affect these. The penalty method defined in the above equation

is conceptually based on inserting a nonlinear spring between the contact points. In other words, in this case, the contact pressure does not change directly due to the load, but changes due to the change in the indenter position (overclosure) caused by the load. Due to the conceptual existence of a nonlinear spring and the relationship between the distances in this calculation, the calculation results in a change in contact pressure even when the same load is applied. The important thing is that these stresses and reaction forces are calculated completely separately from the contact pressure, and the calculation formulas for stress etc. do not change whether there is a spring on the surface as in this case, or whether there is no spring and the surfaces are completely bonded.

3-3-2 Simulation results

The calculated frictional force is shown in Fig. 3-12 (a, b). The calculated frictional force correlated well with the actual measured frictional force, decreasing in the flat > wrinkle > MLAs > Mimicked order. The frictional force in this calculation was determined by the product of the friction constant and contact pressure applied to the indenter and the model interface. As the friction constant was set to the same value for all structures, the applied pressure to the model have changed by the surface microstructure. Fig. 3-12 (c) shows the results of the calculation of the deformation stress applied during sliding. A larger stress concentration was observed at the tip of the structure compared with the flat surface owing to the microstructure. In the flat model, stress attenuates in proportion to the distance. However, in the model exhibiting a surface microstructure, significant stress was applied to the base of the structure far from the contact point. Particularly, the mimicked structure exhibited widely dispersed stresses in the upper wrinkled structure and lower MLAs structure. This difference in stress dispersion may indicate that the forces acting on the surface, which are prone to deformation due to pressure concentration, have changed depending on the structure. Fig. 3-13 (a, b) show the applied contact pressure to the interface during sliding (at 14 μm from the initiation of sliding). The abscissa shows the coordinate of the PSt model surface. The PSt model was set up so that the center of the PSt model was at 0. The applied contact pressure by each structure was different, with the flat structure having the lowest maximum value, followed by the wrinkle and MLAs structures; the mimicked structure having the highest maximum value. Contrarily, the stress range was smallest for the mimicked structure and increased in the following order: MLAs < wrinkle < flat. If the total value of this contact pressure (the colored part in Fig. 3-13 (b)) was not changed based on the structure, the frictional force would be unchanged. However, force calculations normalizing the total contact pressure on the surface to the flat revealed a different ratio on the shape of the structure even when the same load was applied (Fig. 3-13(c) total contact force). The total contact force decreased in the following order: flat > wrinkle > MLAs > mimicked, similar to the actual measured and calculated friction force results, and the ratio of decrease corresponded well with the amount of decrease in friction force. Conversely, in contrast to the contact force, which looked at the force applied only to the interface, the reaction force applied to the entire PSt model was constant regardless of the structure; therefore, the structure may have reduced the applied

force to the surface by dispersing the force to locations other than the surface (Fig. 3-13 (c), reaction force rate). Differences were observed in the stress dispersion based on the structure (Fig. 3-12). Stress dispersion to the bottom and sides of the structure avoids stress concentration on contact surfaces where deformation tends to occur. As a result, it is harder for the material to reach the stress required for deformation, which is thought to

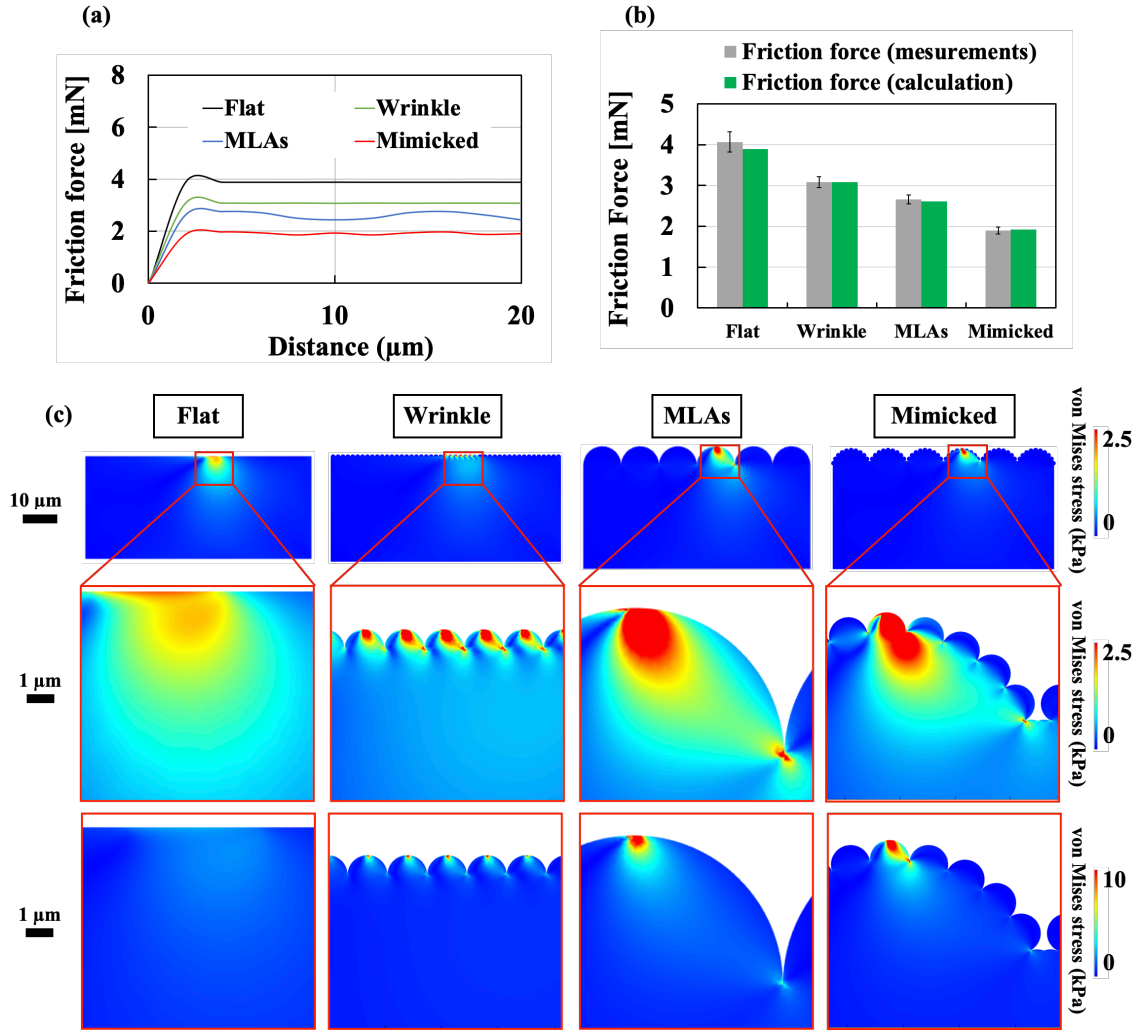


Fig. 3-12 (a) the graph of calculation friction force vs sliding displacement curve. (b) Calculation and measurements results of friction force. (c) Simulation results showing deformation stress applied during sliding. The indenter was omitted. The top row were an overall view, and the bottom two rows surrounded by a red frame were magnified views. The two magnified images had different scale bar ranges, with the upper one being up to 2.5 kPa and the lower one being 10 kPa (see left side of the figure). On the right side of the images were a scale bar that shows the size of each row. All error bar were standard deviation.

suppress deformation and real contact area increase and reduce friction. In addition, when

looking at the direction of stress applied to the surface, there are arrows at the base of the structure that repel load and sliding, and the mimicked structure was seen in both the wrinkled structure and the MLAs structure (Fig. 3-14). Therefore, by distributing the stress to the bottom and sides of the structure, it is possible that stress concentration on the contact surface where deformation is likely to occur was avoided. As a result, it is thought that the material is less likely to reach the stress required for deformation, leading to an increase in the effective contact area and a reduction in friction.

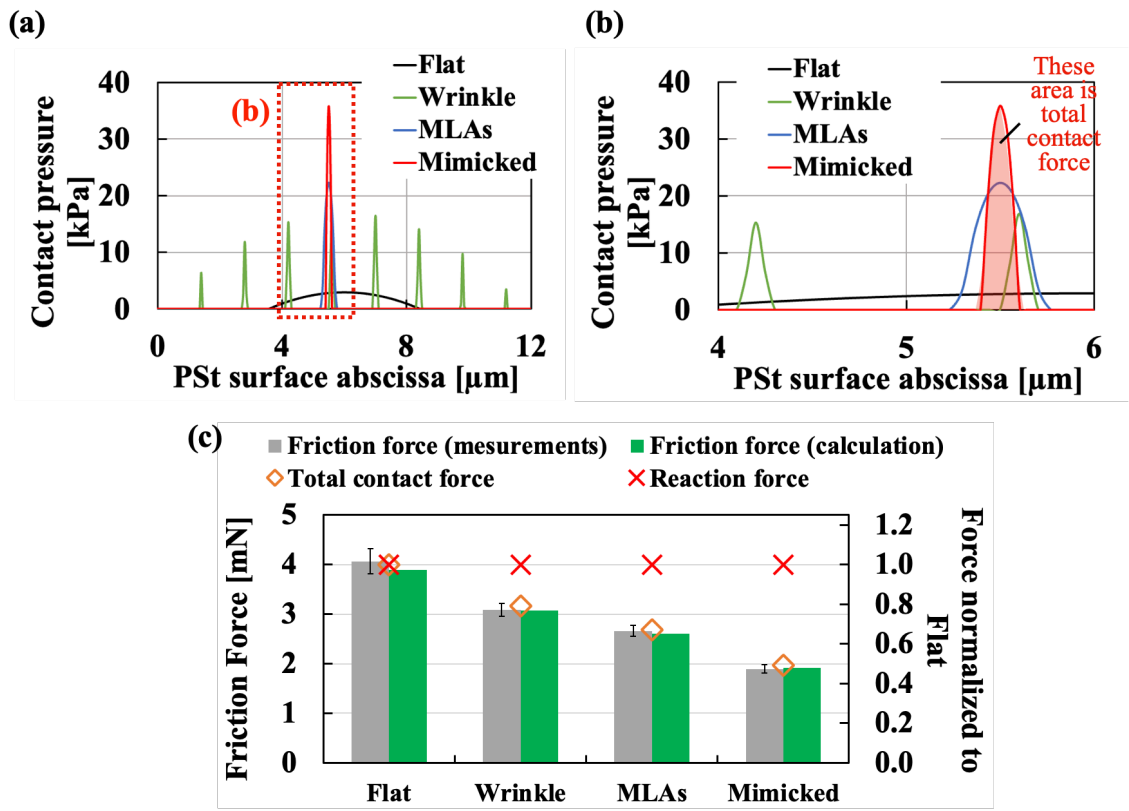


Fig. 3-13 (a) Contact pressure during sliding (when the tip of the indenter was at 6 μm on the horizontal axis of the graph) and (c) measured and calculated friction force results (left vertical axis) and ratio of contact force pressure and reaction force to flat (right vertical axis). The contact force and reaction force are normalized to the flat value. (a, b) The horizontal axis of the graph was the coordinate, and 0 μm was the center position of the PSt model. (b) was an magnified view of (a) (the area surrounded by the red dot shown in (a)). Total contact force pressure was the area of pressure applied to the surface as shown in (b). The reaction force was the total force applied to the PSt model as shown in Figure 4 (d). All error bar were standard deviation.

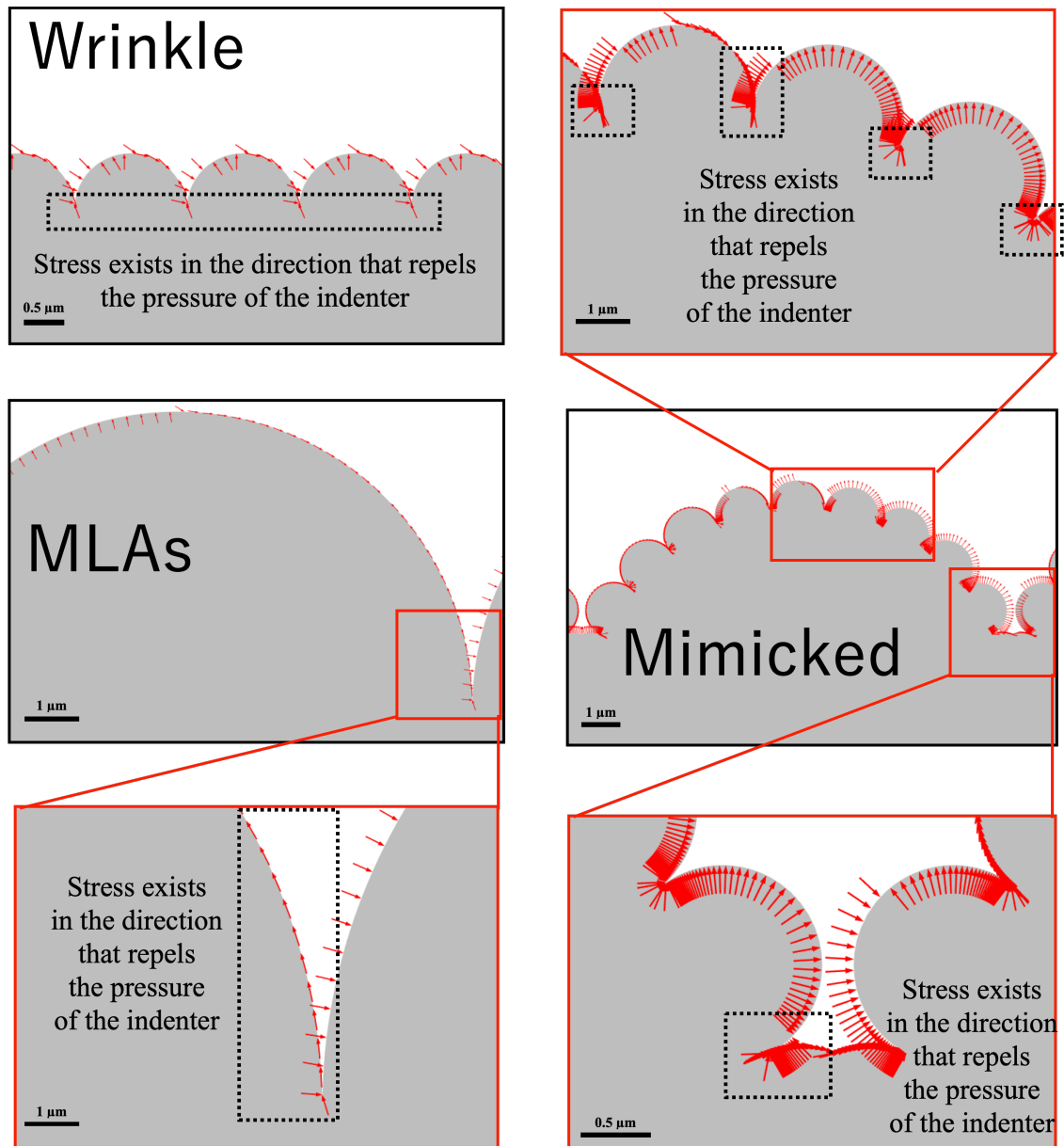


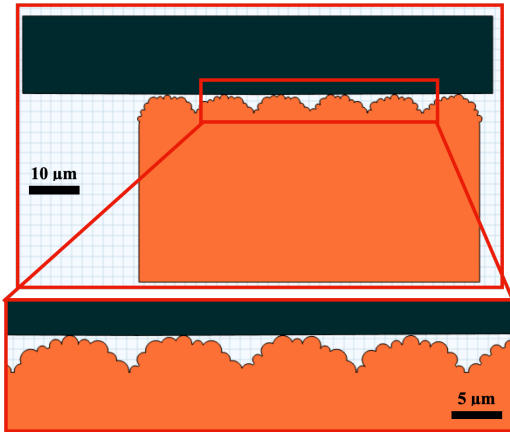
Fig. 3-14 The stresses acting on each structure during sliding (principal stress directions 3) are indicated by arrows.

These results indicate that the hierarchical surface microstructure mimicking *Necrophila japonica* significantly reduces friction compared to flat and single-layer microstructured surfaces. The friction-reduction effect by the mimicked structure can be probably explained by the load-dispersion effect. There are many factors that affect friction force, but in dry friction such as the one we conducted, it has been reported that the adhesive force acting on the real contact area between the indenter and the friction surface has a large effect on the friction force¹⁰⁹. In the case of the contact between hard

plastic deformation materials, the real contact area is extremely small compared with the apparent contact area owing to the influence of roughness on the surfaces. When the load is increased, pressure is concentrated on the roughness and deformation increases the real contact area and frictional force ^{132–134}. In other words, the deformation of the roughness in proportion to the load is the basis of normal friction. Conversely, the simulations revealed that the microstructure reduced the contact area and concentrated the pressure at the tip of the structure. The decrease in the contact area was related to the structure shape, with the area where the mimicked structure contacted the indenter being the narrowest and the pressure applied to the model surface being the highest (Fig. 3-13(a, b)). In such a case, the apparent and real contact areas may almost be the same at the tip of the microstructure ¹¹⁷ (microcontact). Artificial and natural rough surfaces of comparable dimensions to the surface structures substantially reduce the frictional force on the surface in contact with the insect legs ^{116,117}. Gorb et al. suggest that this reduction in frictional force is due to a decrease in the real contact area, which can be calculated as the sum of distances between all virtual contact points on the nondeformable terminal plate and substrate ¹¹⁷. Indeed, the observed trends in calculated and measured values align consistently, underscoring the significant variation in frictional force contingent upon structure size ¹¹⁷. From these results, it is apparent that contact on the surface of the surface microstructure is primarily concentrated at the tip, where the apparent and real contact areas are nearly identical. According to the Amonton–Coulomb law, even after microcontact, the frictional force will not change with the structure if the real contact area increases owing to the deformation of the microstructure. However, in the actual and simulated measurements, the frictional forces decreased by the microstructure (Fig. 3-12 (a, b)). Notably, although the reaction force, which is the force applied to the entire model, was the same for all structural models, the contact force applied to the structure-indenter interface decreased with flat, wrinkle, MLAs, and mimicked, as did the friction force (Fig. 3-13(c)). Despite applying the same load, the fact that the total contact force applied to the surface prone to stress concentration was small indicated that deformation and increase in real contact area was difficult to occur on the actual surface. The reason for the decrease in contact pressure was that the load was dispersed to areas other than the surface, as seen in bridges and metamaterials. In a sample exhibiting a microstructure, the stress was dispersed to the roots of the structure without attenuation proportional to distance, which reduced the contact pressure (Fig. 3-12 (c)). Additionally, although the

stresses in the wrinkle and MLAs structures were dispersed within and at the root of each structure, the stresses in the hierarchical mimicked structure were widely dispersed in the upper wrinkle structure and lower MLAs structure (Fig. 3-14). Even when checking the direction of stress, arrows were present in the direction of repulsion against load and sliding at the base of the structure, and in the mimicked structure, a repulsive force was widely observed in both the wrinkled structure and the MLAs structure (Fig. 3-14). For the above reasons, the mimicked structure had the strongest pressure concentration due to the reduction in contact points (Fig. 3-13(a, b)), but the frictional force decreased the most with the difference in apparent contact area because the deformation (large increase in real contact area) was suppressed by effective load dispersion. Although the conditions in our study are significantly different, these results are similar to those of studies showing behavior that differs from the Amontton-Coulomb law in under low load conditions where even plastic deformation materials only undergo elastic deformation¹⁰³. The region where deformation is difficult can be extended by the surface microstructure, and this may be more effective with a hierarchical structure than with a single layer structure. In our case, the indenter was sufficiently large in relation to the size of the structure to reduce the frictional force due to the simple reduction of the contact points. If the indenter and microstructure sizes were closer, the frictional force may increase owing to the interlock caused by the penetration of the structures by the indenter^{109,110,135}. Conversely, if the hierarchical structure was used, the contact point might be lowered without increasing the gap to penetrate. As a supplementary note, the friction reduction effect was also confirmed in a random-sized model close to the actual mimicked structure, and the impact of the non-uniformity of the structure was small (Fig. 3-15). The random mimicked structure model shown in Fig. 3-15 has wrinkles of random size in the range of 1 to 2 μm . The range of 1 to 2 μm was chosen because the measurement results of the size of the wrinkles on the top of the mimicked structure that was actually created were in the range of 1 to 2 μm . The calculation results did not differ significantly from the actual measurement results, and were within the error range. We believe that the slight difference is due to the difference in contact area caused by the difference in structure size.

(a)



(b)

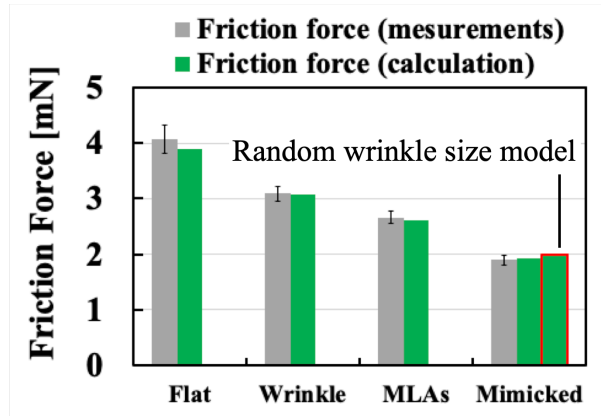


Fig. 3-15 (a) Mimicked structure model with random wrinkle sizes and (b) calculation results.

3-4 Application of the principle of friction reduction through load dispersion

3-4-1 Creating a surface that can reduce friction even under high loads using metallic materials

The above research has shown that the robustness of the structure itself is effective in reducing friction. This robustness can be increased by creating a hierarchical structure, but depending on the physical properties of the material itself, it may be destroyed when the load is increased, and the friction reduction effect may be lost. In fact, when the load on the sample measured in 3-2-3 was increased, the friction coefficient increased and became the same as that of a surface without a structure (Fig. 3-16 Friction coefficient of PSt carrion beetle mimicked sample for each load). This is thought

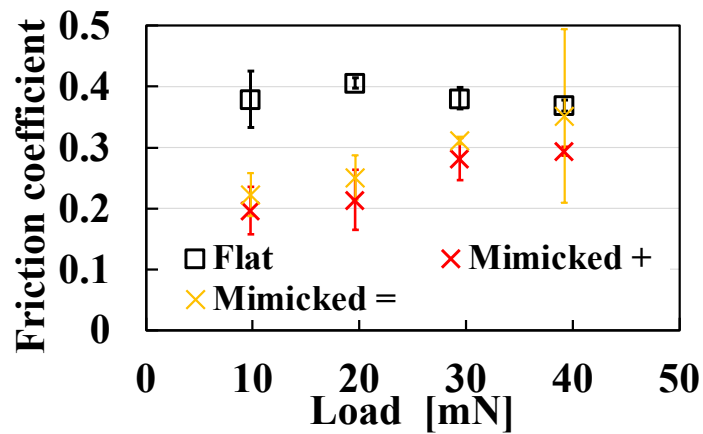


Fig. 3-16 Friction coefficient of PSt carrion beetle mimicked sample for each load

to be due to the destruction of the structure.

Conversely, by combining a structure with a hard material such as metal, friction in the high load range may be reduced. Although there are few species of metallic materials that are used by living organisms, they are very important materials for humans and are still used in various places around the world. If the friction reduction principle of this study could be applied to metallic materials, the weakness of microstructures, that is, loss of function due to destruction of the structure, could be overcome, and it would be possible to apply it to various fields. Therefore, we focused on the moth-eye structure mentioned in 3-1-4. The moth-eye structure is a structure found in the eyes of moths and the wings of cicadas, and is known to exhibit non-reflective properties due to the gradual change in refractive index caused by the structure, as well as super-water repellency and anti-fogging properties. In addition, as mentioned in 3-1, the moth-eye structure has been shown to have low insect adhesion, so it is highly likely to have a friction reduction effect.

The moth-eye structure has been commercialized as MOSMITE™®¹³⁶ by Mitsubishi Rayon Co., Ltd. (currently Mitsubishi Chemical Co., Ltd.). MOSMITE™® is not made by conventional photo-nanoimprinting or top-down microfabrication methods, but by self-organization in which aluminum is electrolyzed in an acid electrolyte (anodic oxidation) to create a regular pore structure, so it is possible to produce a large-area film with low energy. We used MOSMITE™® as a mold and performed electroforming to fabricate a metal moth-eye structure, and investigated its friction reduction effect under high load conditions.

3-4-2 Fabrication of metallic moth-eye structures

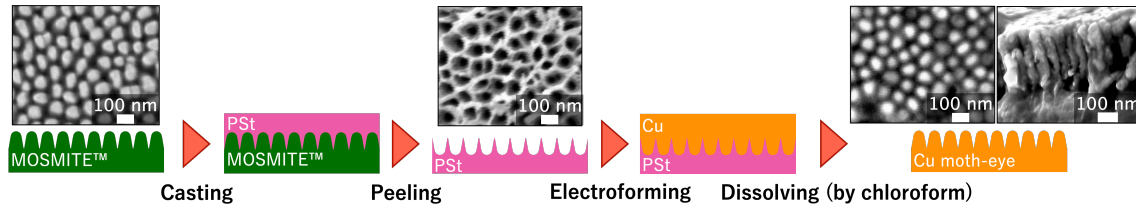


Fig. 3-17 (a) Preparation procedure of the Cu moth-eye structures. Cu moth-eye structures were prepared via electroforming, employing a polystyrene (PSt) reverse structure as a template. The insets depict FE-SEM images, with the left panel showcasing a top view and the right side illustrating a cross-sectional view of the Cu moth-eye

In this chapter, we describe the preparation method of the Cu sample (Fig. 3-17). The Cu moth-eye structure used in the measurement went through a two-step transfer process. First, a solution of 100 g/L of polystyrene (PSt, average molecular weight by GPC about 280,000) dissolved in chloroform (FUJIFILM Wako Pure Chemical Industries, Ltd., Japan) was evaporated onto the surface of MOSMITE™[®] provided by Mitsubishi Chemical Corporation. The solution was then air-dried and carefully peeled off to obtain an inverted (hole) moth-eye structure. Next, platinum-palladium was sputtered onto the PSt hole structure using an ion sputter (E-1030, Hitachi High-Tech Corporation, Japan) to impart conductivity and enable electroforming, after which the sample was attached to a cathode-side electrode and immersed in an electrolytic copper sulfate plating solution (Kiyokawa Plating Industry Co., Ltd., Japan). At the same time, a copper plate (1-126-0111, KENIS, LTD., Japan) with an anode side electrode attached was immersed in the same solution, and a current of 0.1 A was applied from a power source for 60 min to perform electroforming at room temperature. Subsequently, chloroform was used to dissolve PSt, and a Cu moth-eye structure was obtained. A conductive glass substrate was used as a mold for electroforming to prepare a flat structure without microstructures. The obtained samples were inspected and measured using a field emission scanning electron microscope system (FE-SEM, SU8230, Hitachi High-Tech Corporation, Japan). In addition, surface composition analysis was performed using an energy dispersive X-ray spectrometer (EDS, QUANTAX, Bruker, Japan) and an X-ray photoelectron spectrometer (XPS, JPS-9200, JEOL, Japan) to confirm the chemical nature of the Cu sample surface. Observation with a field emission scanning electron microscope (FE-SEM) revealed that the height of the protrusions of the moth-eye structure was about 200

nm, the width of the tip was about 70 nm, and the width of the base was about 120 nm. In addition, using the same method as the moth-eye structure, a flat structure without fine structures was fabricated by electroforming on a conductive glass substrate. EDS and XPS analyses confirmed that the fabricated sample was a simple copper oxide (Fig. 3-18).

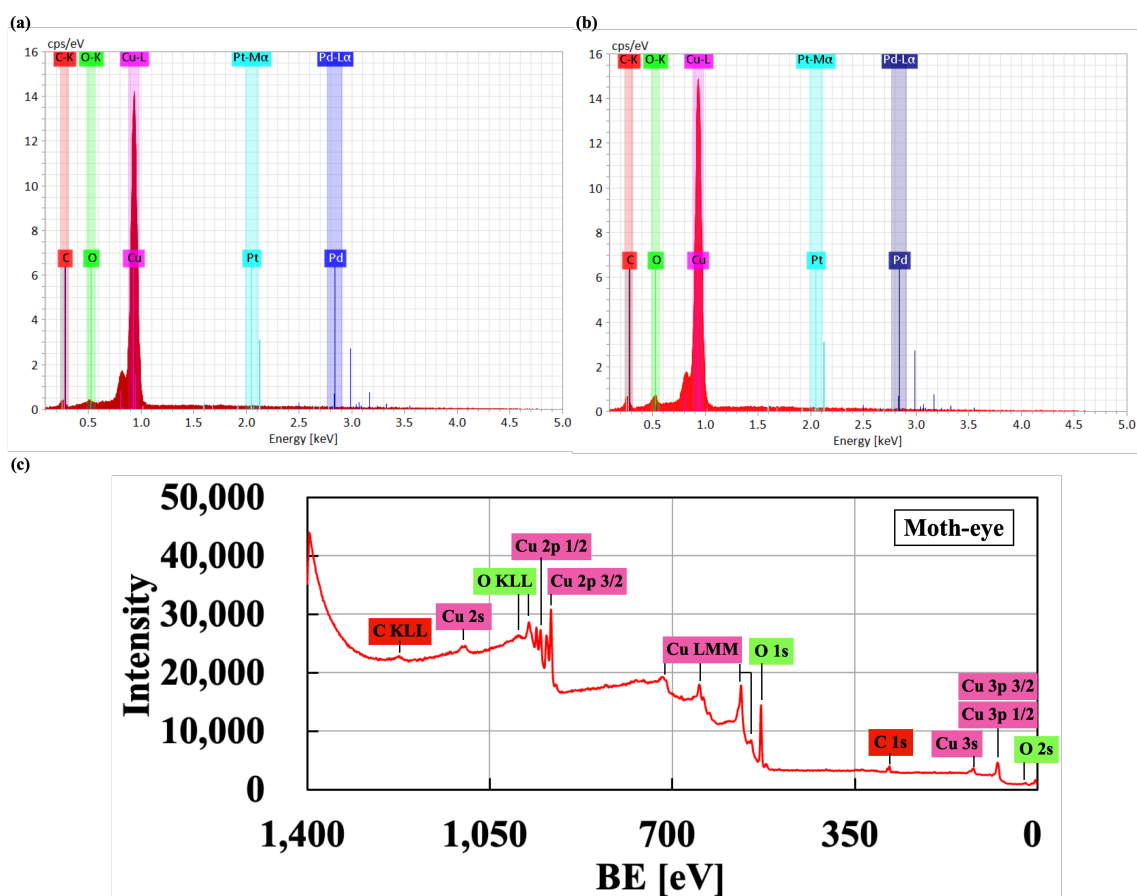


Fig. 3-18 (a, b) EDS and (c) XPS spectra of the Cu (a) flat and (b, c) moth-eye surfaces

3-4-3 Friction test results

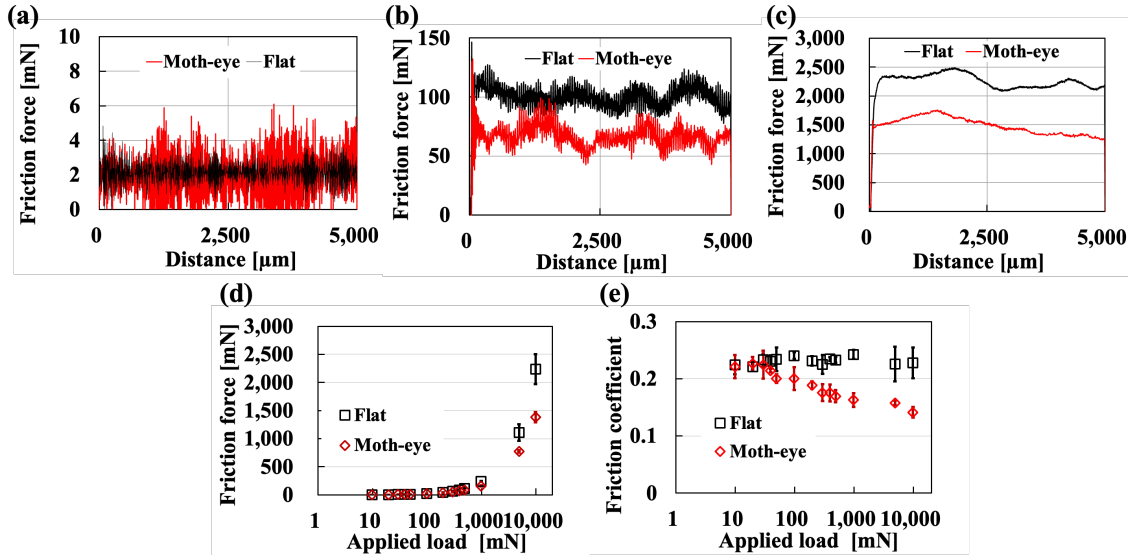


Fig. 3-19 (a, b, c) The frictional force of the Cu flat and moth-eye structures during sliding under varying loads of (a) 9.8 mN, (b) 490 mN, and (c) 9.8 N. (d) Friction force and (e) friction coefficient for each load (the horizontal axis was logarithmic). All error bars are S. D.

To investigate the effect of the moth-eye structure on friction, the prepared Cu samples were slid in one direction using a friction tester within a load range of 9.8 mN to 9.8 N, and the friction force was measured. Fig. 3-19 shows the measured friction force. The presence of the moth-eye structure and the applied load had a significant effect on the friction force. In the load range of 9.8–29.4 mN, there was no discernible difference in the friction force between the flat and moth-eye structures (Fig. 3-19 (a, d, e)). However, when the load exceeded 39.2 mN, the friction force no longer increased linearly with load in the moth-eye samples. At the same time, the friction coefficient increased exponentially, in contrast to the flat samples, where the friction force changed linearly with load and the friction coefficient decreased constantly (Fig. 3-19 (d, e)). Even under a load of 9.8 N, many moth-eye structures showed only minimal damage (Fig. 3-20). These findings highlight the friction-reducing effect of the moth-eye structure under high-load conditions. The real contact area is important in determining the friction force in dry friction such as that investigated in this experiment^{109,110}. For elastically deformable materials, the real contact area does not increase linearly with load, and the friction coefficient decreases exponentially even for flat surfaces^{98,104,105}. Our results show a similar trend for the moth-eye structure, where the real contact area does not increase linearly with load.

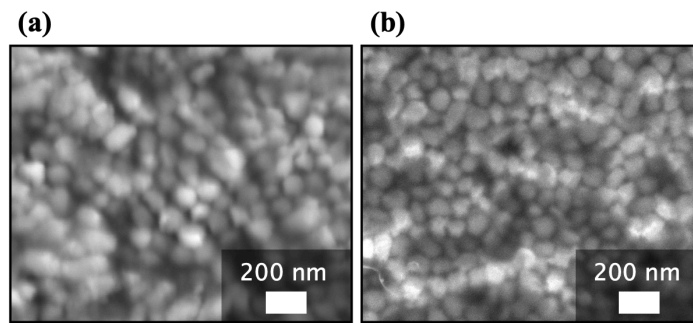


Fig. 3-20 FE-SEM images of the Cu moth-eye surfaces (a) before and (b) after friction measurements with a load of 1000 g

3-4-4 Simulation results

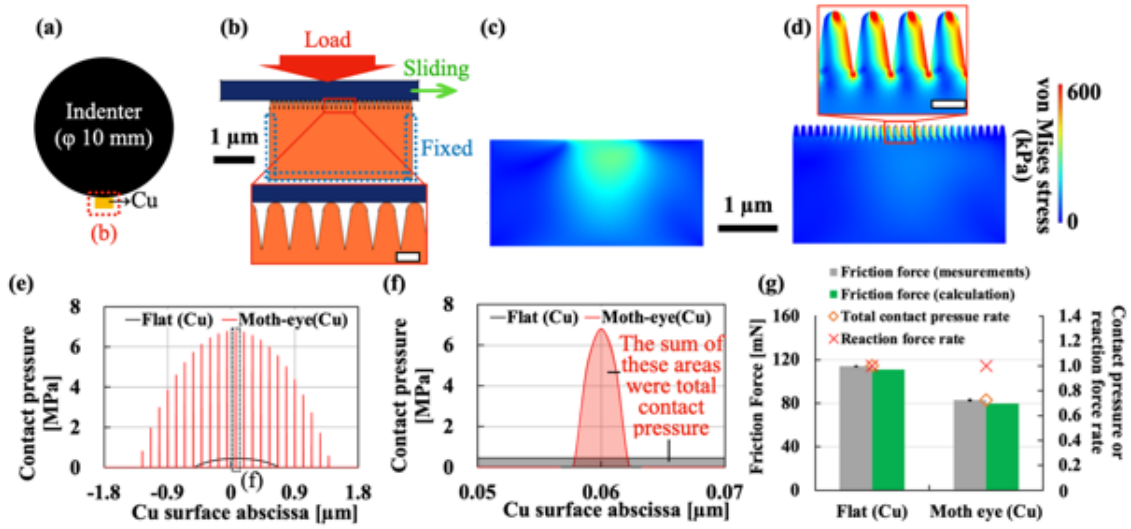


Fig. 3-21 (a) Schematic representation of the simulation process. (b) Simulation model of the moth-eye structure. The inset image provides a magnified view, focusing on a portion of the mode. Simulation results showing (c, d) deformation stress and (e, f) contact pressure during the sliding of the (c) flat and (d) moth-eye structures. The inset image provides a magnified view, with a white scale bar denoting 100 nm. (c, d) The indenter is not shown. (e, f) The horizontal axis of the graph represents the coordinate system, with 0 μm indicating the central position of the Cu model. Panel (f) is an enlarged view of (e) (the area surrounded by the red dot shown in (e)). (g) Measured and calculated friction force results (left vertical axis) and the ratio of total contact pressure and reaction force to the flat (right vertical axis). The total contact pressure is the pressure applied to the surface, as shown in (e, f). The reaction force is the total force applied to the Cu model.

Experimental measurements have confirmed that the presence of moth-eye structures reduces the friction force, making it difficult to increase the real contact area on surfaces featuring such structures. Therefore, in this section, we discuss the factors that hinder the increase in real contact area due to moth-eye structures. To facilitate this investigation, a simplified friction simulation model was developed, as shown in Fig. 3-21. In this model, the friction force is calculated by multiplying the contact pressure generated by an ultrathin virtual spring inserted at the interface with a fixed friction constant. Essentially, this calculation is consistent with Coulomb's law, which considers the real contact area to be independent of the apparent contact area and to vary directly proportional to the applied load¹²⁸. Simulation and Methodology A detailed schematic of

the model is shown in Fig. 3-22. The simulation model was constructed by placing a stainless steel ball indenter, whose dimensions match those of the actual measurements, on top of a flat moth-eye structure model. Fig. 3-21(b) shows a close-up view of the indenter and the cross section of the Cu model. The side and bottom of the Cu model are fixed, and the indenter is slid laterally under a load of 490 mN. The stress, contact pressure, and friction force are calculated.

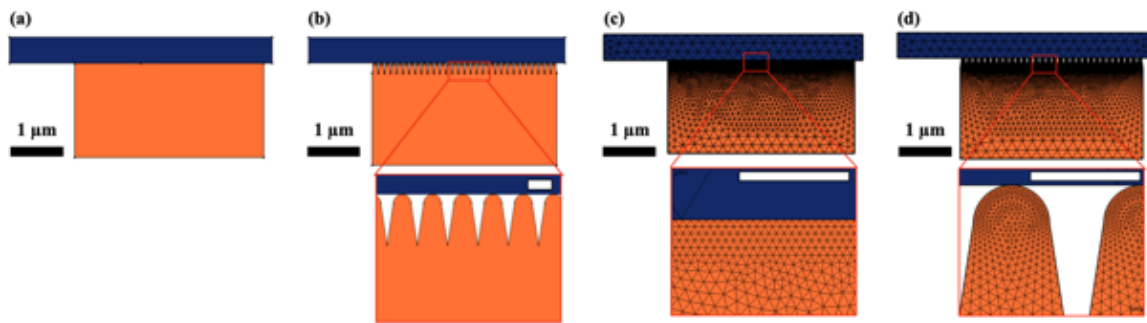


Fig. 3-22 (a, b) Simulation and (c, d) mesh models of (a, c) the flat and (b, d) moth-eye structures. Only a portion of the indenter is shown. Magnified images are provided in insets. The white scale bar represents 100 nm.

The calculation results are shown in Fig. 3-21 (c–g). In the planar structure, the contact points were wide and the stress was distributed uniformly (Fig. 3-21 (c)). On the other hand, in the moth-eye structure, the stress dispersion was non-uniform, concentrating at the tips of the structure protrusions and dispersing toward the sides and base (Fig. 3-21 (d)). The contact pressure acting on the surface during sliding showed a significant difference with and without the moth-eye structure. The planar structure showed various contact pressures resembling a mountain shape with peaks coinciding at the tip of the indenter, but the moth-eye structure had a much stronger contact pressure peak at the tip than the planar structure (Fig. 3-21 (e)). Notably, the stress acting on the tip of the moth-eye structure was mainly on the nanometer scale (Fig. 3-21 (f)). The calculated friction force of the moth-eye structure was smaller than that of the flat structure and was almost consistent with the measured friction force (Fig. 3-21 (g), friction force). This calculated friction force was derived from the product of the friction constant and the contact pressure. Considering that the friction constant was constant between the structures, the change in the total contact pressure of the surface was caused by the presence of the moth-eye structure. This total contact pressure represented the sum of the values, as shown in Fig. 3-21 (f). Analysis of the total contact pressure and its

ratio on the Cu surface revealed that, despite the same load, the moth-eye structure exhibited a lower total contact pressure than the flat surface. Correspondingly, both the friction force and the contact pressure ratio were consistently reduced (Fig. 3-21 (g), total contact pressure ratio). These results suggest that the structure is less likely to deform on the real surface. The reduced deformation results in a smaller real contact area, which contributes to the reduction of friction. Therefore, the simulation results confirm that the moth-eye structure reduces the friction force by reducing the real contact area compared to the flat surface.

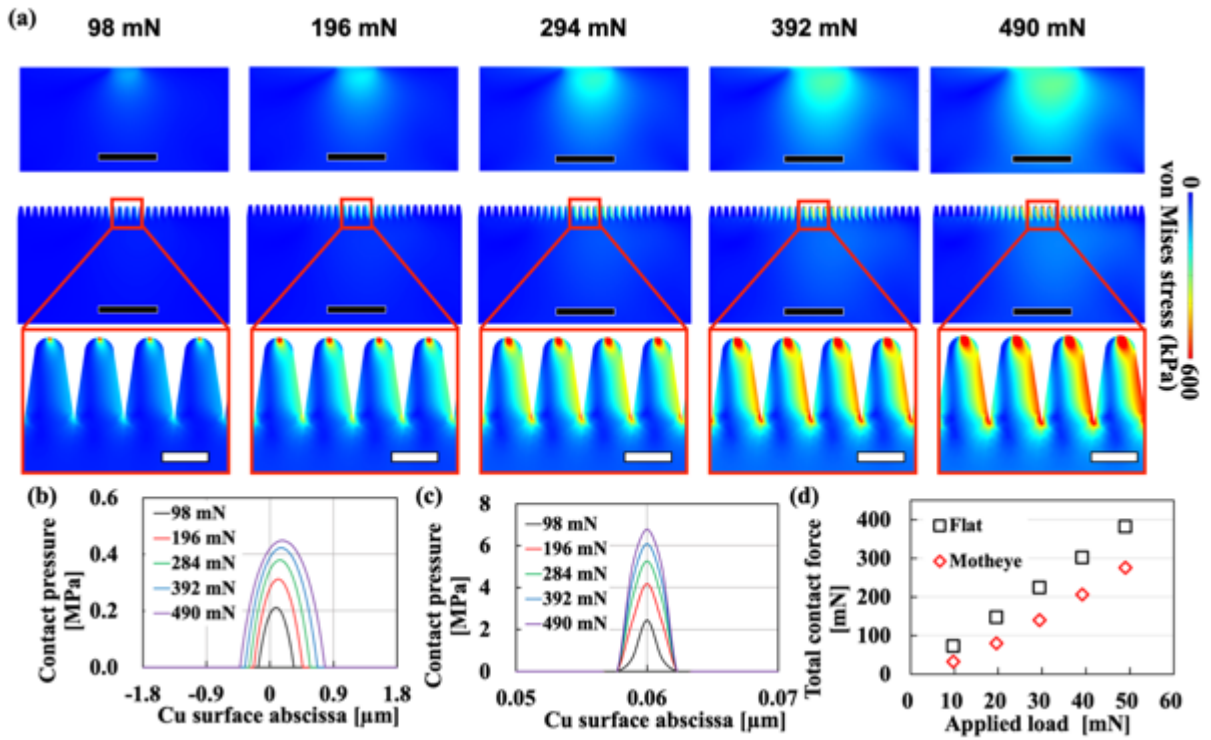


Fig. 3-23 (a) Simulation results illustrating the deformation stress applied during sliding for each load. For clarity, the indenter has been omitted. The upper image represents the flat surface, while the lower image depicts the moth-eye structure. The numbers above the images denote the applied load. Images enclosed within a red frame are a magnified view of the moth-eye surface. A black scale bar indicated 1 μm and white scale bar indicated 100 nm. Simulation results detailing contact pressure during sliding of the (b) flat and (c) moth-eye structures. Panel (c) shows the pressure on a single moth-eye protrusion. (d) Integral value of total contact force of flat structure and moth-eye structure for each load calculated using simulation software

The above research results suggested that the moth-eye structure inhibited the increase in the real contact area (deformation). As shown in Fig. 3-21, the force acting on the entire Cu model was not affected by the presence or absence of the moth-eye structure (Fig. 3-21 (g), reaction force rate). At the same time, the total contact pressure changed (Fig. 3-21 (g), total contact pressure rate). Therefore, the reduction in contact pressure may be attributed to the load dispersion on the surface and the moth-eye structure itself. Fig. 3-23 (a) shows the calculation results of the deformation stress of the Cu model for each load. On the plane, the area where the stress acts uniformly expands with each load increase, whereas in the moth-eye structure, the stress is locally concentrated and distributed not only to the tip but also to the side and root. In addition, in the plane structure, the contact area increases in proportion to the increase in load, whereas in the moth-eye structure, the contact is limited to the tip and does not expand any further (Fig. 3-23 (b, c)). These results support the claim that the moth-eye structure is resistant to deformation. Furthermore, it was found that the increase in contact pressure is smaller in the moth-eye structure than in the plane structure (Fig. 3-23 (d)). These observations indicate that the load on the side and bottom of the structure is distributed apart from the surface. In other words, the moth-eye structure resists deformation and causes deformation resistance (load dispersion effect). This effect reduces the total contact pressure on the surface and suppresses the increase in the real contact area.

The measurement and simulation results show a friction-reducing effect attributable to the moth-eye structure. This is due to the non-proportional increase in real contact area with respect to the load, a phenomenon explained by microcontact and load dispersion. Fig. 3-24 shows a schematic representation of this hypothesis. For surfaces lacking a microstructure, such as hard plastic deformation materials, the friction coefficient is constant because the roughness deformation and increase in real contact area are proportional to the load (Fig. 3-19 (e), Fig. 3-24 (a,b), n_{flat}). Offline, the friction coefficient of the moth-eye structure decreased exponentially with increasing load on the moth-eye surface. Simulation results revealed that the real contact area is in the nanoscale range at the tip of the structure (Fig. 3-21 (e,f)), and the contact area does not change with increasing load (Fig. 3-23 (c), microcontact). Essentially, the microcontact changes the typical nanoscale surface roughness to incorporate the contact of the microstructure and the indenter (Fig. 3-24 (a)). Even under microcontact conditions, the friction coefficient is constant if the moth-eye structure itself deforms proportionally to the load.

However, the simulations showed that the total contact pressure of the moth-eye structure was smaller than that of the planar structure, and the increase with load was also smaller (Fig. 3-21 (g), Fig. 3-23 (d)). The reduction in pressure on the surface suppresses the deformation of the actual surface. These findings suggest that the moth-eye structure is less likely to deform than the planar model. The reduction in contact pressure, even though the same force is applied to the moth-eye and the planar model, is due to the load being distributed to areas other than the surface, as in bridges and metamaterials. In the simulations, we confirmed the load dispersion from the contacting tip to the side and bottom of the structure (Fig. 3-23 (a) bottom row). These observations suggest that after microcontact, the real contact area did not expand proportionally to the load. This is due to the inherent resistance to deformation of the moth-eye structure and the load dispersion effect (Fig. 3-24 (a,b), n_{motheye}). In tests, the moth-eye structure promoted the reduction in friction.

The lack of change in the friction coefficient under low load conditions for the copper moth-eye structure can be attributed to the change in the height of the moth-eye (Fig. 3-19 (a, e)). In essence, the pressure concentration resulting from this change in the designed structure leads to deformation, making the friction force comparable to that of a flat surface. Moreover, because ants tend to slide down the surface of the moth-eye structure¹³⁷, it is conceivable that such deformation may be reduced and friction reduced under very low load conditions¹⁰³. Studies by Gorb et al. have shown that frictional forces are reduced on the rough surface of the insect's moth-eye scale^{116,117,138–141}. These results are similar to those of studies showing behaviors different from the Amont-Coulomb law in microtribosystems¹⁴² or low load conditions where even plastically deforming materials experience only elastic deformation¹⁰³. Another report suggests that the force exerted by the ants on the substrate was about 70 μN ¹⁴³. Even if the surface roughness is random, this force may be insufficient to induce deformation, providing evidence that there is no deformation under very low load conditions such as those experienced by ants. Furthermore, these reports also show different loading conditions than our experiments, where the friction coefficient of flat surfaces is constant. There are other microstructural friction studies carried out under high load conditions, but the principles of friction and contact are different for elastically deforming materials^{95,104,144–147}, and the friction coefficient decreases exponentially with increasing load even for unstructured surfaces due to the flexibility of the material^{98,104,105}. Although studies of the surface

microstructure of plastically deforming materials have been carried out ^{108–111}, the reason for the decrease in the contact area remains unclear. Alternatively, the experiments were not carried out under dry conditions as we did, but in the presence of a lubricant^{102,148}. Our findings are in agreement with the findings of K. Holmberg et al., where friction decreases as roughness increases¹²³. The measurements from the study of K. Holmberg et al. were close to the results of our study and that of Gorb et al. Although the contact area changes with the change in roughness, it is possible that the roughness does not deform due to the dispersion of the load. A recent report by M. Sedláček et al. also highlighted the influence of surface topography, especially the height and shape of the structures, on friction forces ^{125,126,149}. It was expected that changes in the height and shape of the structures would affect how forces are applied to the roughness when a load is applied, suggesting that load dispersion effects may affect friction forces. Furthermore, the size of the indenter is also important in such studies. If the indenter is not large enough relative to the microstructure, as in our experiments, factors such as drilling may prevent friction reduction from occurring^{109,135}. The relationship between the size and shape of the indenter and the microstructure is also important from a microcontact perspective. If the indenter is square rather than spherical and the size of the microstructure is on the sub-millimeter scale, nanoscale roughness on the microstructure may deform proportionally to the load, leaving the friction coefficient unchanged¹⁵⁰. Offline, identifying regions that suppress deformation while considering the load and strength of the microstructure and topography allows for independent control of the real contact area, regardless of the size and shape of the surface microstructure.

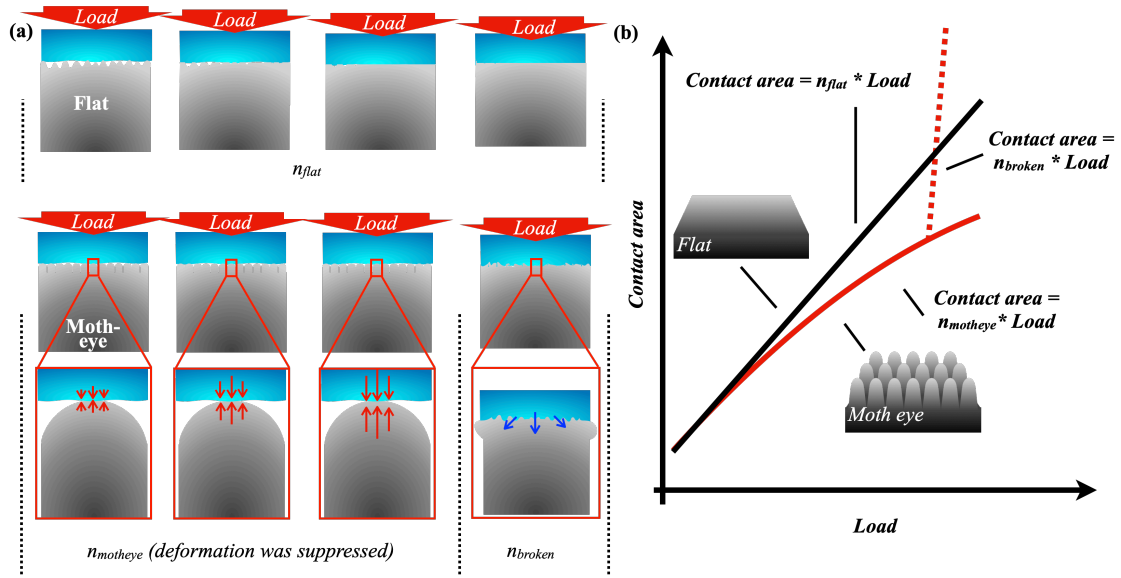


Fig. 3-24 (a) Schematic comparison of deformation between flat and moth-eye surfaces with respect to load. As the diagram moves further to the right, the load increases. In flat surfaces, the deformation of the roughness and the real contact area increase are proportional to the load (n_{flat}). Conversely, on moth-eye surfaces, the real contact area is the tip of the structure (microcontact), which resists deformation by load dispersion (indicated by the red arrow). (b) Therefore, the real contact area of the moth-eye structure proves challenging to increase with respect to the load ($n_{motheye}$), resulting in a smaller frictional force compared to that of a flat surface. In contrast, when the microstructure is disrupted, as observed in the case of the PSt sample (Figure S5), the frictional force increases (n_{broken}).

3-5 Application of the surface microstructure of the carrion beetle to other materials

3-5-1 Friction in elastic deformation materials

Elastically deformable materials such as vulcanized rubber and silicone rubber are known for properties such as flexibility, stress absorption, electrical properties, and barrier properties. These attributes make them ideal for use in biomedical devices¹⁵¹, soft robotics¹⁵², and flexible electronics¹⁵³, among others¹⁰⁴. However, the increase in adhesion friction due to their flexibility presents a concern¹⁵⁴. In addition, during sliding, the elastomer can deform in the sliding direction, creating friction hysteresis, and in some cases the digging friction required to climb over uneven surfaces becomes significant¹⁰⁴. As a result, elastic deformation materials typically exhibit a higher coefficient of friction than plastic deformation materials such as polymers and metals¹⁵⁴, requiring effective friction reduction measures.

A common method to reduce frictional forces is the application of lubricants; however, lubricants must be replenished periodically and can introduce new issues, such as the risk of combustion⁹⁹. In addition, the , the leakage of harmful lubricants poses significant environmental risks^{99,100}. Lubricant-free friction reduction methods are therefore increasingly sought after in various fields due to work environment constraints and concerns over environmental pollution.

Certain lubricant-free friction reduction methods inspired by the surface microstructures found in living organisms have attracted recent attention. Many species have evolved nm– μ m scale surface microstructures that provide remarkable friction-reducing functions^{58,106,155,156}. The microstructures of snakes¹⁵⁷, firebrats⁹⁴, and bush crickets⁹⁵ are known to reduce friction without lubricants. Friction can also be reduced in artificial materials by adding such surface microstructures to the materials^{108,109}, and extensive research on friction reduction by surface microstructures in flexible elastic deformation materials has been conducted^{104,144–146,158}. Flexible elastic deformation materials differ from hard plastic deformation materials, which follow the Amonton–Coulomb law¹³², and the surface roughness of these materials is often completely compressed under slight loads due to the flexibility of the material. This causes the apparent contact area to be nearly identical to the real contact area.^{105,154,159} As a result, the frictional force tends to decrease when the apparent contact area is reduced by microstructure, surface texture, and roughness^{104,144–146,158,159}. However, while a reduction in contact area is more likely to occur with larger-scale microstructures,

increasing the size of the microstructures can lead to digging friction when the mating surface enters the grooves of the structure ¹⁶⁰. This limitation suggests that there is a ceiling to the friction reduction achievable with single-layer structures. This study focused on the surface microstructure of the carrion beetle (*Necrophila japonica*) to reduce friction in elastic deformation materials. This species of carrion beetle lives on the forest floor and has a hierarchical surface microstructure composed of submicron-scale wrinkles on micron-scale microstructures on its elytra. Our research has shown that this structure provides a greater friction-reducing effect than that of a single-layer structure in rigid plastic deformation materials. We hypothesized that a hierarchical surface microstructure, such as that of the carrion beetle, could significantly reduce friction by maintaining the microstructure spacing, while still significantly reducing the contact area due to the large number of voids. In addition, we examined the discontinuity and deformation at the contact related to microstructure, which suppresses the compression difference due to sliding and reduces hysteresis friction ^{104,161}, so that the wrinkle structure at the top of the hierarchical structure may also reduce friction.

3-5-2 Mimicked structure preparation

The preparation method of the epoxy inverse structure as the template is the same as in 3-2-2. The PDMS sample was prepared by dropping the PDMS solution (crosslinking ratio = base: curing agent = 10:1) onto the epoxy resin mold and thermally crosslinking it at 70 °C for 12 hours. The structure of the prepared sample was observed and measured using a laser microscope (VK-X200, Keyence Corp., Japan) (n = 20).

The procedure for preparing the PDMS sample is shown in the following figures (Mimicked : Fig. 3-25, Wrinkle : Fig. 3-27, MLAs : Fig. 3-28). The dimensions of the obtained structures were wrinkles (diameter $1.668 \pm 0.459 \mu\text{m}$, depth $0.306 \pm 0.101 \mu\text{m}$), MLAs (diameter $12.225 \pm 0.233 \times 9.768 \pm 0.326 \mu\text{m}$, depth $1.721 \pm 0.171 \mu\text{m}$), and mimicked structures (wrinkles: $1.382 \pm 0.330 \mu\text{m}$, MLAs: $12.100 \pm 0.342 \times 9.803 \pm 0.293 \mu\text{m}$) (Fig. 3-29 (a-d)). The above sizes are the average values (n = 20) for each structure, and the numbers following $\pm$ represent the standard deviation. The wrinkle structure size was also changed by changing the number of spin coats as shown in Fig. 3-26.

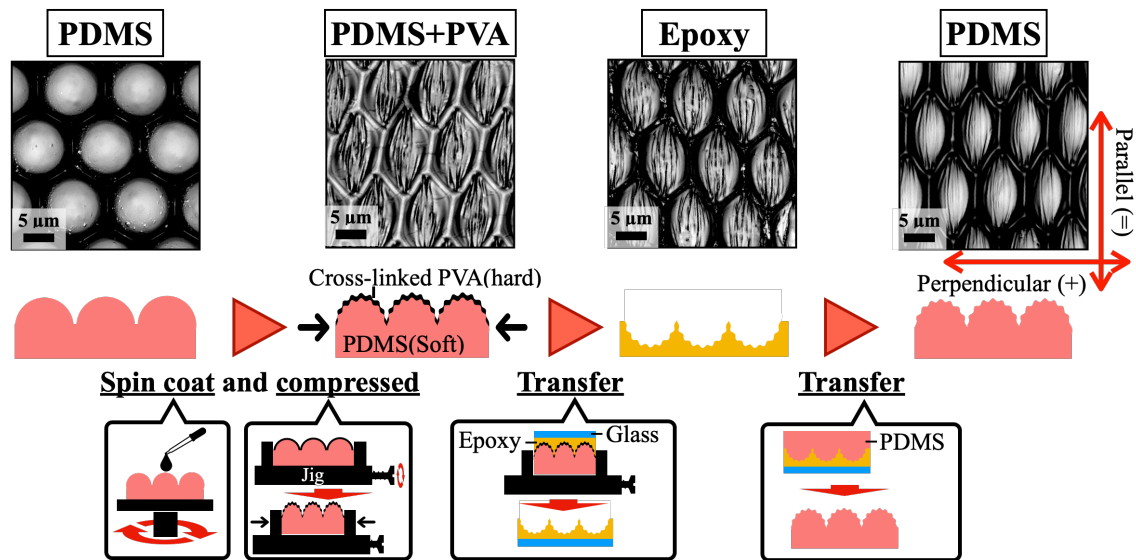


Fig. 3-25 Procedure for preparing the polydimethylsiloxane (PDMS) structures that mimic the carrion beetle surface. Wrinkles were formed by preparing a hard PVA layer on soft PDMS and compressing it. Although the PVA layer significantly affects friction, preparing a mimicked structure in a single material using two transfers allows investigation of the microstructure's effect on friction. Inset images are laser microscope images of each sample.

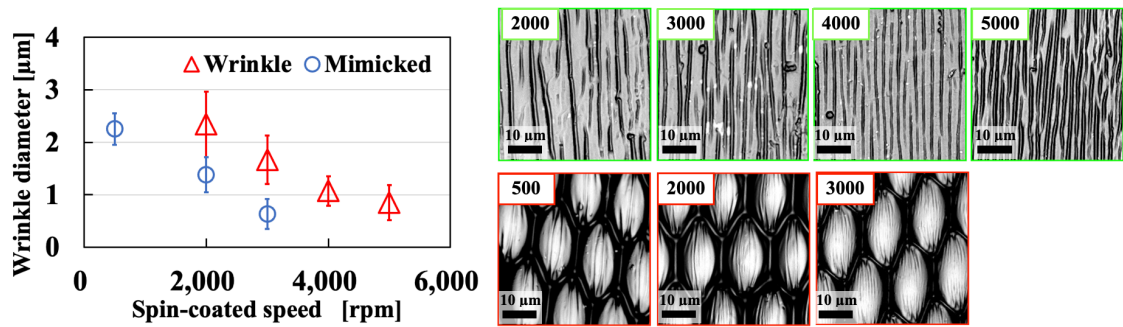


Fig. 3-26 Wrinkle structure diameter for each spin coating speed and the laser microscope images of PDMS wrinkle and mimicked structures with 11 μm MLAs. In the laser microscope images, the green outline indicates the wrinkles, the red outline indicates the mimicked structure, and the number surrounded by the colored outline in each image indicates the spin-coating speed. As the spin-coating speed increases, the PVA layer thickness and the wrinkle diameter both decrease.

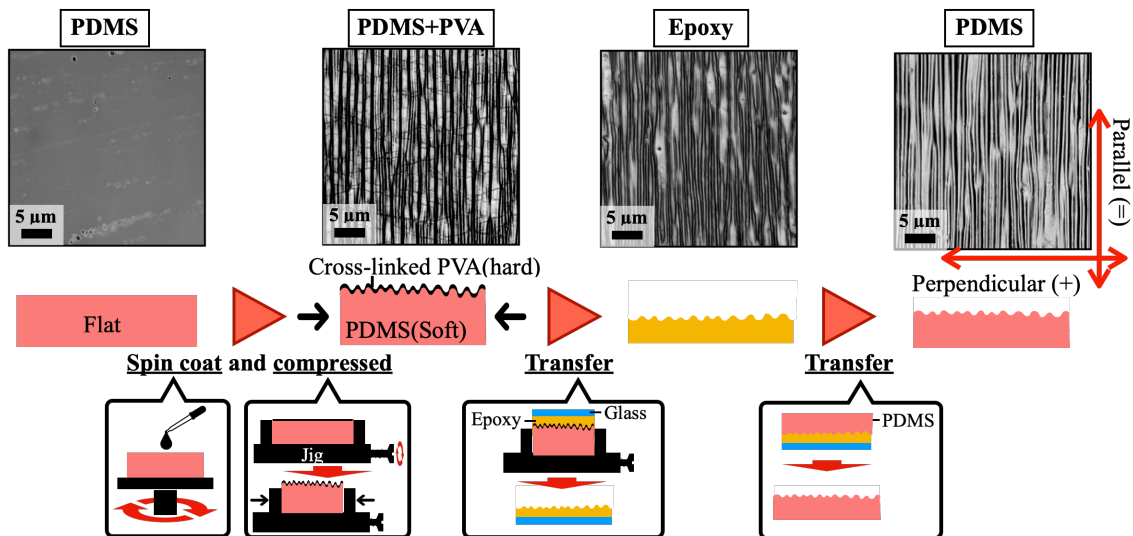


Fig. 3-27 Preparation procedure for the PDMS wrinkle structures. Wrinkles were formed by preparing a hard PVA layer on a soft PDMS flat surface and compressing it. Inset figures are laser microscope images of each sample. Red arrows indicate the sliding direction.

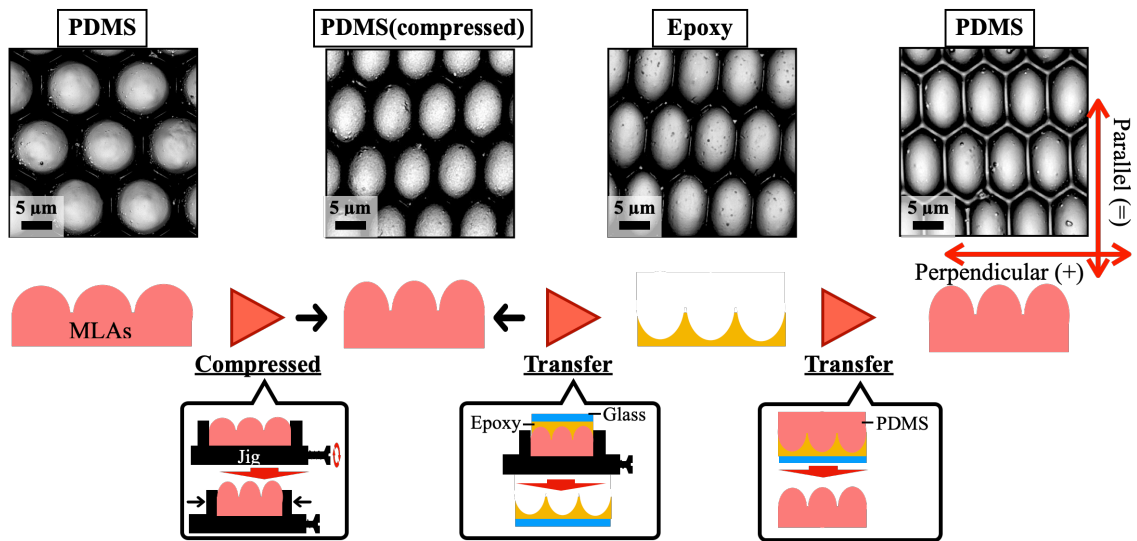


Fig. 3-28 Preparation procedure for the PDMS MLAs structures. The MLAs used in the measurements were formed by compressing circular structures onto PDMS MLAs. PVA was not spin coated. Inset figures are laser microscope images of each sample. Red arrow indicate the sliding direction.

3-5-3 Friction test results

The method for measuring friction force is the same as that shown in 3-2-3, except that the load is 98 mN-480 mN.

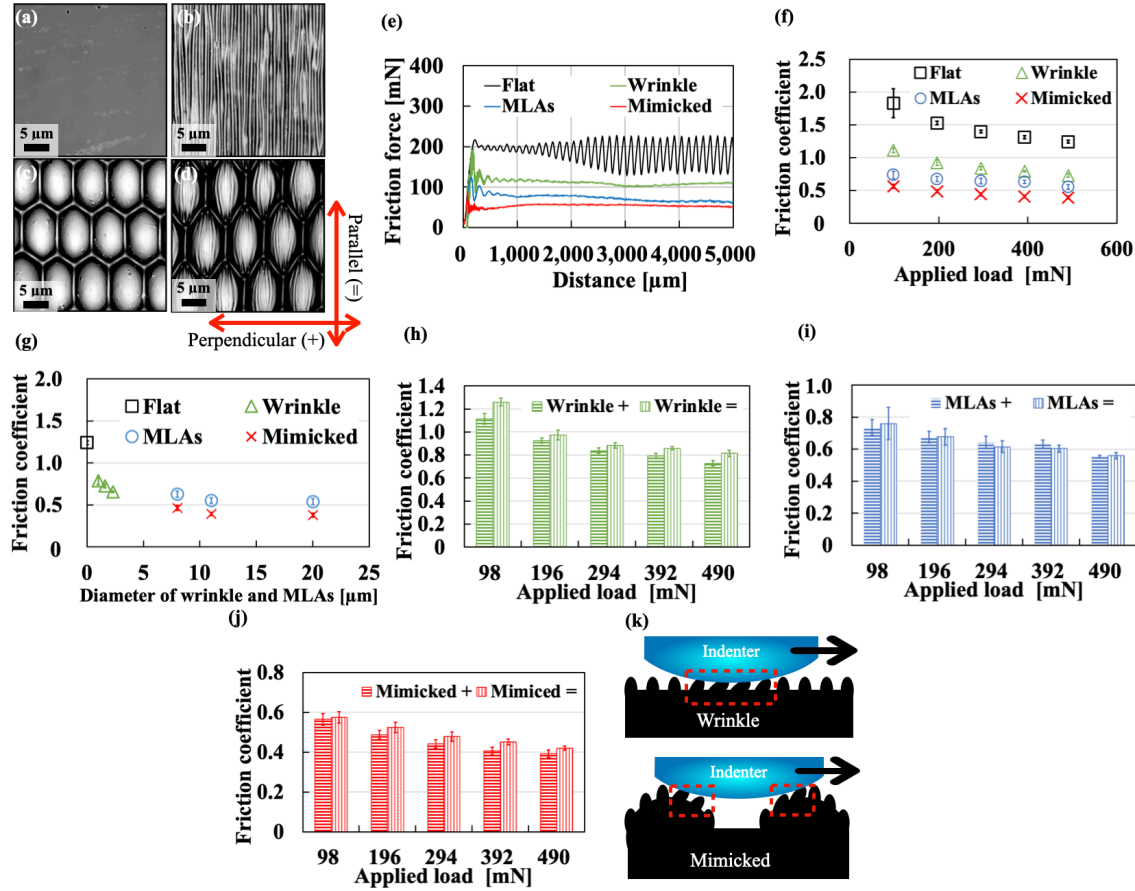


Fig. 3-29 (a-d) Laser microscope images and (e) frictional force measurements of (a) flat, (b) wrinkle, (c) MLAs, and (d) mimicked structures (wrinkle on MLAs). The load was 98 mN. (f) Friction coefficient for each load. (g) Friction coefficient of samples with varying MLA and wrinkle structure diameters. In the mimicked structure, only the MLA diameter was changed, while the wrinkle diameter was fixed at 1.6 μm. (e-g) The sliding direction in the graphs is perpendicular. Frictional force of (h) wrinkle, (i) MLAs, and (j) mimicked structure when changing sliding direction (+; perpendicular, =; parallel). (k) Schematic diagram of the structure deforming.

To examine the effect of structure on frictional force, the frictional forces of four types of samples were measured by sliding them in one direction with a load of 98 mN using a friction tester. The measurement direction was set perpendicular (+) across the structure. The results, shown in Fig. 3-29(e), indicated significant variation in

frictional force depending on the microstructure type, decreasing in the order of flat, wrinkle, MLAs, and mimicked. This trend was consistent across all loadings (Fig. 3-31), with the mimicked structure always exhibiting the lowest frictional force and coefficient of friction, up to 70% less than the flat surface (Fig. 3-30). The coefficient of friction of PDMS was not uniform across all samples, decreasing with increasing load (Fig. 3-29 (f)). This decrease was not influenced by differences in sliding direction, structure type, or size and was observed across all samples in this study (Fig. 3-30). On soft elastic material surfaces, even a slight load can cause surface unevenness to collapse, preventing a proportional increase in the real contact area with load ^{104,105,154,159}. Therefore, the observed reduction in friction is attributed to the difference in real contact area due to the microstructure.

At a load of 98 mN, stick-and-slip behavior was observed on the flat surface, but this was significantly suppressed in the microstructured surfaces, especially the mimicked structure. In Fig. 3-29 (e), large error bars for the 98 mN load of the flat surface indicate stick-and-slip effects, while no significant stick-and-slip was observed for other structures or loads, except for the 98 mN load of the flat surface. These results demonstrate that all structures—wrinkle, MLAs, and mimetic—effectively suppress stick-and-slip, with the mimetic structure showing the greatest effect. Furthermore, the mimetic structure maintained a consistently low coefficient of friction with no significant change in frictional force after 100 round-trip friction tests (Fig. 3-32). The samples produced exhibit improved repeatability and structural durability, overcoming typical weaknesses of microstructures, demonstrating very effective advantages for practical applications. When the size of the microstructure was varied, the frictional force decreased in proportion to the structure size, regardless of geometry. However, the rate of decrease diminished as the structure size increased. Notably, the mimicked structure consistently reduced friction more than the MLAs structure across all MLAs diameters and loads (Fig. 3-29 (g), Fig. 3-31, Fig. 3-30). While larger microstructure diameters reduce contact area, they can also increase digging, which limits friction reduction. However, the mimicked structure effectively reduces contact area while minimizing increases in microstructure spacing that lead to digging, thereby achieving the greatest reduction in friction.

Next, differences in frictional force due to varying sliding directions were examined (Fig. 3-29 (h, i, j), Fig. 3-31, Fig. 3-30). The results demonstrated anisotropy in both the wrinkle structure (Fig. 3-29 (h), Fig. 3-31 (a), Fig. 3-30 (a)) and the mimicked structure (Fig. 3-29 (j), Fig. 3-31 (c), Fig. 3-30 (c)). The frictional force was consistently lower in the perpendicular direction compared to the parallel direction, irrespective of the load, with both structures showing an average difference of slightly less than 10% in frictional force. Conversely, MLAs exhibited a less pronounced trend in frictional force difference by direction (Fig. 3-29 (i), Fig. 3-31 (b), Fig. 3-30 (b)). The anisotropy of frictional force in different sliding directions has been discussed in a previous study 7. According to that study, the anisotropy in frictional force in asymmetric structures is attributed to the difference in friction hysteresis caused by the discontinuity in contact between the structure and the indenter. In the parallel direction, the indenter and the structure maintain continuous contact, and the differential compression between the leading and trailing surfaces of the indenter in the sliding direction likely causes friction hysteresis. In the perpendicular direction, the contact becomes discontinuous, and the structure deforms during sliding, which suppresses friction hysteresis (Fig. 3-29 (k)).

In this experimental system, the indenter is large relative to the wrinkle structure, allowing the structure to collapse under the sliding force, thereby reducing friction hysteresis and digging due to stress relief and deformation in the sliding direction. The minimal anisotropy observed in MLAs can be attributed to the smaller structure size in the sliding direction compared to the wrinkle structure and the difficulty of deformation due to the larger structure size. The difference in frictional force with respect to deformation was also suggested by the difference between dynamic and static frictional forces. Under high load conditions, where stick-and-slip effects were minimal, the difference between dynamic and static friction coefficients was the smallest for flat surfaces, followed by MLAs, mimicked structures, and wrinkles (Fig. 3-33). Static frictional force is primarily determined by the difference in contact area at rest, so the difference in frictional force is mainly due to the difference in contact area. In contrast, in the case of dynamic frictional force, which causes deformation of the structure due to sliding, the effect of digging and friction hysteresis is smaller in wrinkle and mimicked structures, which are easily deformed, leading to a larger difference between static and dynamic frictional forces.

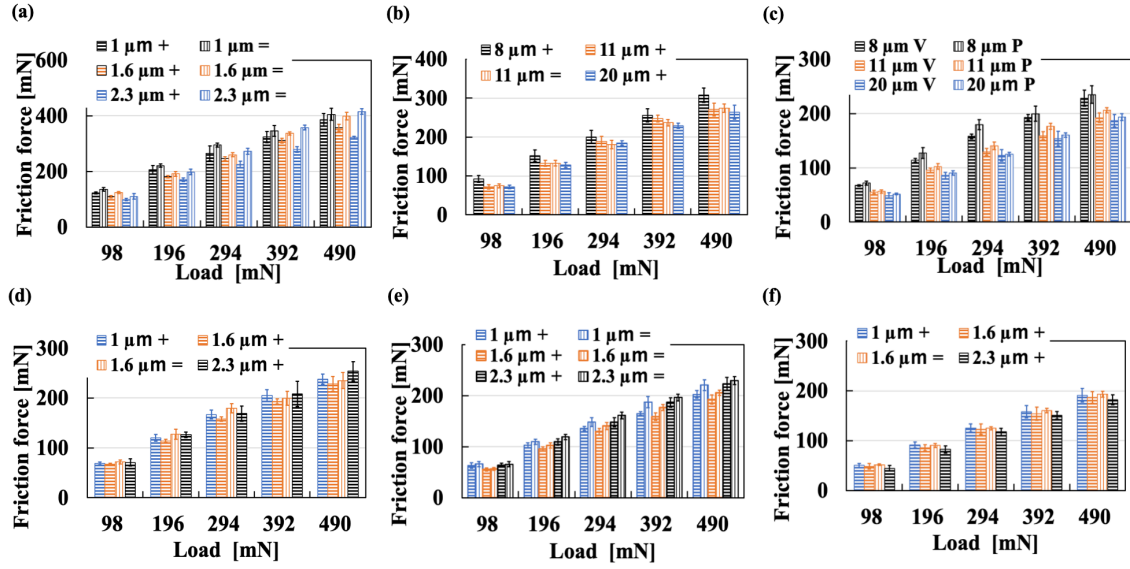


Fig. 3-31 Measured frictional force of (a) wrinkles, (b) MLAs, and (c-f) mimicked structure samples with varying MLAs and wrinkle structure diameter under each load. The legend indicates either (a) wrinkle or (b) MLA size. (c) In the mimicked structure, only the diameter of the MLAs was changed, while the diameter of the upper wrinkle structure was fixed at 1.6 μm .

(d-f) In the mimicked structure, only the diameter of the wrinkle was changed, and the diameter of the MLAs structure was fixed at (d) 8 μm , (e) 11 μm , and (f) 20 μm (+ ; Perpendicular, = ; parallel).

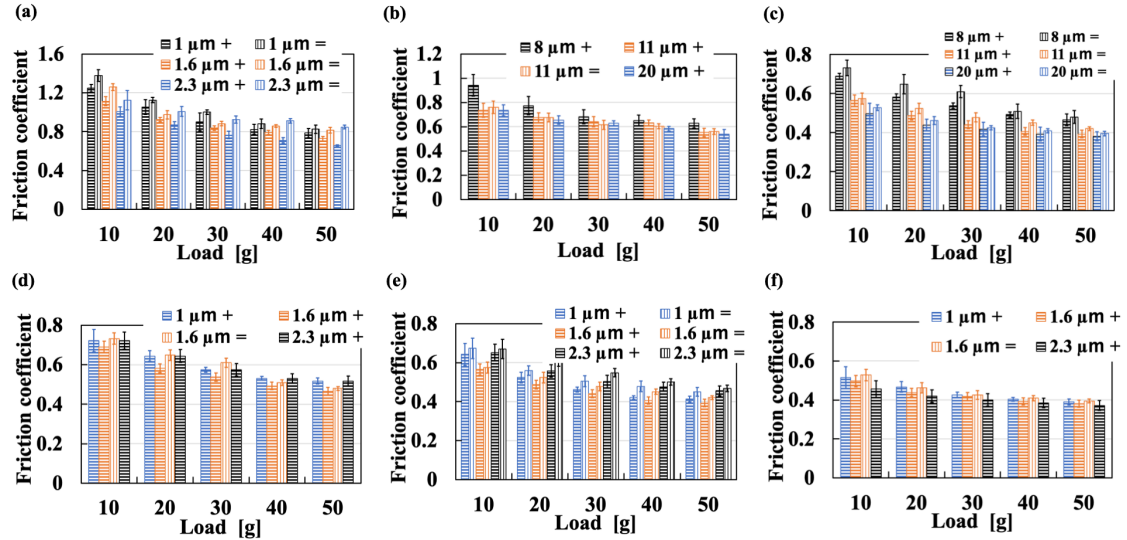


Fig. 3-30 Measured coefficient of friction for (a) wrinkles, (b) MLAs, and (c-f) mimicked structure samples with varying MLAs and wrinkle structure diameter under each load. The legend indicates either (a) wrinkle or (b) MLA size. (c) In the mimicked structure, only the diameter of the MLAs was changed, and the diameter of the upper wrinkle structure was fixed at 1.6 μm . (d-f) In the mimicked structure, only the diameter of the wrinkle was changed, and the diameter of the MLA structure was fixed at (d) 8 μm , (e) 11 μm , and (f) 20 μm .

(+ ; Perpendicular, = ; parallel).

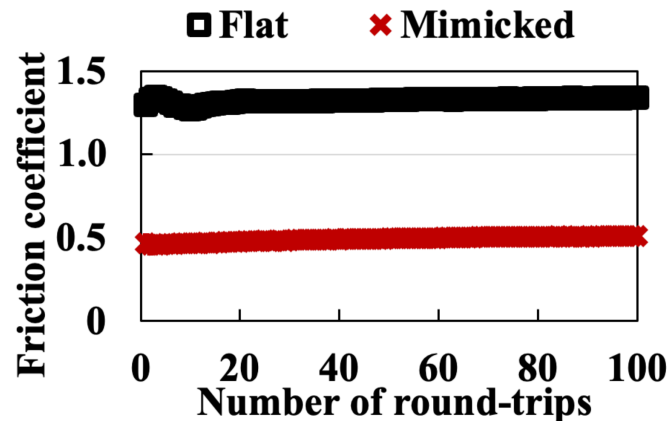


Fig. 3-32 Reciprocating friction measurement results. The mimicked structure featured 1.6- μm

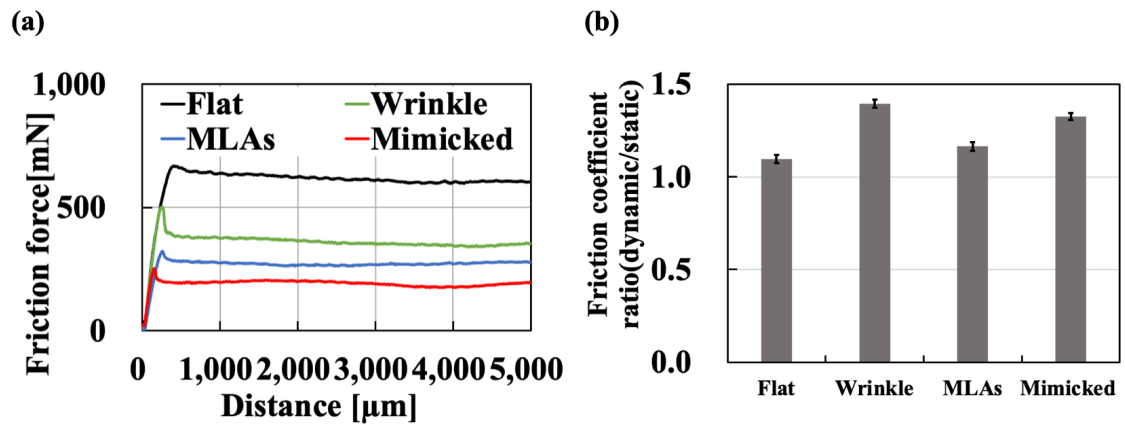


Fig. 3-33 (a) The frictional force of each PDMS sample during sliding under a load of 490 mN.

The sliding direction was perpendicular (+). (b) Ratio of dynamic coefficient of friction to static coefficient of friction. The dynamic coefficient of friction was the average of the coefficient of friction at 1000–5000 μm , while the static coefficient of friction was the maximum value of the coefficient of friction.

3-5-4 Simulation results

The simulation model and calculation method are basically the same as those in 3-3-1. The material properties of the structural model are changed to PDMS, and the load is changed to 490 mN. The calculation parameters are set as follows (Contact pressure penalty factor (Pn): 1e2 [N/mm³], Friction force penalty factor (ft): 1 (Automatic), Spring foundation (ktot): 2e6 [N/m], diagonal).

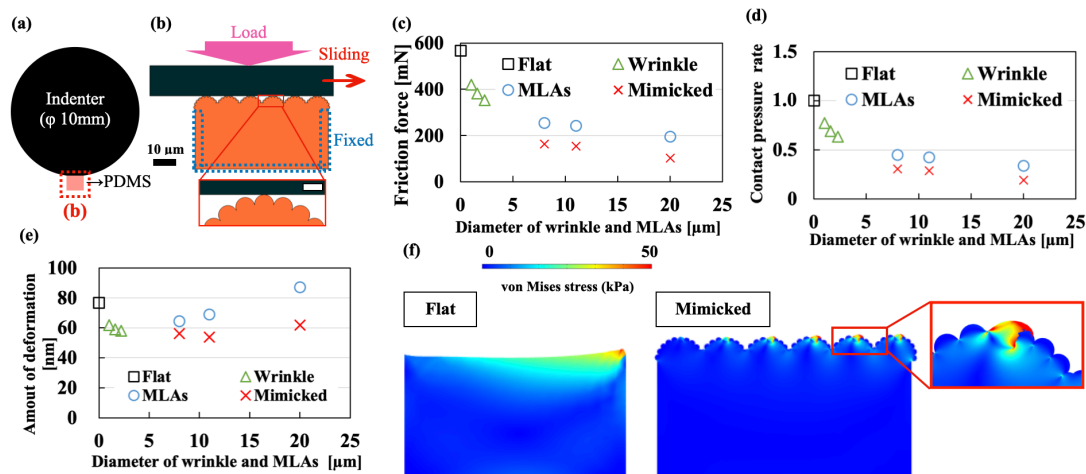


Fig. 3-34 (a) Schematic diagram of the simulation. Only a portion of the indenter is shown. The PDMS to indenter ratio is not accurate. (b) Simulation model of the mimicked structure. The frictional force is calculated when the top of the orange PDMS part and the bottom of the indenter are in contact, a load is applied from the top of the indenter, and it slides sideways. The inset image is a magnified view. The white scale bar is 1.6 μm. (c) Calculation results of frictional force and contact pressure ratio for each sample. The contact pressure is obtained by integrating the contact pressure generated at the interface during sliding, and the ratio of the contact pressure to the flat surface is shown in the graph as the contact pressure ratio. The contact pressure ratio in this model indicates the real contact area.

The actual measurement results suggest that the mimicked structure effectively reduces frictional force, likely due to a decrease in contact area and structural deformation. To further explore these effects, a simplified friction model was employed to analyze the impact of microstructures on frictional force, stress, and deformation (Fig. 3-34). An indenter of the same size used in the actual measurements was positioned on top of a PDMS structure representing the microstructure sample. The simulation calculated the frictional force, deformation amount, and deformation stress when a load of 490 mN was applied to the indenter, with the side and bottom edges of the PDMS fixed. The frictional

force followed Coulomb's law and was calculated using the product of the interfacial contact pressure, generated by a virtual spring at the interface, and the friction constant

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The results shown in Fig. 3-34 (c) indicate a reduction in frictional force due to the microstructure, consistent with the measured data. For single-layer structures like wrinkles and MLAs, the frictional force decreased proportionally with structure size, but this reduction lessened as the structure size increased. However, the mimicked structure consistently showed a greater reduction in friction than the MLAs structure across all sizes. This alignment with the measured results suggests that the force applied to the surface varies with the microstructure. The frictional force calculated in this study also involved calculating the ratio of the total contact pressure applied to the model structure surface during sliding to that on a flat surface. The contact pressure was reduced by the microstructure and was almost proportional to the reduction in friction. Additionally, when the apparent contact area between the indenter and the structural model during sliding was calculated, the frictional force decreased proportionally to the reduction in the apparent contact area, as well as the ratio of the forces acting on the surface, for all structures and structural sizes, in both calculations and actual measurements (Fig. 3-35). This is thought to be because, as stated in previous papers, the apparent contact area and the real contact area are almost equal on the surface of soft, elastic deformation materials 104,105,145,154,158,159. These results suggest that the microstructure reduces the real contact area, with the greatest effect observed in the mimicked structure.

The calculated deformation during sliding is shown in Fig. 3-34 (e). In single-layer structures, deformation decreased up to a certain structure size but then increased proportionally with structure size, with MLAs of 20 μm showing more deformation than a flat surface. This is likely due to increased voids from larger structure sizes, leading to digging. However, the results suggest that the mimicked structure can suppress increased digging while reducing contact area more effectively than larger MLAs structures. From the deformation and stress dispersion results during sliding (Fig. 3-34 (e)), the flat surface showed significant elevation in the sliding direction, indicating potential digging and friction hysteresis. In contrast, the mimicked structure exhibited deformation where the structure escaped in the sliding direction, potentially reducing hysteresis and digging friction. This effect is supported by the reduction in stick-and-slip, anisotropy, and difference between static and kinetic friction coefficients observed in the mimicked

structures (Fig. 3-29, Fig. 3-30, Fig. 3-33). Additionally, the lower frictional force in the perpendicular direction compared to the parallel direction, despite the same contact area, indicates reduced hysteresis and digging friction due to structural deformation. As shown in Fig. 3-35, the mimicked structures with the same apparent contact area as the MLAs consistently had lower frictional forces than the MLAs in measurements. This suggests that the deformation of the wrinkle structure on the top of the mimicked structures is a key factor in reducing friction, beyond the reduction in contact area.

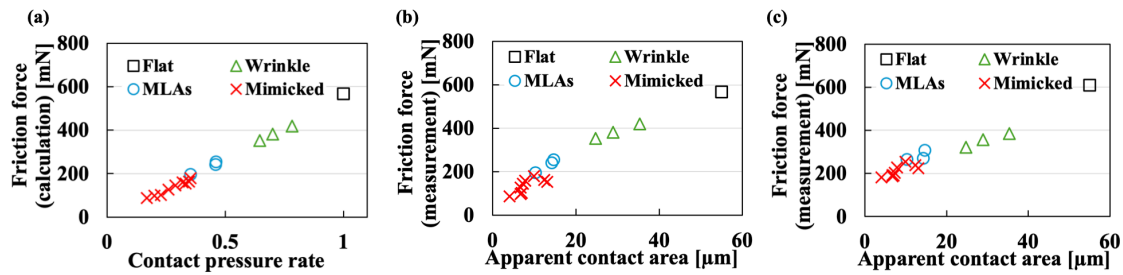


Fig. 3-35 (a) Calculated contact pressure rate and calculated frictional force, (b) calculated apparent contact area and calculated frictional force, and (c) calculated apparent contact area and measured frictional force. The graph includes measured and calculated results for all sizes of flat, wrinkle, MLAs, and mimicked structures. Apparent contact area is the calculated range with which the structural model was in contact during the slide.

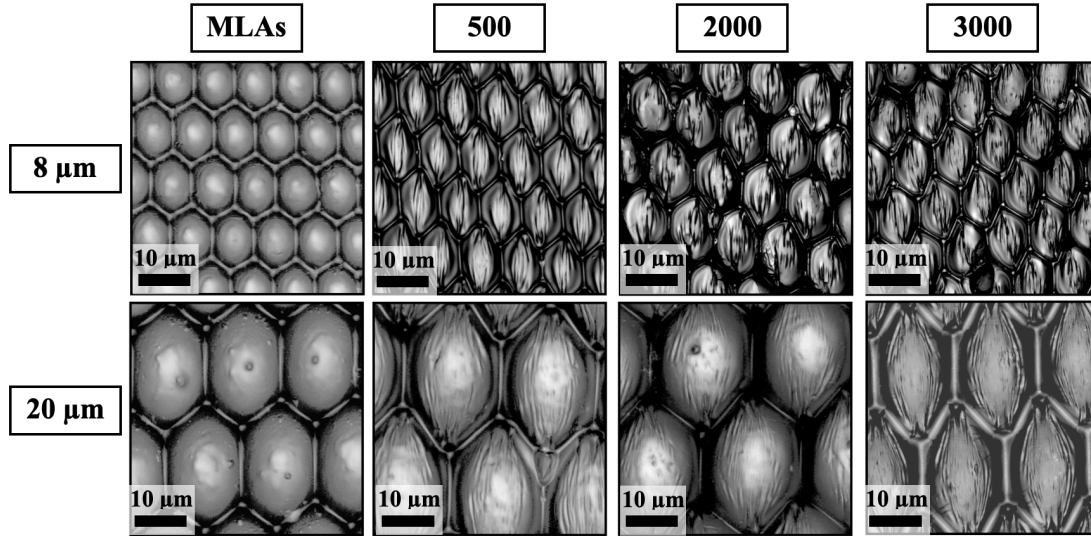


Fig. 3-36 Laser microscope images of PDMS MLAs and mimicked structures with 11- μ m MLAs. In the laser microscope images, the green outline indicates the wrinkle, the red frame indicates the mimicked structure, and the number surrounded by the colored outline in each image indicates the spin-coating speed. As the spin-coating speed increases, the PVA layer thickness and the wrinkle diameter both decrease.

Next, we investigated the effect on friction by using mimicked structures in which the MLAs size and wrinkle structure size were changed, as shown in Fig. 3-36. The wrinkle size was adjusted by altering the spin-coating speed. The results, shown in Fig. 3-37, Fig. 3-31, Fig. 3-30, indicate that an optimal combination of microstructure sizes exists for reducing friction. For MLA sizes of 8 and 11 μ m, the smallest frictional force was observed when the wrinkle size was 1.6 μ m. However, at 8 μ m, a wrinkle size of 1 μ m resulted in lower friction than a size of 2.3 μ m, whereas at 11 μ m, the wrinkle size of 1 μ m led to higher friction than 2.3 μ m. When the MLAs were 20 μ m, the frictional force was smallest at 2.3 μ m, which was the lowest among the conditions tested (Fig. 3-37 (a), friction coefficient: 0.371). This demonstrates a successful reduction of the coefficient of friction for high-friction materials, with values exceeding 1, down to levels comparable to general polymers and metal materials, without the use of lubricants or composites of other materials. The trend observed in the actual measurements was consistent with the simulation results, which compared the ratio of the frictional force to the force applied to the surface (Fig. 3-37 (b, c)). Again, the frictional force was roughly

proportional to the apparent contact area (Fig. 3-35). For MLAs of 8 μm , the smallest deformation occurred with a wrinkle size of 1 μm , while for 11 μm , the smallest deformation was at a wrinkle size of 1.6 μm , matching the actual measured frictional force (Fig. 3-37 (d)). In contrast, at 20 μm , the smallest deformation was observed with a wrinkle size of 1 μm (Fig. 3-37 (d)). These results suggest that as the lower MLA structure size increases, the upper wrinkle size can also be increased to reduce friction, which is expected to decrease the true contact area and thus reduce friction. However, there appears to be a limit to friction reduction, as increasing the upper wrinkle size too much can lead to increased digging, potentially limiting the reduction in friction. These effects are likely influenced by factors such as the indenter size, and further detailed and systematic research is needed to fully understand them.

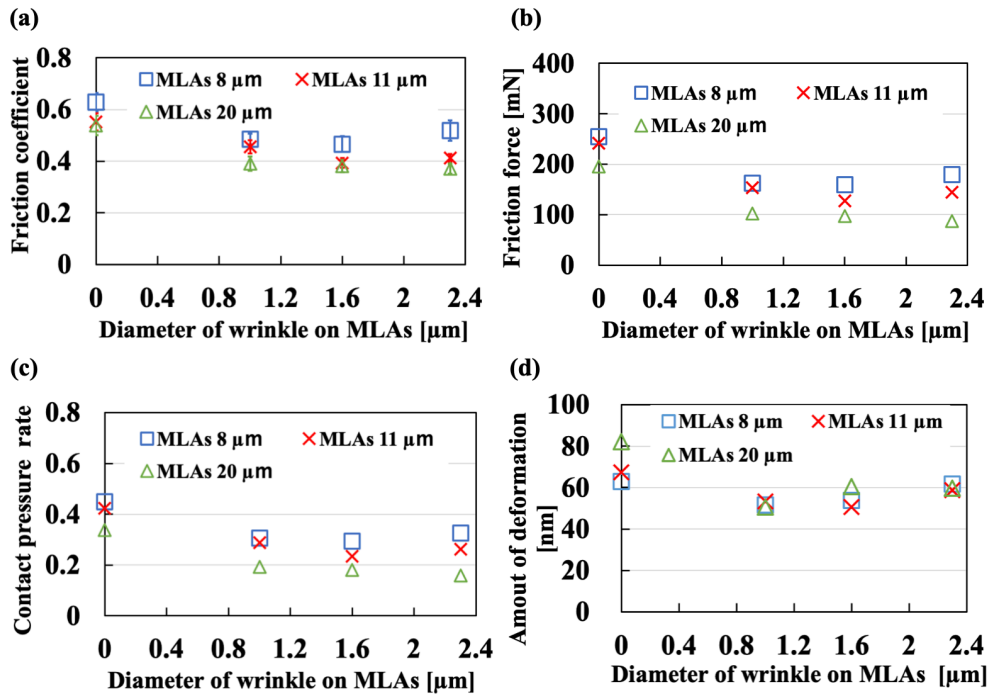


Fig. 3-37 (a) Friction coefficient results from actual measurements and (b) calculated frictional force, (c) contact pressure ratio, and (d) deformation amount for varying mimicked structure sizes. All results displayed in the graph are for the mimicked structure; the horizontal axis represents the wrinkle structure diameter on MLAs, and the legend shows the MLA size. The load is 490 mN, and the sliding direction is perpendicular.

The studies shown in 3-2 and 3-3 have revealed that hierarchical structures such as those seen in *Necrophila japonica* reduce friction. In hard plastic deformation materials,

the real contact area is smaller than the apparent contact area due to roughness and hardness, and when a load is applied, the roughness deforms in proportion to the load. This means that the frictional force is proportional to the load rather than the apparent contact area, resulting in a constant coefficient of friction regardless of the load ^{132–134}. Adding a hierarchical structure to plastic deformation materials decreases friction by reducing contact points and suppressing deformation due to the load-dispersing effect. In contrast, the real and apparent contact areas of elastic deformation material surfaces tend to be equal, even on flat surfaces, due to the material's flexibility. Thus, frictional force is not proportional to the load, and the coefficient of friction decreases exponentially. On hierarchically structured surfaces, the microstructure not only reduces the contact area but also reduces excavation and friction hysteresis due to deformation in the sliding direction. The frictional force on the surface of plastic deformation materials is reduced by the hardness of the microstructure, while the frictional force on the surface of the mimicked structure is reduced by the flexibility of the microstructure.

In addition, the hierarchical structure may offer benefits beyond friction reduction through decreased contact area, digging, and friction hysteresis. The prevention of stick-and-slip by the mimicked structure may help prevent adhesion over a large area, potentially aided by the local deformation of the wrinkle structure in the sliding direction. This local deformation may also alleviate pressure concentration due to the reduced contact area, so repeated use may not lead to a significant increase in the friction coefficient. Previous research on reducing friction using elastically deformable materials has focused on single-layer structures ^{104,144–146,159}. Therefore, increasing microstructure spacing to reduce friction may lead to increased digging. Conversely, studies on friction reduction through hierarchical structures mimicking snakeskin have shown that decreasing the contact area and adjusting compressive force can reduce friction ¹⁶¹. However, these studies often involve hard surface layers, and the observed reduction is not solely due to the structures.

In this study, we have created a hierarchical structure on the surface of a single material by sandwiching two transfers, a technique that can be applied in various fields where mixing with other materials is undesirable and has the potential for broad applicability in other materials. In future applications, the ability to reduce friction without using lubricants, which can cause environmental issues, could be highly beneficial ^{99,100}, especially in soft robotics, wearable electronics, and biomedical devices.

Another important environmental consideration is the use of self-organization to prepare mimicked structures, akin to those found in living organisms, rather than relying on lithography or etching, which require substantial cost, time, and specialized equipment. This combination of solutions to major friction-related challenges is a novel and outstanding property of our carrion beetle-inspired hierarchical structure.

3-6 Other functions and structures

3-6-1 Water repellency due to microstructure

The friction-reducing function of the surface microstructure of *Necrophila japonica* has been studied and discussed. However, the function of the hierarchical structure in the previous studies is superhydrophobicity, as seen in the ^{86,91} (Fig. 3-45) and lotus leaf (Fig. 3-47) ^{3,9,90}. Self-cleaning is important for *Necrophila japonica*, which, in addition to worms, eats decomposing corpses, and for beetles, which live on the surface of moist forests and cannot breathe as humans do, self-cleaning is important. For these beetles, which live on the surface of moist forests and cannot breathe like humans, securing a breathing opening (stomatal opening) by water repellency is a matter of life and death. Therefore, we investigated the wettability of *Necrophila japonica*.

Wetting is a phenomenon caused by the surface (interfacial) tension of a liquid when it touches a solid. The shape of a droplet when a liquid touches a solid is determined by the balance between the surface tension of water γ_L , the surface tension of the solid γ_s , and the interfacial tension between the liquid and the solid γ_{sL} , (Fig. 2 16). The equation for this balance is expressed by the following Young's equation ^{15,162}, where θ is the contact angle and γ_s is the interfacial tension between the liquid and the solid (Fig. 3-38). where θ is the contact angle.

$$\gamma_s = \gamma_{sL} + \gamma_L \cos\theta, \quad \cos\theta = \frac{\gamma_s - \gamma_{sL}}{\gamma_L} \quad \cdot \cdot (11)$$

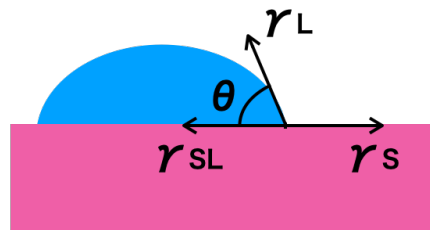


Fig. 3-38 Balance of surface (interface) tension forces when a liquid is placed on a solid

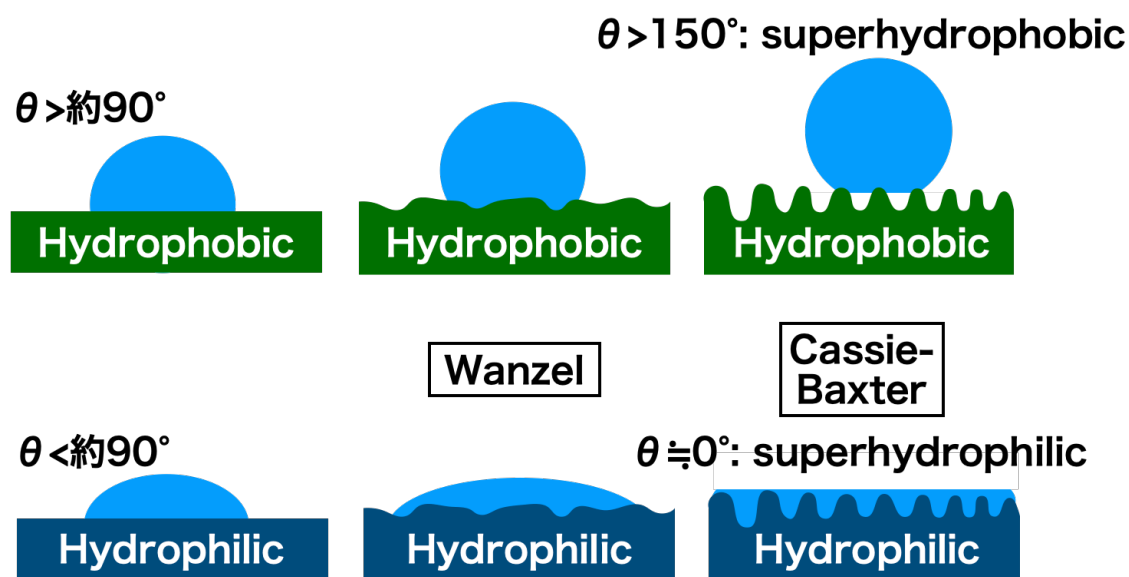


Fig. 3-39 Changes in wetting behavior due to surface roughness and chemical properties

When the liquid is water, the surface tension of the solid is high, and the interfacial tension with water is low, such as with a glass substrate, which has a low contact angle (easy to wet). On the other hand, materials such as Teflon have a low surface tension, and the liquid tends to curl up, resulting in a high contact angle (difficult to wet = easy to repel). In general, a contact angle of 90° or less is called hydrophilic, and a contact angle of 90° or more is called hydrophobic (Fig. 3-39).

If the substrate surface is flat, the contact angle when CF_3 groups, which have the smallest surface tension of any substance known to date, are arranged on the surface is approximately 120° ¹⁶², which is the maximum contact angle that can be generated by chemical factors such as surface tension. However, as mentioned above, lotus leaves have a very large contact angle of over 150° . This is related to the roughness of the surface. If the substrate surface is rough to a certain extent, the contact area with water increases accordingly. Therefore, the effect of surface (interfacial) tension increases by a factor of R , which is the surface roughness compared to a flat substrate.

$$\cos\theta_w = \frac{R(\gamma_s - \gamma_{sL})}{\gamma_L} = R\cos\theta \quad \cdot \cdot (12)$$

For these reasons, the contact angle on a rough surface is θ_w , as shown in equation (12). Since $R \geq 1$, if $\cos\theta$ is positive ($\theta < 90^\circ$, hydrophilic), the contact angle is further decreased, and if $\cos\theta$ is negative ($\theta > 90^\circ$, hydrophobic), the contact angle is further increased, as shown in Fig. 3-39. That is, on a rough surface, a wettable surface becomes more wettable, and a repelling surface becomes more repelling. This is called the Wenzel state. However,

this Wenzel state does not always hold, since it assumes that the water and the solid are in contact with each other. For example, when the water repellency of the substrate increases and the water can no longer reach the bottom of the grooves, a different theory is applied. This is called the Cassie-Baxter state. In the Cassie-Baxter state, the substrate is assumed to be a mosaic of materials with contact angles θ_1 and θ_2 . The total contact angle in this case is expressed by the following equation.

$$\cos\theta_c = f_1\cos\theta_1 + f_2\cos\theta_2 \quad \cdot \cdot (13)$$

In this case, f_1 and f_2 are the ratios of the material with contact angles θ_1 and θ_2 ($f_1+f_2=1$). Since the state in which the liquid cannot penetrate into the groove can be considered as a mosaic structure with air, equation (13) can be transformed as follows.

$$\cos\theta_c = f\cos\theta - 1 + f \quad \cdot \cdot (14)$$

As water bends under weightless conditions, the contact angle between water and air can be assumed to be 180° , so $\cos\theta_2(\text{air}) = -1$, and $f_1 + f_2 = 1$, so $f_2 = 1 - f_1$, which gives us equation (14). If the solid area ratio f is reduced, the contact angle approaches 180° , and contact angles of 150° or more are called superhydrophobic (Fig. 3-39). The surface of lotus leaves is covered with wax and is hydrophobic. As shown in Fig. 3-47, the surface has a large contact angle due to the innumerable protruding structures, and water easily rolls off because the surface area in contact with the leaf is small.

• Water contact angle measurements

To investigate the wettability of the microstructure of *Necrophila japonica*, the static water contact angle (WCA) was measured. The WCA of the sample surfaces was measured using a contact angle measuring device (FAMAS, Drop Master, Kyowa Interface Science, Japan). The static WCA of the beetle elytra (Fig. 3-40(a)) was measured by dropping 1 μL of ultrapure water on the sample surface, and the static WCA of the PSt sample surface (Fig. 3-40 (b)) was measured by dropping 3 μL of ultrapure water on the sample surface. The contact angle was distinguished as being perpendicular or parallel to the wrinkle structure (Fig. 3-40 (c)). Perpendicular (+) was used to observe the perpendicular water drop, and parallel (=) was used to observe the parallel water drop. The average of six measurements was taken for the elytra of *Necrophila japonica*, and the average of four measurements was taken for the PSt sample. The error bars in the graphs represent the standard deviation. The results are shown in Fig. 3-40 (d). The WCA was about 90° , and the superhydrophobicity was zero in either direction. Anisotropy was

observed in different directions, with the WCA being slightly higher in the perpendicular direction. Numerous studies have examined the anisotropy of the static contact angle due to the anisotropy of the microstructure⁶⁻⁹. According to these studies, the anisotropy of the contact angle is due to the anisotropy of the roughness. In other words, the WCA increases according to Wenzel's law, as the structures exist continuously and discontinuously in the parallel and perpendicular directions, respectively. The contact angle of the mimicked polystyrene (PSt) structure was higher than that of a flat surface (water droplet of 3 μL) without any structure, indicating that the mimicked structure exerted a water-repellent effect (Fig. 3-40 (e)). However, the mimicked structure did not exhibit superhydrophobicity. This is thought to be because a smooth structure such as that of *Necrophila japonica*, even if it is a hierarchical structure, does not reach the Cassie-Baxter state required for superhydrophobicity. From these results, although superhydrophobicity was not observed, the improvement in wettability may lead to ensuring breathing, etc. Also, it is thought that it suggests that friction reduction is more important than superhydrophobicity for *Necrophila japonica*.

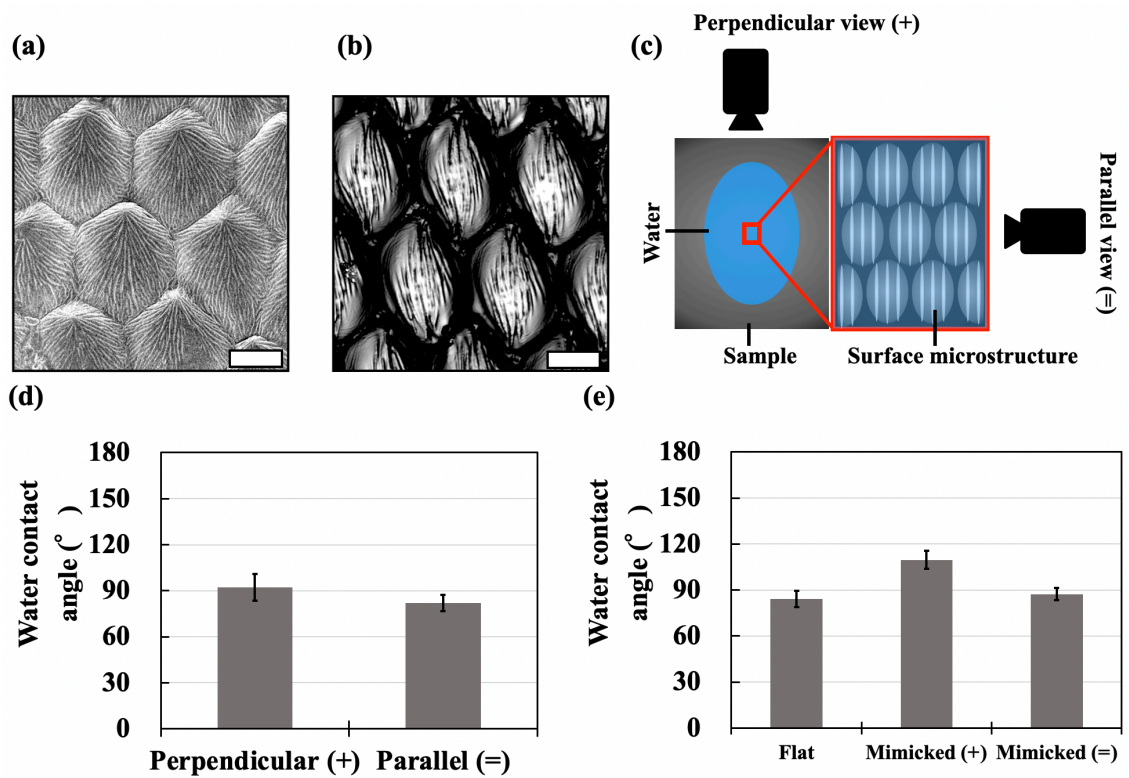


Fig. 3-40 (a) FE-SEM image, (b) laser microscopy image, and (d, e) WCAs of the (a, d) carrion beetle elytra and (b, e) PSt carrion beetle elytra mimicked structure. For comparison, the WCA of a flat PSt surface was also measured. (c) Contact angle of the mimicked structure was divided into the perpendicular view (+) when observed from a direction perpendicular to the wrinkle structure and parallel view (=) when observed from a parallel direction. The white scale bar was 5 μm .

3-6-2 Structure and function of the elytra of the carrion beetle, as seen on the underside and in cross section

We have been studying the functions of the surface microstructures of *Necrophila japonica*. However, beetles and other insects have microstructures not only on the surface but also on the underside and cross-section, and these structures have functions^{17,163–165}. Therefore, in this article, we observed the underside and cross-section of the elytra of *Necrophila japonica*.

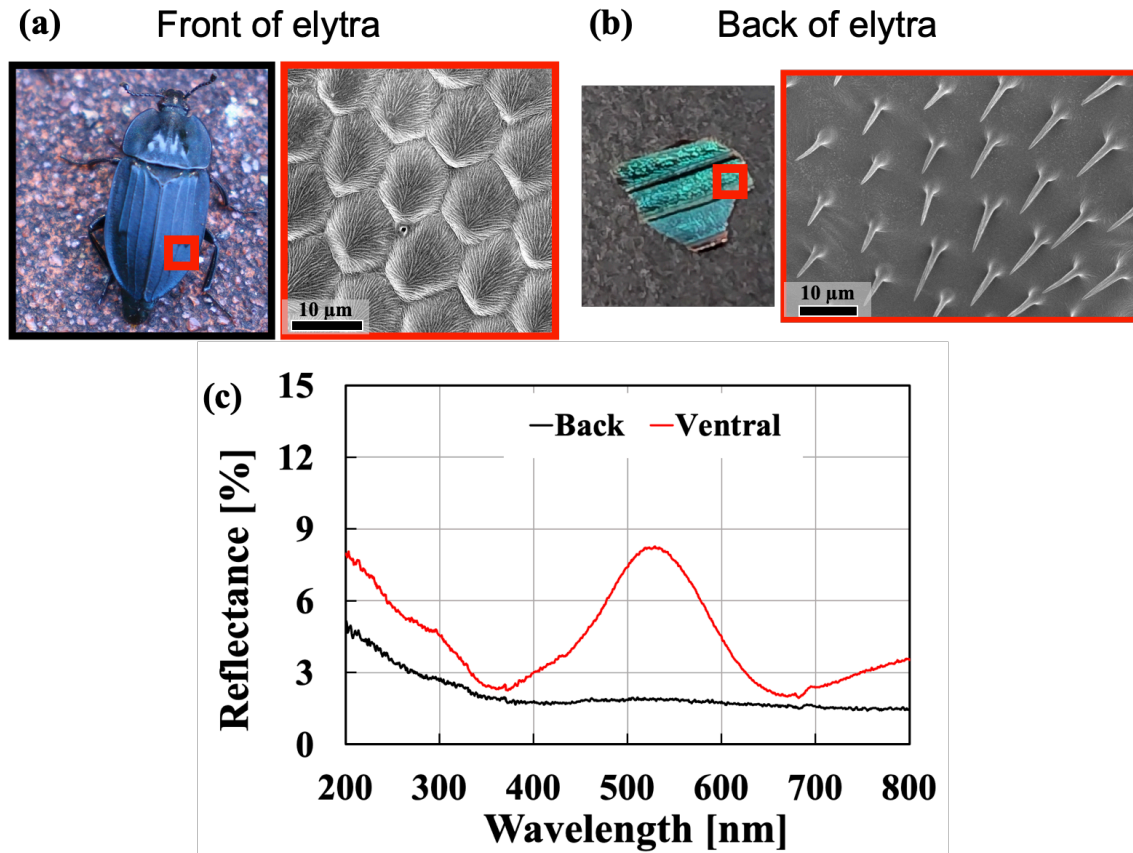


Fig. 3-41 (a, b) Photographs, (a, b) SEM images and (c) reflectance measurements of the (a) front and (b) back of the elytra of the carrion beetle

First, the underside was observed to be a beautiful glossy green color, while the front side was bluish black(Fig. 3-41 (a, b)). When the reflectance was measured using a microreflection spectrophotometer (MSV-5200, JASCO, Japan), the front side showed no particular peak and the reflectance only increased as the wavelength approached the ultraviolet range, while the back side showed a reflection peak at about 530 nm and low reflectance at about 350 nm and 660 nm(Fig. 3-41 (c)). It is highly likely that this glossy color is a structural color that is expressed by a microstructure rather than a pigment.

When the surface of the back side was observed with an SEM, it was found that there was no hierarchical structure like the front side, and that long, thorn-like structures were present all over the surface (Fig. 3-43). This structure is called microtrichia, and has been observed and studied in other beetles in the past^{163–165}. According to those reports, the function of this microtrichia is to interlock the elytra (outer wings) and the inner wings, making it easier to open the wings for flight. *Necrophila japonica* is characterized by dimorphism of flight muscles, with some individuals capable of flight and others unable to fly, but both groups show fully developed wings^{4,5}. Research has also shown that *Necrophila japonica* is not attracted to food placed in the air, suggesting that even individuals capable of flight rarely fly. Therefore, although the micro-nanofilaments exert an interlocking function, they may actually have the micro-nanofilaments as a vestige of their flight ability. In addition, since they are present on the entire surface of the wings, they are likely to function as a cushion. By sandwiching an air layer between them, it is possible that they may have an insulating effect. However, there have been no reports that microtrichia show structural color, and even considering the principle of structural color^{7,8}, microtrichia are too large to show structural color. In fact, when the microtrichia of *Necrophila japonica* were transferred to PVA and PSt to create a mimicked structure, and the reflectance was measured in combination with a surface without a structure, no reflection peak was observed (Fig. 3-42). Therefore, it is highly likely that the microstructures resulting from structural color are not on the surface. Some organisms, such as jewel beetles and Japanese beetles, produce structural color due to structures (multilayer structures, gyro structures) that can be seen by observing a cross section^{7,8,166}. When a cross section was observed, a multilayer structure was confirmed on both the front and back. This is similar in size to the multilayer structures seen in jewel beetles, and even when a multilayer structure was created in a simulation, a reflection peak close to that of an actual measurement was obtained, so it is highly likely that the beautiful green color seen on the back of *Necrophila japonica* is structural color due to a multilayer structure.

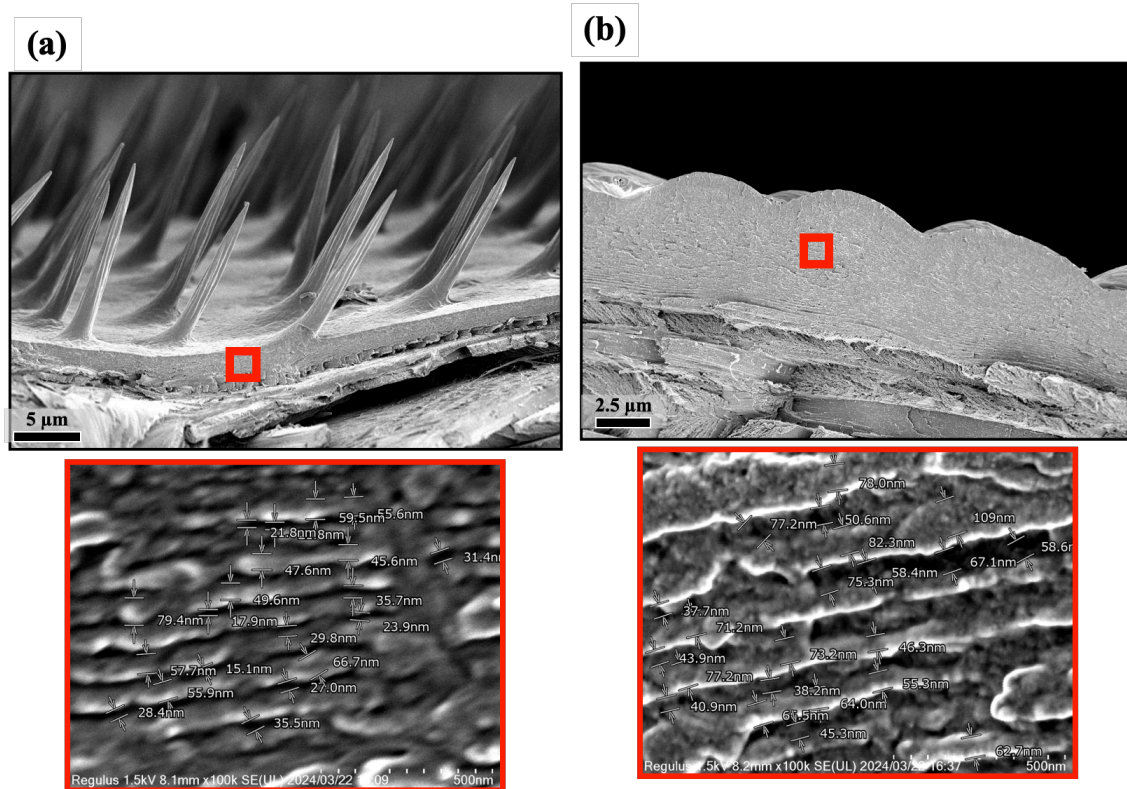


Fig. 3-43 (a, b) Cross-sectional SEM images of the (a) back and (b) front surfaces of a carrion beetle elytra. The red boxes are enlarged views, and the numbers in the figures indicate the thickness of each layer.

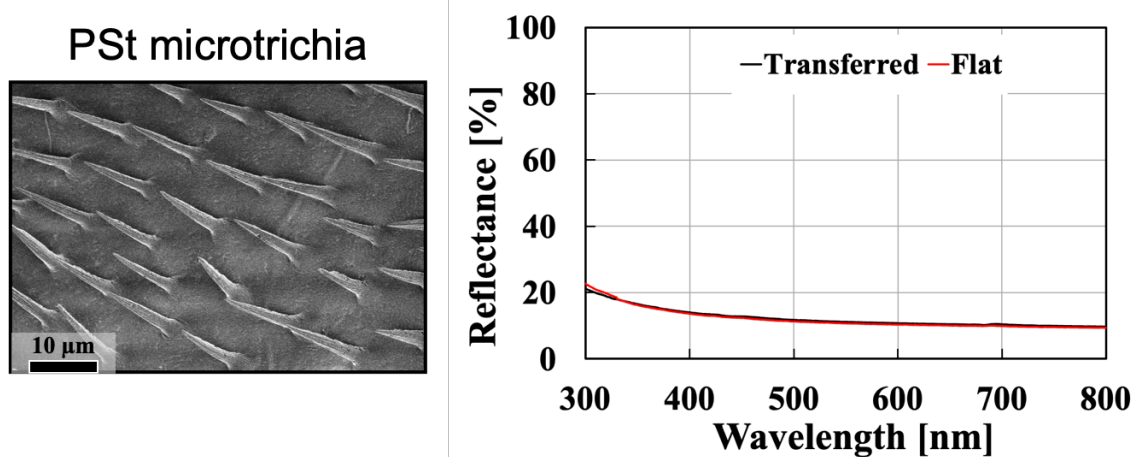


Fig. 3-42 SEM images and reflectance of microtrichia-mimicked structures prepared in PST

However, if the green color observed this time is structural color, the question arises as to why it is only present on the underside, which cannot be seen by other organisms, and why there is no reflection peak on the front side, which also has a multi-layer structure. Organisms that exhibit structural color include Morpho butterflies⁷, jewel beetles^{7,167}, and carabid beetles¹⁶⁸, and all of them benefit from showing their structural color to others. The benefits include blinding predators, warning colors, and visual appeal to the opposite sex. In areas where carabid beetles, which show red structural color, are found to live, red models are attacked by predators such as birds less frequently than black models, which is said to be a repellent behavior against carabid beetles that spray harmful body fluids¹⁶⁸. In addition, it has been reported that jewel beetles do not approach models made from metallic paper or leaves, but do approach jewel beetle wings, indicating that jewel beetles can distinguish structural color¹⁶⁹. However, as mentioned above, *Necrophila japonica* is said to hardly fly, so it is difficult for it to appeal to others while flying. Therefore, *Necrophila japonica*, which has structural color in a place that is difficult for other organisms to see, may not have had a multi-layer structure to produce structural color, but to obtain other functions. The color appeared because the multi-layer structure was the size that showed the structural color, but if the structural color was conspicuous, *Necrophila japonica*, which does not have a defense mechanism like the Japanese snail beetle, would likely be eaten by predators. Therefore, it is possible that the structural color was canceled out by the hierarchical structure, making the color of the front side, which is noticeable to others, less conspicuous. In particular, there are organisms that see color by contrast rather than peaks, and for them, a reflectance spectrum with no undulations, such as that shown in Fig. 3-41, may be effective. When the reflectance of the hierarchical structure was actually calculated, no reflectance peak was found regardless of whether the multi-layer structure was present or not (Fig. 3-44). The benefit of the multi-layer structure is thought to be an increase in the strength of the wings. In other words, the multi-layer structure avoids stress concentration, resulting in stronger wings like shell³. In addition, when combined with the results of friction research, it may also be involved in reducing friction by deformation resistance. There are also reports that the layered structure reduces friction. Further research will be required to discover detailed functions and clarify principles.

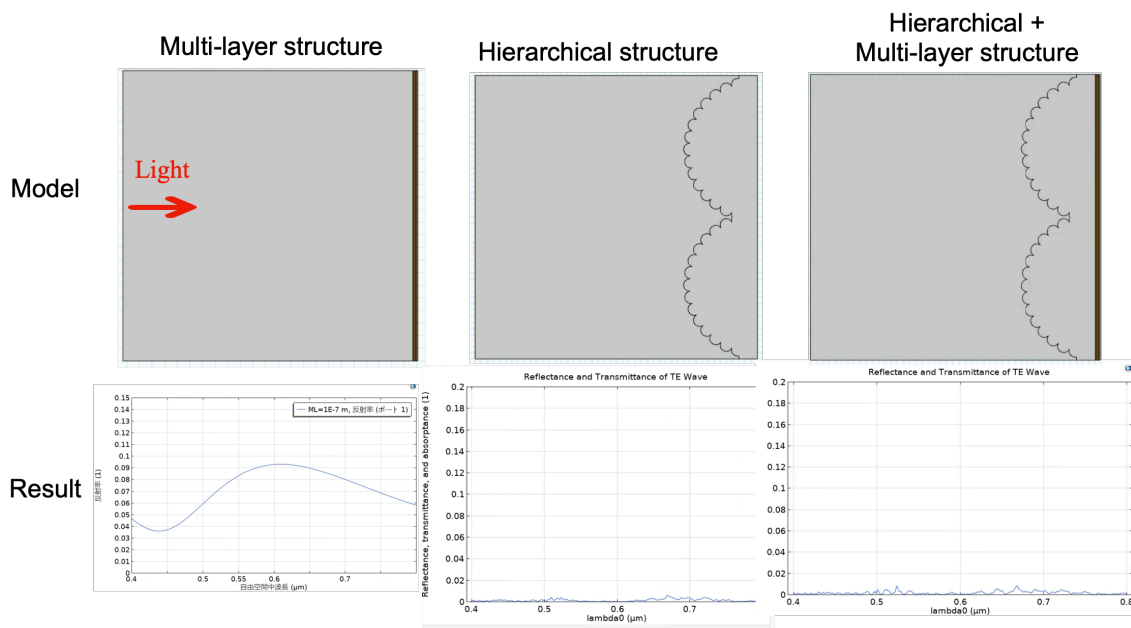


Fig. 3-44 Models and calculation results of reflectance simulation for multi-layer structure, hierarchical structure, and structure that has both multi-layer and hierarchical structures

3-7 Comparison with other organisms and studies

From the results of 3-2 and 3-3, the microstructures arranged on the back of *Necrophila japonica* may reduce energy loss due to friction when moving on forest surfaces where friction is likely to occur, such as under fallen leaves. Furthermore, such hierarchical structures are also present in springtails (Fig. 3-45)^{86,91} and slithering geckos (Fig. 3-46)⁹², which share the same habitat on the ground surface and in soil environments. Hierarchical structures such as those in lotus leaves (Fig. 3-47)⁹³ and slithering geckos⁹² were thought to be effective in reducing friction, but detailed studies had not been conducted. However, based on the results of this study, their hierarchical structures may also reduce friction. Springtails live in soil, so friction against the soil is a matter of life and death for them. The mushroom-shaped hierarchical structure may enable the simultaneous achievement of superhydrophobicity and friction reduction by reducing contact points and suppressing deformation, which was not observed in *Necrophila japonica*. As shown in Fig. 3-46 the slithering geckos also show a hierarchical structure on the dorsal side where friction is reduced, and in some cases this structure is raised against the abdominal skin and lateral sides where friction is increased. Our research has shown that the dome-shaped hierarchical structure is more effective in reducing contact points and suppressing deformation, so the surface microstructure of this organism is likely to have a friction-reducing effect. As for lotus leaves, they do not move and therefore do not reduce energy loss, but previous research has reported that plant wax and microstructures reduce the frictional force of insects. Therefore, it is possible that the moth-eye structure, which reduces friction and has sliding properties, prevents insects from eating plants. There are several studies that have investigated friction in hierarchical structures inspired by snake scales, but they do not consider the effect of the structure itself because they are not composed of a single material like ours. Furthermore, the materials used are elastically deformable materials, which have a different friction principle from the plastically deformable materials investigated in this study. A snake-scale-inspired material made of a plastically deformable material showed a friction reduction effect of ~40%, but this is a single-layer structure¹⁰⁹. The friction reduction effect of various other structures has been studied, but most of them are single-layer structures^{108,110}. In addition, lubricants have been used and mixed with different materials^{102,148,170}. Regarding dry friction reduction by surface microstructures, there has been much discussion about the reduction in contact area represented by the Hertz contact theory due to the microcontact state¹⁰⁹. However, there has been little discussion about deformation due to pressure concentrated in the reduced contact area, and we have explained it by incorporating knowledge from the field of structural geology. In this way, the result that the hierarchical structure is advantageous for suppressing deformation by load dispersion and reducing friction by reducing the contact area may enable feedback

to biology. In addition, anisotropic structures show anisotropy in friction force^{109,171,172}. This may provide a survival strategy for organisms by restricting or selecting the movement direction. Compared with previous studies¹²⁷, the structural shape differs depending on the sliding direction, which may affect the length and stiffness of stiction. The length of stiction was the same as the microstructure and indenter scale ratio in our study. In particular, for stiffness, the authors use a penalty method like ours to show the difference between the force acting on the entire surface and the force acting on the surface. Therefore, the anisotropy of the structure may also affect the load dispersion effect. Friction anisotropy due to structural anisotropy has also been demonstrated in snakes. The size of these anisotropic microstructures is submicron scale, similar to the wrinkled structure of *Necrophila japonica*^{109,171,172}. Since the load dispersion effect has not been considered in detail, our results may advance research on anisotropy in the future. Further research is needed to elucidate the effect of structure on anisotropy and the behavioral patterns of beetles, and future progress is expected. In addition, the results of this study can be applied to various fields such as architecture, medicine, and industry. Considering future applications to human society, it is advantageous to be able to reduce friction without using lubricants that cause environmental pollution¹⁰⁰. In addition, it is also important to create mimicked structures using self-organization like living organisms, rather than lithography and etching, which require cost, time, and special equipment, as a consideration for the environment³⁶⁻⁴⁰. Using high-cost, high-energy processing methods to reduce friction is counterproductive because it contradicts the purpose of friction reduction, which is to save energy. From this perspective, this research is different from others, and there is a high possibility that it has come closer to a living organism in terms of the method of production.

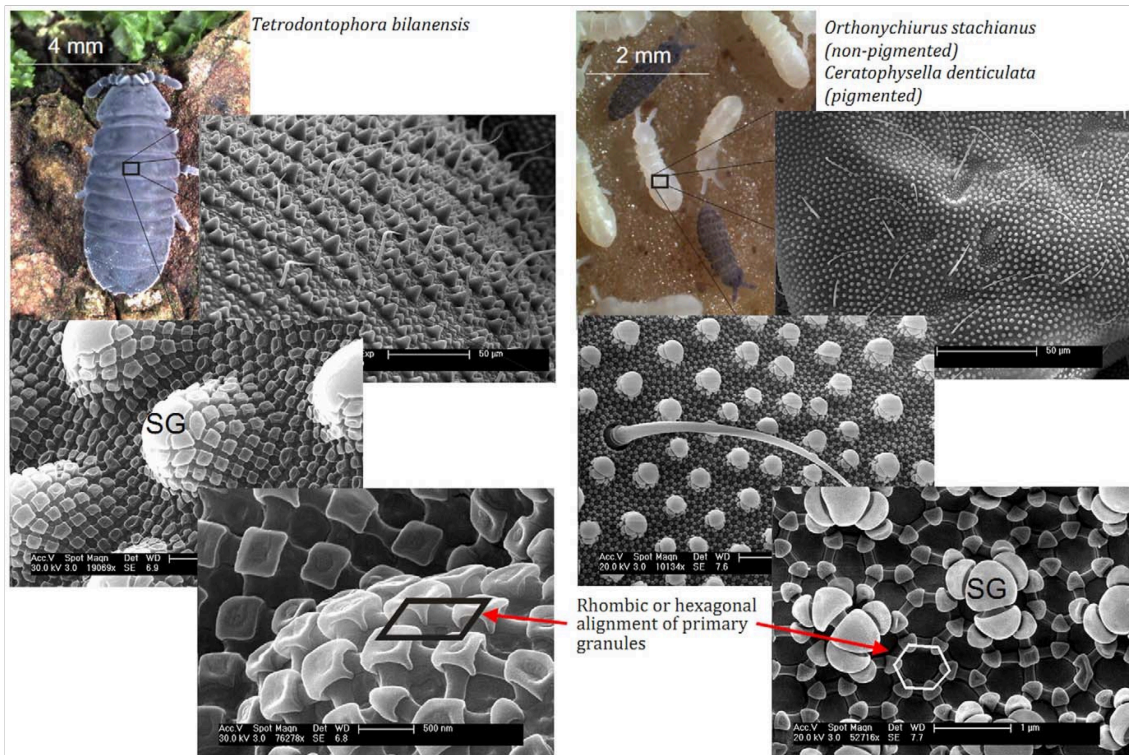


Fig. 3-45 Springtail skin combines bristles and a unique nanoscopic comb pattern. The rhombic or hexagonal comb pattern is formed by small primary granules connected by ridges. Additionally, some - but not all - species possess papillous secondary granules (SG), which can significantly differ in shape, depending on the specific habitat and body size of the respective species ^{86,91}

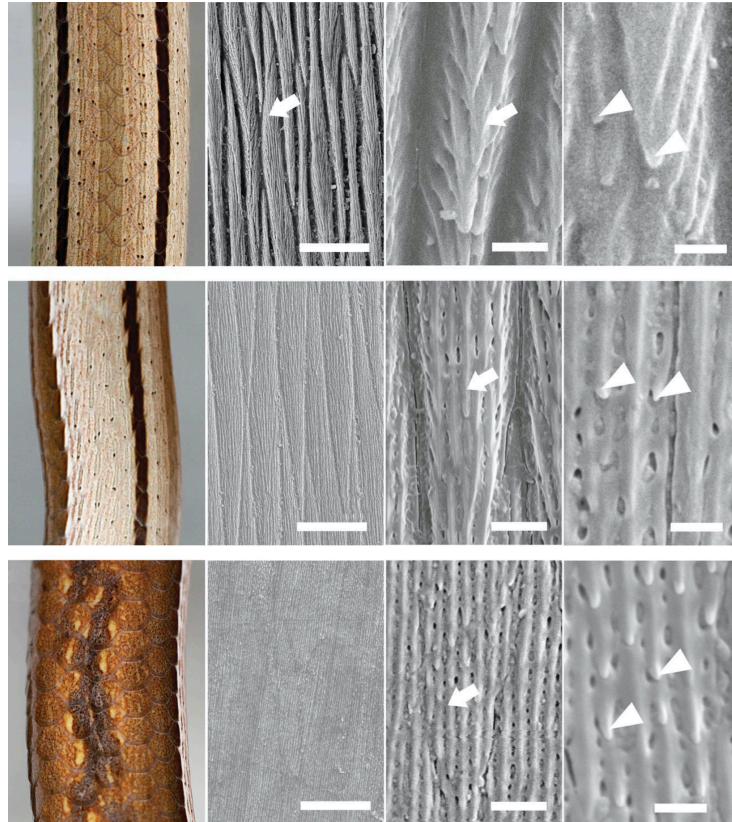


Fig. 3-46 Scales of *L. jicari* and scanning electron micrographs of their microornamentation at different magnifications at the (a) back, (b) the flank and (c) the belly. Back scales (a) feature bulges (B) covered with nanoridges (N). Ventral scales (c) feature only nanoridges (N). Free tips on nanoridges are marked in all scales of all regions with white arrow heads. In the images, the rostrad is on the upper part of the figure.⁹²

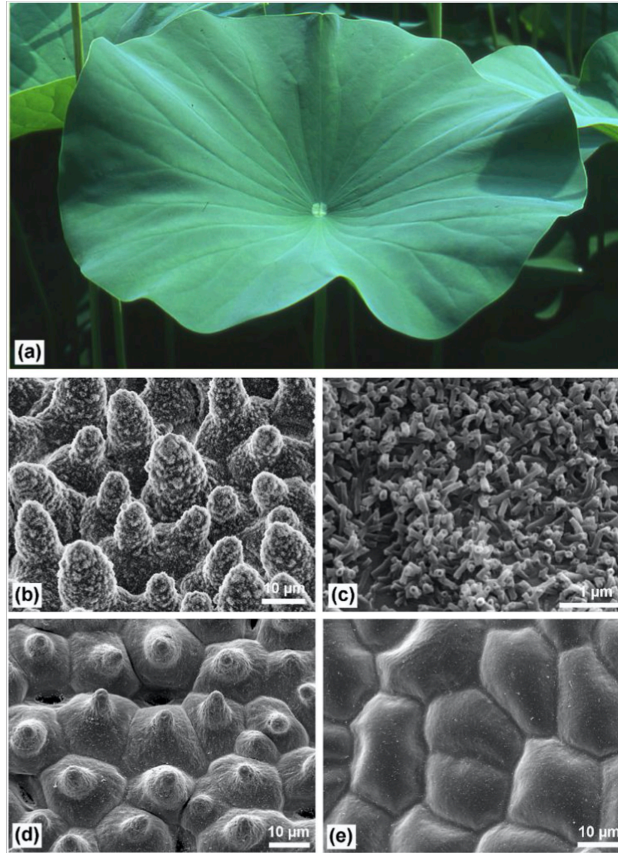


Fig. 3-47 The lotus plant. (a) Lotus leaves, which exhibit extraordinary water repellency on their upper side. (b) Scanning electron microscopy (SEM) image of the upper leaf side prepared by ‘glycerol substitution’ shows the hierarchical surface structure consisting of papillae, wax clusters and wax tubules. (c) Wax tubules on the upper leaf side. (d) Upper leaf side after critical-point (CP) drying. The wax tubules are dissolved, thus the stomata are more visible. Tilt angle 15°. (e) Leaf underside (CP dried) shows convex cells without stomata.¹⁷⁹

As a biological feedback, we compared the microstructure of *Silphidae perforata*, a closely related species that lives in the same environment (Fig. 3-48). *Silphidae perforata* inhabits Russia and northern China, and is thought to have migrated from the north to Hokkaido 90,000 years ago, and inhabits only Hokkaido in Japan^{83,173,174}. *Necrophila japonica* is found throughout Japan, but has never been found overseas¹⁷⁵. These two species are in the same family but different genus. They look quite similar, but the differences are that *Silphidae perforata* is slightly smaller than *Necrophila japonica*, and the upper wings of *Silphidae perforata* are bulging when viewed from above, while the upper wings of *Necrophila japonica* are almost parallel to the left and right until near the center. There is also a large difference in body color, with *Silphidae perforata* being black with a slight bronze tint, and *Necrophila japonica* being black with a slight bluish tint. In addition, the vertical grooves on the pronotum are clearly visible in *Necrophila japonica*, but are hardly seen in *Silphidae perforata*. In addition, by observing the upper surface of the elytra with SEM, we newly discovered that *Silphidae perforata* also has a microstructure (Fig. 3-48). The microstructure of *Silphidae perforata* is different from that of *Necrophila japonica*, and is only a dome structure, not a hierarchical structure. As the results of 3-2 and 3-3 show that the dome structure also has a friction reducing effect, it is highly likely that *Silphidae perforata* also reduces friction by using a microstructure. However, when the friction of *Silphidae perforata* was measured using the method of 3-2-1, it was shown that the microstructure of the body surface of *Silphidae perforata* does not have a friction reducing effect (Fig. 3-49). This is thought to be due to the influence of structures other than the dome structure. Both species have protrusions that resemble antennae, but while the area around the antennae of *Necrophila japonica* is a hole, the area around the antennae of *Silphidae perforata* is raised like a hill (Fig. 3-50). The spacing between the hills is as much as 100 μm . Previous studies have shown that when the size ratio between the indenter and the microstructure spacing is less than 100, friction increases due to the occurrence of interlocks and energy barriers^{109,127}. In this study, a 3 mm ball was used for the indenter so that the body surface could be measured without being hindered by nodes, and it is thought that friction did not decrease due to the hill-like structure being caught. In contrast, *Necrophila japonica* has a hole-like structure of several tens of microns, which makes it difficult for it to be caught, and the friction reduction effect of the hierarchical structure is more likely to occur. Therefore,

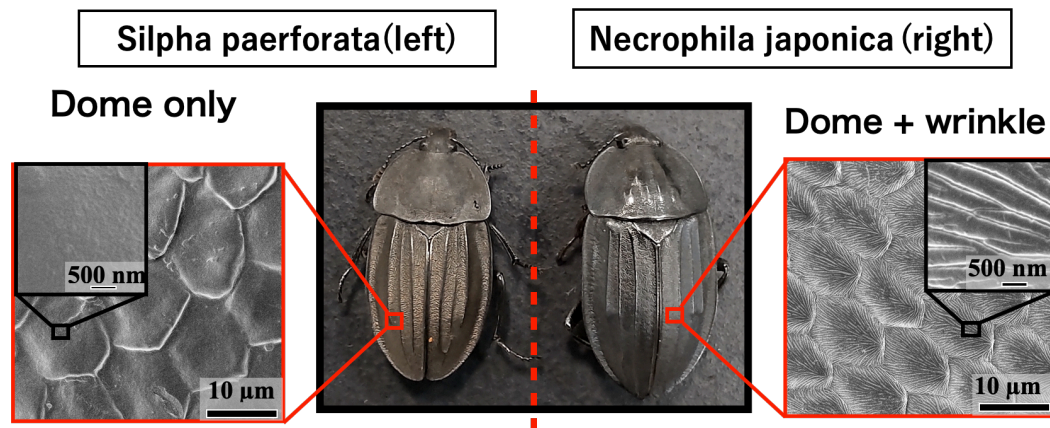


Fig. 3-48 Photographs and SEM images of *Silphidae perforata* and *Necrophila japonica*.

considering the results of this study, it can be said that *Necrophila japonica* has an elytra surface that is more likely to reduce friction than *Silphidae perforata*. The wrinkled structure formation process is thought to be a factor in the difference in the structure of *Silphidae perforata*. The wrinkled structure is created by the compression of two layers with different hardness^{121,122}. It is highly likely that the carrion beetle is compressed during metamorphosis, and there may be differences in the metamorphosis process between the two species mentioned above. Another difference between the two species is their flying ability. *Necrophila japonica* shows two types of flight muscles, but *Silphidae perforata* does not have developed flight muscles^{83–85,173,174}. Therefore, it is highly likely that *Necrophila japonica* uses its flight muscles to control turbulence during flight^{3,13} in addition to reducing friction, but since many papers have reported that *Necrophila japonica* does not fly even though it has flight muscles^{84,176}, we believe that its current function is mainly to reduce friction. There is little research on the ecology of *Silphidae perforata* and *Necrophila japonica*, so we cannot provide any definitive results at this time. By examining these two species and other species, we may be able to clarify how they have adapted to their environment during the course of evolution.

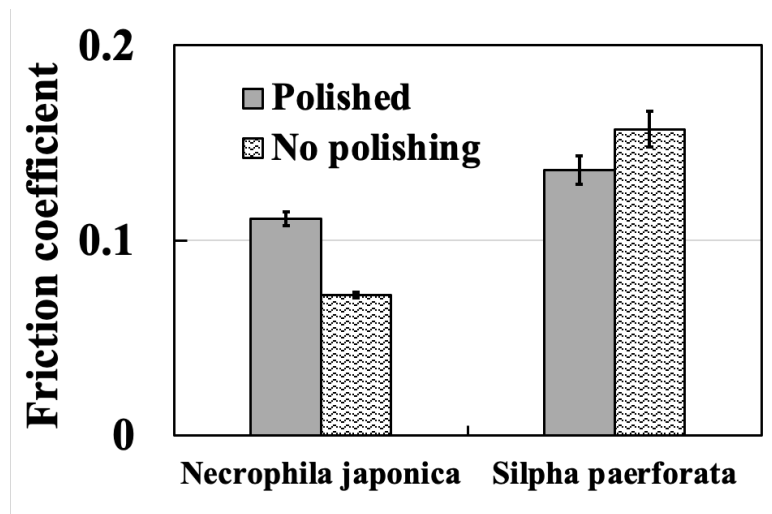


Fig. 3-49 Friction measurements of polished and no polishing samples of Silphidae perforata and *Necrophila japonica*.

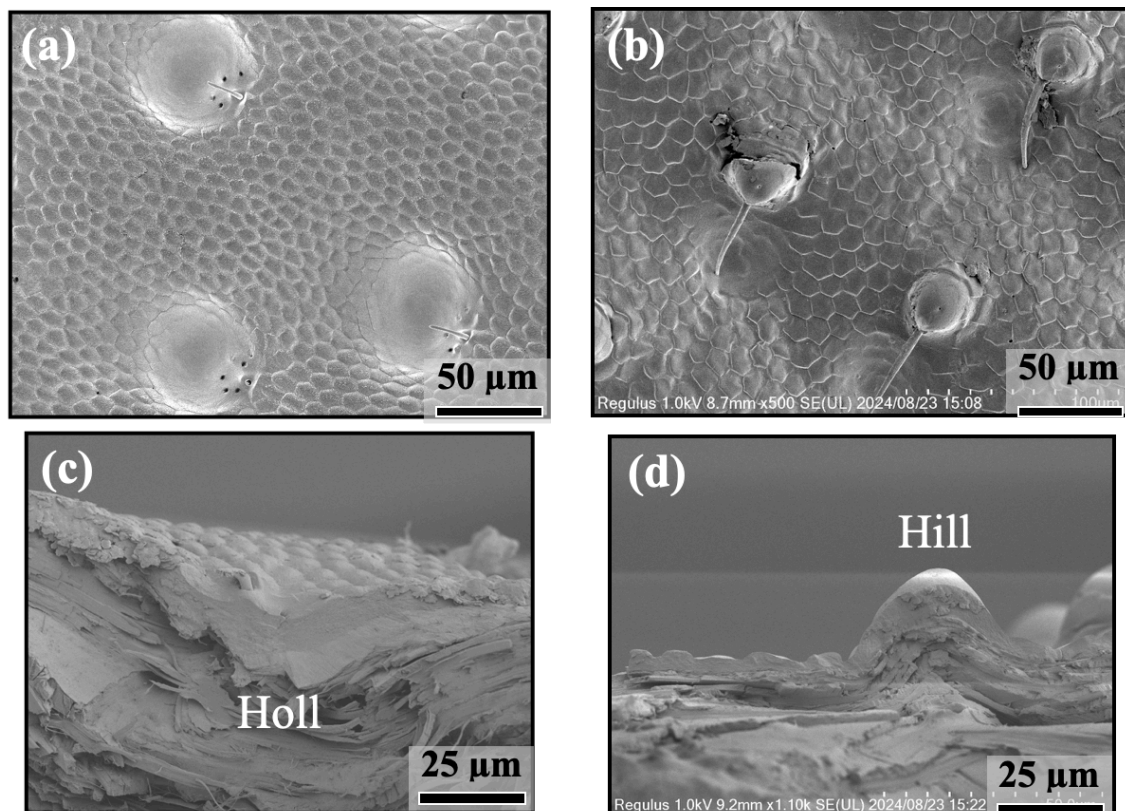


Fig. 3-50 SEM images of elytra of *Silphidae perforata* and *Necrophila japonica*. Both species have antennae, but in *Silphidae perforata* the antennae are hill-like, whereas in *Necrophila japonica* they are holes.

3-8 Summary

In conclusion, we have clarified for the first time the wettability-improving and friction-reducing functions of the hierarchical surface microstructure of *Necrophila japonica*. Friction force tests of mimicked structures made of the same plastic deformation material as living organisms showed that the hierarchical surface microstructure had a friction-reducing effect greater than that of a monolayer structure. By combining knowledge from the field of structural mechanics that the shape of the structure affects deformation with the basic principles of friction, we have newly clarified the friction characteristics and mechanisms of the hierarchical surface microstructure. The hierarchical surface microstructure of *Necrophila japonica* reduces friction more than a monolayer structure because it suppresses deformation and increases in the real contact area due to the effective hierarchical load dispersion effect on both domes and wrinkles, despite the large reduction in contact points. This will lead to the elucidation of the microstructural functions of many organisms, not just *Necrophila japonica*, and provide useful knowledge for reducing non-lubricated friction in human society. Our research focusing on living organisms makes it possible to find new connections between knowledge from different fields, namely friction reduction and load dispersion. In the field of metamaterials, excellent load-bearing properties due to various complex hierarchical structures have been reported^{119,120,177}, and further systematic research is expected to increase the degree of freedom in friction design (anisotropy and selective expression of required friction force) by selecting the size, shape, and material required to control friction. This load-dispersing effect has been demonstrated in our research using moth-eye structures¹⁷⁸, and this research confirms this and demonstrates the advantages of a hierarchical structure.

We also applied it to an elastically deformable material, which has a different material from *Necrophila japonica* and is a material that humans often use. The results showed that a hierarchical surface microstructure like that of *Necrophila japonica* has a greater friction reduction effect than a single-layer structure. This is because the hierarchical structure can suppress the increase in friction hysteresis and digging-in friction by deforming the upper wrinkle structure in the sliding direction, while at the same time reducing the effective contact area to a greater extent. These findings are supported by friction force tests across various structure sizes, the difference between static and kinetic friction forces, the anisotropy of friction force with respect to the sliding

direction, and simulation analysis. In particular, the friction force of the mimicked structure remained constant even after 100 sliding cycles, highlighting the excellent durability of the elastically deformable material. This mimicked structure also prevents stick-and-slip behavior. This means that the advantages of hierarchical structures can be obtained in materials other than living organisms. In hard materials, the microstructure provides hardness, preventing deformation and reducing friction, but in soft materials, the microstructure provides softness, making the material more flexible and easier to deform, reducing friction. This is similar to the microstructure of a lotus leaf, which becomes superhydrophilic if its surface is hydrophilic, and superhydrophobic if its surface is hydrophobic. The properties of the material and the properties of the microstructure enhance each other, successfully creating a highly functional and multifunctional material. This type of hierarchical structure is expected to be applied in various fields such as architecture, medicine, and industry. The key to reducing friction is reducing the contact area and mitigating friction hysteresis and digging in due to deformation in the sliding direction. If systematic research is further advanced, it is expected that it will be possible to precisely control friction properties (such as anisotropy and selective expression of the required friction force) by selecting the size and shape of the microstructure.

In addition, both of the above methods can reduce friction without using lubricants. In addition to solving problems such as environmental pollution, this is likely to contribute to reducing friction in situations where lubricants are difficult to use, such as in medical care. The fabrication process also has a major advantage. In this study, the fabrication was achieved using the same low-energy self-organization approach as seen in living organisms, and it is expected to be of great help in solving problems such as carbon neutrality. In particular, the breath figure method and wrinkle method used in this study are advantageous for industrialization compared to lithography and etching in that they can be easily scaled up. We believe that we have achieved results that go one step further than conventional biomimetic research, such as the low environmental impact and easy-to-apply fabrication process, and the fact that we have been able to express multifunctionality and high functionality by utilizing the properties of materials and structures without compromising them. We believe that this embodies the knowledge of ecomimetics and “perspective of living things”, which study not only the structure of living organisms but also their ecosystems and think from the perspective of insects.

Chapter 4

Conclusion

In this study, we used the surface microstructure of living organisms as an information source to conduct research on the harmony between the natural environment and human society. First, we observed and discovered the functions of the surface microstructure of living organisms, and clarified their principles through simulations, etc., with the aim of producing highly functional materials. This makes it easier to apply the technology of living organisms to human society, and by combining it with human knowledge, we can produce highly functional and multifunctional materials. In addition, by using a low-environmental-impact production method, we can learn about sustainability from living organisms by not simply imitating living organisms, but also imitating the ecosystems that surround them. Finally, by comparing the principles and functions obtained with those of other organisms, we clarify the importance of the information held by living organisms and ecosystems. This type of research is important for increasing the sustainability of human society.

In Chapter 2, we clarified a new adhesion mechanism learned from the surface microstructure of clingfish suckers and evaluated the function of a functional mimetic adhesive material. In 2-2, we produced a clingfish mimetic material, produced a polymer nanofilament covered with PDMS, and performed an adhesion test. As a result, it was revealed that the adhesive strength of the mimetic material is generated by PDMS, and that the nanofilament strengthens the adhesive strength of PDMS. The simulation results also revealed that the adhesive strength was enhanced by stress dispersion to the nanofilament. After that, each parameter of the mimicked structure was changed. The longer the nanofilament, the larger the contact area between the PDMS and the nanofilament, and the greater the amount of stress dispersion, which increased the adhesive strength, but the deformation stress was concentrated at the tip of the nanofilament, so there was a limit to the increase in adhesive strength. The harder the nanofilament, the greater the stress dispersion, and the greater the adhesive strength. Due to the relationship between the adhesive strength increase effect due to stress dispersion and the contact area, there was an appropriate PDMS thickness at which adhesive strength increased. The softer the PDMS, the greater the adhesive strength. Although the physical properties of mucus and PDMS are different, even when grease, which is not a solid like mucus, was used, the adhesive strength was higher with the nanofilament. From these results, it became clear that clingfish achieves both softness and hardness, which is the basic principle of adhesion, by using mucus and nanofilament, and that the adhesive

strength changes depending on the parameters of the mucus mimicking part and the nanofilament. In addition, the clingfish mimicked structure created in this study could be bonded underwater and on rough surfaces, and could be repeatedly bonded due to the protective effect of the nanofilaments in PDMS. The mimicked structure succeeded in shortening the bonding time underwater by transferring the MLAs structure to the surface. Based on the principle elucidated, a simple method using fine particles was used to create a high-performance adhesive material. These had the same or greater adhesive strength as the mimicked structure using nanofilaments, even on rough surfaces underwater, and could be repeatedly bonded. Simulations revealed that the fine particles were responsible for the hardness, as in the case of nanofilaments, but it is highly likely that the compliance changed in the case of fine particles, while the compliance did not change in the case of nanofilaments. The fact that it is possible to achieve excellent functions using the same principle even with fine particles, which have a structure different from clingfish, and that it has greater advantages in process and after use than nanofilaments, could greatly contribute to the research and development of functional adhesive materials in the future. This was achieved by strengthening the structure without compromising the material property of underwater adhesiveness of PDMS, as in living organisms. Therefore, it is easy to reproduce multi-functions and high functions, and the range of applications is wide. Finally, we compared it with other organisms, and we believe that this has made a great advance in the previously unclear effect of nanofilaments on adhesion in water. Our research has clarified this in engineering, but further investigation is needed in biology.

In Chapter 3, we clarified the friction reduction function of the hierarchical structure of the carrion beetle by combining knowledge from the field of structural mechanics that structural shape affects deformation, knowledge from the field of friction, and knowledge from the field of biology that it lives in an environment where friction is likely to occur. The hierarchical structure reduced friction more than single-layer structures such as wrinkled structures and MLAs against a surface without structure. Simulation revealed that the hierarchical structure reduced friction more than a single-layer structure because the effective hierarchical load dispersion effect on both domes and wrinkles suppresses the increase in deformation and real contact area, despite the large reduction in contact points. This will lead to the elucidation of the microstructural functions of many organisms, not just *Necrophila japonica*, and provide useful knowledge for reducing friction without lubrication in human society. We then applied

the hierarchical surface microstructure seen in *Necrophila japonica* to elastically deformable materials that are commonly used by humans. The results showed that the hierarchical structure had a greater friction reduction effect than the single-layer structure. This is because the wrinkled structure at the top deforms in the sliding direction, significantly reducing the contact area while suppressing friction hysteresis and digging-in friction. Friction force tests and simulation analysis showed that the friction coefficient of the mimicked structure remained constant even after repeated sliding, emphasizing its durability. This structure also prevented stick-and-slip behavior, confirming that the advantages of the hierarchical structure can be obtained even with different materials. In hard materials, the hardness of the microstructure suppressed deformation, while in soft materials, the ease of deformation due to the softness of the structure contributed to friction reduction. This was similar to the characteristic of the surface microstructure emphasizing wettability. Like living organisms, material properties and microstructures interact with each other, and we succeeded in developing a highly functional and multifunctional material. The advantage of this research is that friction can be reduced without the use of lubricants, which not only contributes to solving the problem of environmental pollution, but also allows for situations in which it is difficult to use lubricants, such as in medical care. In addition, there are great advantages in the fabrication process, and it is expected that utilizing the same low-energy self-organization approach as living organisms will contribute to the realization of carbon neutrality. In particular, the breath figure method and wrinkle method used in this study are easy to scale up and are more suitable for industrialization than lithography or etching.

In both of the adhesion enhancement and friction reduction methods used in this study, the stress applied changed depending on the structure, resulting in differences in contact force. Research that examines the effect of structure on deformation and stress dispersion in this way has been mainstream in the field of structural mechanics such as metamaterials, and has not been conducted much in the field of biomimetics, and there have also been few studies in the field of contact force. In this study, promoting the fusion of different fields, which is the key to biomimetics research, led to the discovery of a new principle. This research, which makes the most of the characteristics of materials and structures, is believed to have various extensibility, such as adjusting the structural shape and chemically modifying the surface. Therefore, we believe that the fusion of principles obtained from living organisms in various situations with the wisdom accumulated by

humans will lead to the creation of even more highly functional and multifunctional materials. Furthermore, the fact that the environmental impact is low and that multifunctionality and high functionality have been achieved by maximizing the properties of the materials and structure can be said to be a step forward in conventional biomimetic research. This research embodies ecomimetics, which learns not only from the structure of living organisms but also from the ecosystem as a whole, and a "perspective of living things" that incorporates the viewpoint of living organisms. We believe that this research will provide significant insight into mastering sustainability, a common challenge facing all of humanity.

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List of Publications

Journal Publication

• Chapter 2

Kazuma Tsujioka, Yasutaka Matsuo, Masatsugu Shimomura, and Yuji Hirai, “A new concept for an adhesive material inspired by clingfish sucker nanofilaments”, *Langmuir*, **2022**, 38, 3, 1215–1222. <https://doi.org/10.1021/acs.langmuir.1c02972>

辻岡一眞 平井悠司, "ウバウオの吸盤に学ぶ接着材料", *高分子(高分子学会出版)*, **2021**, 70 巻 7 月号 p365-366. In Japanese, <https://main.spsj.or.jp/c5/kobunshi/kobu2021/2107.html#365>

平井 悠司, 辻岡 一眞, 松尾 保孝, 下村 政嗣 ”模倣構造作製によるウバウオ吸盤表面の構造機能の解明” *日本接着学会誌*, **2023**, 59, 9, 310-317.

• Chapter 3

Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura and Yasutaka Matsuo, “Friction-reduction effect of the hierarchical surface microstructure of carrion beetle by controlling the real contact area”, *Nanoscale*, **2024**, 16, 45, 21021-21030. <https://doi.org/10.1039/D4NR02892A>

Tsujioka, K.; Koda, A.; Hirai, Y.; Shimomura, M.; Matsuo, Y. “Friction Reduction Effect Caused by Microcontact and Load Dispersion on the Moth-Eye Structure”. *Advanced Engineering Materials*, **2024**, 26 (20), 2401405. <https://doi.org/10.1002/adem.202401405>.

• Chapter 3 Derivation

Shimada, Y.; Tsujioka, K.; Matsuo, Y.; Shimomura, M.; Hirai, Y. “Influencing Role of Anisotropic Metal Microstructures on Friction Force”, *Molecular Crystals and Liquid Crystals* **2024**, 768 (5), 158–167. <https://doi.org/10.1080/15421406.2024.2323753>.

Nosaka, M.; Tsujioka, K.; Matsuo, Y.; Okamatsu, T.; Arita, T.; Shimomura, M.; Hirai, Y. “Effect of the Microstructures on Vulcanized Rubber Frictions”, *Langmuir* **2021**, 37 (21), 6459–6467. <https://doi.org/10.1021/acs.langmuir.1c00590>

Awards

- 1) 優秀ポスター賞 受賞(Poster award), 「有限要素法シミュレーションを用いたウバウオ接着メカニズムの解明」, 辻岡一眞, 松尾保孝, 平井悠司, 下村政嗣, 第 70 回高分子学会討論会, オンライン,ポスター, 2021.9
- 2) POSTER AWARD, “The effect of the deformation stress dispersion to the nanofilaments for strong adhesion in clingfish”, Kazuma Tsujioka, Yasutaka Matsuo, Yuji Hirai, Masatsugu Shimomura, 21st Chitose International Forum on Science & Technology (CIF21), Chitose (Japan), poster, (2021.10)
- 3) Student Poster Award (Material Award), “Analysis of resistance against deformation stress of nanofilaments inside clingfish mimicked tape by using FEM simulation”, Kazuma Tsujioka, Yasutaka Matsuo, Yuji Hirai, Masatsugu Shimomura, The 22nd International Conference on Nanoimprint and Nanoprint Technology(NNT2022), Toyama (Japan), poster, (2022.10)
- 4) IPC2023 Young Scientist Poster Award, “The effect of hierarchical surface microstructures of *Necrophila japonica* on friction properties”, Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura, Yasutaka Matsuo, The 13th SPSJ International Polymer Conference (IPC 2023), Sapporo (Japan), poster, (2023.7)
- 5) 最優秀講演賞 受賞(Oral award), 「ウバウオのように水中の粗い面でも強固に接着可能な高機能性粘着テープの作製」, 辻岡一眞, 平井悠司, 下村政嗣, 松尾保孝, 2023 年度 北海道高分子若手研究会, 札幌,オーラル, 2023.8
- 6) 2024 年度 Student Poster Award, オオヒラタシデムシ階層表面微細構造が示す摩擦低減効果の原理解明」, 辻岡一眞, 平井悠司, 下村政嗣, 松尾保孝, 2024 年第 2 回ナノインプリント技術研究会, 墨田区(東京),ポスター, 2024.5
- 7) 優秀ポスター賞 受賞 (Poster award), 「シデムシ科の上翅に見られる表面微細構造のもつ機能と原理の調査」, 辻岡一眞, 平井悠司, 松尾保孝, 2024 年北海道高分子若手研究会, 札幌,ポスター, 2024.9

Conference presentation (Only for which I am a speaker)

① International conference

- 1) “Preparation of Cellulose Nanocrystal/Chitosan Composite Multi-Functional Films”, Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura, 33th International Microprocesses and Nanotechnology Conference (MNC2020), online, poster, (2020.11)
- 2) “Characterization of hydrochloric acid hydrolyzed Cellulose Nanocrystal (CNC) / Chitosan composite film”, Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura, 21st Chitose International Forum on Science & Technology (CIF21), Chitose (Japan), oral, (2021.10)
- 3) “The effect of the deformation stress dispersion to the nanofilaments for strong adhesion in clingfish”, Kazuma Tsujioka, Yasutaka Matsuo, Yuji Hirai, Masatsugu Shimomura, 21st Chitose International Forum on Science & Technology (CIF21), Chitose (Japan), poster, (2021.10)
- 4) “A new adhesive mechanism learning from the nanofilaments and mucus of a clingfish sucker”, Kazuma Tsujioka, Yasutaka Matsuo, Yuji Hirai, Masatsugu Shimomura, 34th International Microprocesses and Nanotechnology Conference (MNC2021), online, oral, (2021.10)
- 5) “Investigation of the influence of the clingfish sucker nanofilament on adhesion”, Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura, The International Chemical Congress of Pacific Basin Societies 2021(Pacifichem 2021), online, poster, (2021.12)
- 6) “Analysis of resistance against deformation stress of nanofilaments inside clingfish mimicked tape by using FEM simulation”, Kazuma Tsujioka, Yasutaka Matsuo, Yuji Hirai, Masatsugu Shimomura, The 22nd International Conference on Nanoimprint and Nanoprint Technology(NNT2022), Toyama (Japan), poster, (2022.10)
- 7) “Investigation of the friction on *Necrophila japonica* surface microstructure”, Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura, Yasutaka Matsuo, 35th International Microprocesses and Nanotechnology Conference (MNC 2022), Tokushima (Japan), oral, (2022.11)
- 8) “The effect of hierarchical surface microstructures of *Necrophila japonica* on friction properties”, Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura, Yasutaka Matsuo,

- The 13th SPSJ International Polymer Conference (IPC 2023), Sapporo (Japan), poster, (2023.7)
- 9) Materials for Electronics and Photonics (KJF-ICOME2023), Fukuoka (Japan), poster, (2023.8)
 - 10) “Particle-embedded underwater adhesive tape based on the adhesive strength increase mechanism inspired by clingfish”, Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura, Yasutaka Matsuo, 2023 KJF-International Conference on Organic Materials for Electronics and Photonics (KJF-ICOME2023), Fukuoka (Japan), poster, (2023.8)
 - 11) “Comparison of clingfish-learning adhesive materials using nanoparticles or nanofilaments on adhesive properties”, Kazuma Tsujioka, Yuji Hirai, Masatsugu Shimomura, Yasutaka Matsuo, The 24th RIES-HOKUDAI International Symposium, Sapporo (Japan), poster, (2023.12)
 - 12) “Friction reduction by load dispersion effects learning from the hierarchical surface microstructure of carrion beetle”, Kazuma Tsujioka, Yuji Hirai, Yasutaka Matsuo, The Third International Conference on Nature Inspired Surface Engineering (NISE 2024), Granada, (Spain), poster, (2024.11)

② Domestic conferences in Japan

- 1) 「セルロースナノクリスタルとキトサンを用いた高透明フィルムの作製」, 辻岡一眞, 平井悠司, 下村政嗣, 2019 年度第 54 回北海道支部研究発表会, 札幌,ポスター, 2020.1
- 2) 「セルロースナノクリスタル/キトサン混合フィルムの作製」, 辻岡一眞, 平井悠司, 下村政嗣, 第 69 回高分子学会年次大会, オンライン, ポスター, 2020.5
- 3) 「ウバウオから着想を得た新規接着メカニズムの解明」, 辻岡一眞, 平井悠司, 下村政嗣, 高分子学会 2020 年度第 55 回北海道支部研究発表会, オンライン,オーラル, 2021.1
- 4) 「透明性、高い材料強度、シール性を有するセルロースナノクリスタル/キトサンコンポジットフィルム」, 辻岡一眞, 平井悠司, 下村政嗣, 第 70 回高分子学会年次大会, オンライン,ポスター, 2021.5

- 5) 「有限要素法シミュレーションを用いたウバウオ接着メカニズムの解明」, 辻岡一眞, 松尾保孝, 平井悠司, 下村政嗣, 第 70 回高分子学会討論会, オンライン,ポスター, 2021.9
- 6) 「オオヒラタシデムシの表面微細構造における濡れ・摩擦特性の調査」, 辻岡一眞, 平井悠司, 下村政嗣, 松尾保孝, 第 71 回高分子学会討論会, 札幌, ポスター, 2022.9
- 7) 「オオヒラタシデムシから学んだ摩擦低減効果を持つ階層的な表面微細構造」, 辻岡一眞, 平井悠司, 下村政嗣, 松尾保孝, 第 72 回高分子学会年次大会, 高崎,ポスター, 2023.5
- 8) 「ウバウオのように水中の粗い面でも強固に接着可能な高機能性粘着テープの作製」, 辻岡一眞, 平井悠司, 下村政嗣, 松尾保孝, 2023 年度 北海道高分子若手研究会, 札幌,オーラル, 2023.8
- 9) 「オオヒラタシデムシから学ぶ階層的な表面微細構造による摩擦低減」, 辻岡一眞, 平井悠司, 下村政嗣, 松尾保孝, 第 72 回高分子討論会, 高松,オーラル, 2023.9
- 10) 「オオヒラタシデムシ階層表面微細構造が示す摩擦低減効果の原理解明」, 辻岡一眞, 平井悠司, 下村政嗣, 松尾保孝, 2024 年第 2 回ナノインプリント技術研究会, 墨田区(東京),ポスター, 2024.5
- 11) 「オオヒラタシデムシ階層表面微細構造が示す摩擦低減効果の原理解明」, 辻岡一眞, 平井悠司, 下村政嗣, 松尾保孝, 第 73 回高分子年次大会, 仙台, ポスター, 2024.6
- 12) 「シデムシ科の上翅に見られる表面微細構造のもつ機能と原理の調査」, 辻岡一眞, 平井悠司, 松尾保孝, 2024 年北海道高分子若手研究会, 札幌,ポスター, 2024.9
- 13) 「オオヒラタシデムシから学ぶ階層的な表面微細構造による摩擦低減」, 辻岡一眞, 平井悠司, 松尾保孝, 第 72 回高分子討論会, 高松,オーラル, 2024.9

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