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2017

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Synthesiology is a journal that describes the objectives and social value of research activities that attempt to utilize the results in society, the specific scenarios and research procedures, and the process of synthesis and integration of elemental technologies. To allow the readers to see the value of the papers in a glance, the highlights of the papers characteristic to *Synthesiology* are extracted and presented by the Editorial Board.

***Synthesiology* Editorial Board**

Development of a compact, onboard slurry icemaker to rapidly produce optimal ice for maintaining freshness of marine products

Hiroshi Nagaishi *et al.*

This paper describes the process whereby the use of frozen seawater in slurry form was considered to maintain the freshness of marine products, and an icemaker that is low in cost, that can supply slurry ice stably, and that can be installed on fishing vessels was developed. To solve the issues that were difficult to solve by conventional technology, a new icemaker was successfully developed by fusing the technologies of mechatronics, icemaking, and freshness assessment. The paper is detailed documentation of a success story of industry-academia-government collaboration where a company, a public research institute, and a national research institute brought the techniques and know-how of their respective expertise together and worked toward one goal.

Standardization of dimethyl ether (DME) fuel specifications

Mitsuharu Oguma

Dimethyl ether is regarded promising for various uses as one of the alternatives to fossil fuel, but standardization of fuel quality is necessary in introducing this product to the market. This paper describes the details of the international standardization efforts. It is a very interesting paper that also discusses the process of defining fuel quality and test methods, details of round robin tests by institutions around the world, the adjustments among multiple countries, and the difficulty of standardization when multiple stakeholders are involved.

A study on high-density recording with particulate tape media for data storage systems
—On the process of introducing barium-ferrite tape media to the market—

Takeshi Harasawa *et al*.

The paper presents the re-accelerated achievement of high density in magnetic tape systems, where barium-ferrite was focused on as new material that surpassed the performance of metal magnetic material that was showing limit in achieving high density. This development was in response to the rapid increase of information and data handled in society. To realize the barium-ferrite tape, materials and peripheral technologies were developed, including tape manufacturing technology within the company and high-sensitive magnetic head and signal processing technologies outside the company. The paper outlines the process by which the de facto standardization of barium-ferrite tape was established for data storage.

Development of a cell microarray chip system for early and accurate malaria diagnosis
—Finding one parasite in 2 million erythrocytes for elimination of malaria—

Muneaki Hashimoto *et al.*

This paper explains the development of a quick and highly sensitive diagnostic method for malaria, which is one of the three major infectious diseases. Under the concept that it is necessary to determine the severity of infection as well as the presence of infection in order to be usable at medical settings in developing countries, a series of research was conducted including comparison of various diagnostic methods, development of a cellular chip, development of a detection method using the chip, and a verification test in Africa jointly with universities. The paper details the efforts toward social implementation of a new diagnostic method.

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Contents

Development of a compact, onboard slurry icemaker to rapidly produce optimal ice for maintaining freshness of marine products

Hiroshi NAGAISHI[1]*, Takaaki INADA[2], Takeya YOSHIOKA[3] and Atsushi SATO[4]

[Translation from *Synthesiology*, Vol.10, No.1, p.1–10 (2017)]

Marine products require optimal cold storage transport to maintain freshness and minimize damage. Slurry ice is suitable for this purpose due to its softness and immediate cooling capability. This paper describes the process from design to commercialization of a compact icemaker that continuously, stably, and rapidly produces slurry ice and can be placed on fishing vessels. This icemaker is expected to increase the competitiveness of both domestic and international cold chains by advancing both food safety and security.

Keywords : icemaker, slurry ice, freshness, marine products, cold chain

1 Introduction

In 1965, the Resources Council of the Science and Technology Agency in Japan announced the need for modernization of the transport and distribution system of food (aka cold chain recommendations) to systematically improve the eating habits of Japanese people by ensuring healthy and desirable dietary modification. Thereafter, quality control of food in the distribution system was enforced. In this quality control, broad distribution and long-term preservation have been achieved by minimizing the deterioration of perishables via temperature control during refrigeration or freezing. The consumer's need for food safety and security in the cold chain system has been attained by technological advances as a result of having improved infrastructure for sufficient traceability of transportation routes and their environmental conditions, and also by implementation of severe quality control procedures such as HACCP[Term 1] and the food safety management system of ISO22000. Furthermore, fresh fish will surely have a higher market value. Suitable processing from the time that fish are caught until consumption is critical for maintaining such freshness. To prevent degradation and to maintain quality of the fish, crushed ice and/or the plate ice made from fresh water is generally loaded onto fishing vessels upon their departure from port, and then the caught fish are placed in a tank (on the vessel) cooled with the ice and kept cold until the ship returns to port. An alternative handling procedure called "ike-jime (spiking)" effectively maintains a higher freshness of raw fish by preventing violent motions by the fish itself. This procedure (1) delays the decrease in ATP,[Term 2] which is an energy source for a living body, (2) delays the increase in lactic acid in the fish body, and (3) delays the time until rigor mortis. An additional process called "bleeding" of spiked fish would delay the breeding of microbes that cause deterioration, and would thus yield a higher level of freshness in the fish for a longer time compared with when a fish dies naturally. Although spiking is generally recognized as effective, it is time-consuming and thus expensive because it must be done fish by fish, and consequently is generally not carried out on fish other than expensive and/or relatively large-scale fish. To maintain freshness as high as that achieved using the spiking procedure, the time in which a fish moves violently until it dies must be as short as possible. Slurry ice with small particle diameter and high fluidity (i.e., sherbet-like ice) is suitable for this purpose due to its high contact frequency and high rate of heat exchange with the fish for rapid chilling. If such slurry ice is used in the marine product industry, the freshness of fish can be kept high by preventing deterioration of the fish. Utilization of slurry ice for freshness produces a competitive advantage in this industry domestically as well as internationally.

In this paper, the development of a compact icemaker that produces slurry ice from seawater stably and continuously and that can be easily installed on a fishing vessel is described along with its design policy and the effects of such ice in maintaining freshness of marine products.

2 Development background

Development of an icemaker that produces ice to maintain the freshness of fish is expected to yield higher fish prices, specifically the development of a compact icemaker made in Japan to rapidly produce optimal ice onboard a fishing vessel. As early as the 1980s, the effectiveness of slurry

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ice was already known[1][2] and had attracted attention in Japan. However, most icemakers at that time were located at facilities at ports and required 20–24 hours to produce the required amount of ice for a fishing vessel. As a result, a large-scale tank was necessary to store the ice, thus increasing the cost on the facility-side (Table 1). An icemaker that could supply suitable ice at low cost was not yet available at the commercial level. Even now, a large amount of ice is still used in the marine product industry. Such systems degrade the efficiency of workers by wasting time having to wait for ice to be produced and to load ice onto a fishing vessel. Furthermore, knowing the amount of ice required by a vessel is difficult because prediction of the amount of caught fish is not always accurate. Excess loading of ice increases the cost of both the ice and fuel, whereas insufficient loading decreases the commercial value of the caught fish because their quality cannot be guaranteed. In contrast, if an onboard icemaker is installed in a fishing vessel to provide ice produced from seawater during the fishing voyage, the following time and cost advantages can be realized: (1) short ice-loading time, (2) reduction in fuel consumption, and (3) onboard adjustment in the amount of ice based on the actual fish catch.

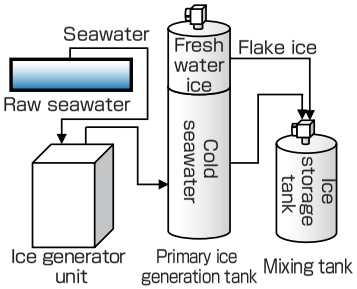
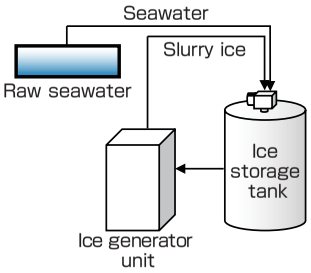
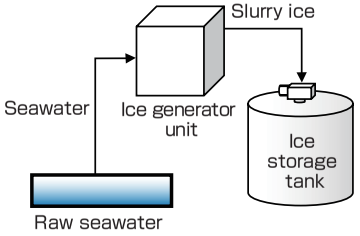
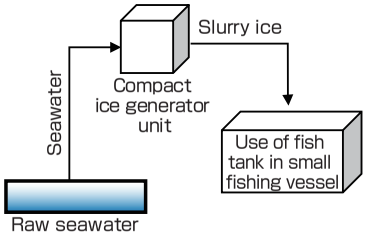
Nikko Co., Ltd. developed technology for fish processing to increase the commercial value of marine products and for the food industry. The company focused on technology to maintain freshness of fish because freshness is considered top priority in terms of commercial value of marine products. The company already had a commercial system for maintaining freshness of vegetables, in which the vegetables were enveloped by pulverized fresh-water ice to maintain an optimum temperature and humidity environment. To apply this technique to maintain freshness of marine products, Nikko surveyed existing icemakers. At that time, several icemakers that could produce slurry ice were developed by other international companies and were commercially available both domestically and internationally. As shown in Table 1, these icemakers produced at that time were all similar in that they used fundamental technologies of ice generation used in Canada, Germany and Iceland, with only a slight difference in the constituting unit and structure of the icemaker. Some patents from these countries were protected, but others were not due to their expiration. The technique to control the actual ice generation mechanism apparently depended on know-how, and thus numerous sections of the mechanism remained unclear, especially details of the ice nucleation process. To determine how to improve these icemakers, starting in 2008, Nikko investigated the ice-making capacity of the icemakers by collecting technical data such as operating temperature, flow rate of feeding sea-water, concentration of sodium chloride and other operating conditions. In addition, interviews were conducted with a person involved in fishery who had used a similar icemaker and ice. Based on the collected data and information, basic specifications of an icemaker and its generated ice were stated, thus defining the (1) performance parameters of the icemaker, such as ice generation rate and capacity per unit time, (2) characteristics of the generated ice, such as ice particle size, ice fraction and temperature, and (3) procedures for using such ice, such as the ice-to-fish ratio and environmental temperature. However, because the internal workings of the generator of the icemaker during ice production could not be observed directly, the ice-generation mechanism and how to improve the ice generation method based on this mechanism remained unclear. Additional unknowns include the technique used to evaluate freshness of the fish, knowledge about the usage of the ice, and knowledge about the effect of the ice in maintaining the freshness of fish and/or marine products. Therefore, Nikko could not completely design the target icemaker alone based on familiar mechatronics techniques.[3][4]

In contrast, the National Institute of Advanced Industrial Science and Technology (AIST) had performed fundamental research on ice nucleation and crystal growth phenomenon, and had gained significant knowledge, such as generation of slurry ice using ultrasonic vibration,[5]–[7] control of generation and growth of ice using antifreeze proteins and a synthetic polymer,[8]–[10] and generation of functional ice that contains bubbles.[11][12] Based on these research activities, in-depth knowledge was obtained on ice-making control technology, especially control of nucleation and crystal growth of ice in the early stages that influences the ice properties, such as cooling capability, that depend on the particle diameter of ice. Such knowledge and technology determined the direction of research of ice used to maintain the freshness of fish. AIST determined the design policy and stated the basic specifications in the development of a prototype icemaker.

The Hokkaido Industrial Technology Center in the Hakodate Regional Industry Promotion Organization (HITEC) has been working on quality preservation technology for fresh fish and shellfish for many years. For example, they quantified freshness and its decline mechanism for squid, and consequently used the resulting data to develop a system to maintain the freshness of squid.[13] Data on maintaining the freshness of fish in general could not be obtained at that time because there was no domestically developed system that could produce enough slurry ice for such a purpose at a reasonable cost. Furthermore, properties of the most suitable ice as well as ice-generation conditions were not yet known.

The above three institutions thus had their respective specialized technical fields, and exchanged information to collaborate in the development of an icemaker as shown in Fig. 1. Such collaboration enabled investigating standards of the ice used for temperature control and freshness management for various species of fish having different body size to plan utilization in the cold chain.

Table 1. Comparison of overseas major slurry icemakers (as of 2008)

Manufacturer	Company A in Canada	Company B in Germany	Company C in Iceland	Developed icemaker (target)
Ice-generation method	Raw seawater is sent to the ice generator unit, and generated slurry ice is sent to an ice generation tank. Ice with no salinity in the separated upper part is scraped off by a scraper, and mixed with cold seawater and flake ice in a mixing tank, resulting in slurry ice. 	Raw seawater is stored in an ice storage tank. Circulation is repeated between this tank and the ice generator unit. About 20–24 hours are needed to generate slurry ice. 	Slurry ice is produced continuously several minutes after startup by direct supply of seawater to the ice generation unit. 	Slurry ice is produced continuously several minutes after startup by direct supply of seawater to the compact ice generation unit. 
Features	・Slurry ice fraction can be adjusted (0–100 %). ・Slurry ice tends to melt easily. ・Large equipment with complex features make inspection & maintenance difficult. ・Slurry ice can be generated continuously. If the generation tank is halted even when ice generation is not needed, the scraper must continue to function because it will freeze otherwise. ・Salinity adjustment of cold seawater of the ice generation tank is necessary.	・Slurry ice generation takes a long time (20–24 hours) due to the circulation system. Adjustment in production schedule is therefore difficult. ・Generation temperature of the sherbet ice can be adjusted.	・Compact facilities can produce slurry ice instantly by connecting seawater to ice generation unit. ・Concentration of slurry ice can be quickly adjusted as requested.	・Compact with high ice-generation rate. ・Compact unit installable on small fishing vessels less than 20 tons. ・Dense sherbet ice generated faster than conventional equipment. ・Refrigeration and ice generation units are separate, resulting in space-saving installation.
Icy property, performance	ca. 50 μm, –1.5 ℃ 10 kw (7.5 t/d ice-generation standard power)	ca. 1 mm, –3.2 ℃ 18 kw (7.5 t/d ice-generation standard power)	ca. 10 μm, –3.2 ℃ 8 kw (7.5 t/d ice-generation standard power)	ca. 10 μm, –3.2 ℃ 8 kw (7.5 t/d ice-generation standard power)
Installation on a fishing vessel	Impractical	Impractical	Practical (100-ton class)	Practical (20-ton class)
Remarks	Large primary ice generation tank needed. Mixing tank for slurry ice generation is required.	Ice storage tank and ice generation tank (combined use) are required.	In land-installation use, ice storage tank is required due to low ice-generation rate.	By using a fish tank as ice storage tank, even a small fishing vessel can utilize this compact icemaker.

3 Development of an icemaker

To determine the optimal shape and size of ice particles in slurry ice produced from seawater, fundamental knowledge of ice formation in seawater must be known. Such knowledge is required to determine the cooling conditions necessary for producing such ice particles, and in developing a built-in control system in the icemaker in which the cooling conditions, which depend on the salt concentration and seawater temperature, can be determined in real time.

Ice formation proceeds via ice nucleation followed by ice growth. For water at subzero temperatures, the solid phase (ice) is more thermodynamically stable than the liquid phase, because the free energy of the solid phase per molecule is lower than that of the liquid phase. However, when a small ice embryo forms and grows in the liquid phase, the total free energy of the system increases as the size of the ice embryo increases, due to the increase in interfacial free energy. Therefore, water is often supercooled without ice nucleation even at subzero temperatures. On the contrary, if the ice embryo size exceeds a critical value, which depends on the degree of supercooling, the total free energy decreases as the ice embryo size increases, and thus ice nucleation proceeds.[14] It is extremely rare that ice nucleation occurs inside homogeneous liquid water. In most cases, ice nucleation occurs on insoluble solid surfaces as heterogeneous nucleation, because the increase in interfacial free energy is mitigated to some extent due to the existence of solid surfaces. After nucleation, ice grows in water accompanied by the diffusion of the latent heat, and also by the diffusion of salt molecules in the case of seawater.

Ice formation from seawater on a cooling solid surface is illustrated in Fig. 2. First, heterogeneous ice nucleation is initiated on the solid surface (Fig. 2a). The ice nucleation temperature is determined by the properties of the solid surfaces, and by the salt concentration of seawater. The subsequent ice growth pattern is influenced by the local growth rate, which is determined by the surface integration of water molecules into ice, the diffusion of latent heat, and the diffusion of solute molecules. When considering ice growth from seawater, the growth rate is dominated by the latter two factors, which are influenced by numerous factors, such as the ice nucleation temperature, salt concentration, flow rate of seawater, and cooling conditions. Therefore, if these factors are adequately controlled, the shape and size of ice particles produced in an icemaker could be optimized. In addition, the force required to peel off the ice from the solid surfaces can be reduced by controlling the ice growth pattern. Based on the above discussion, a built-in control system for an icemaker was developed here to control the cooling conditions in real time.

In the design of a prototype icemaker, the focus was on compact size and on ice generation rate, which are in a

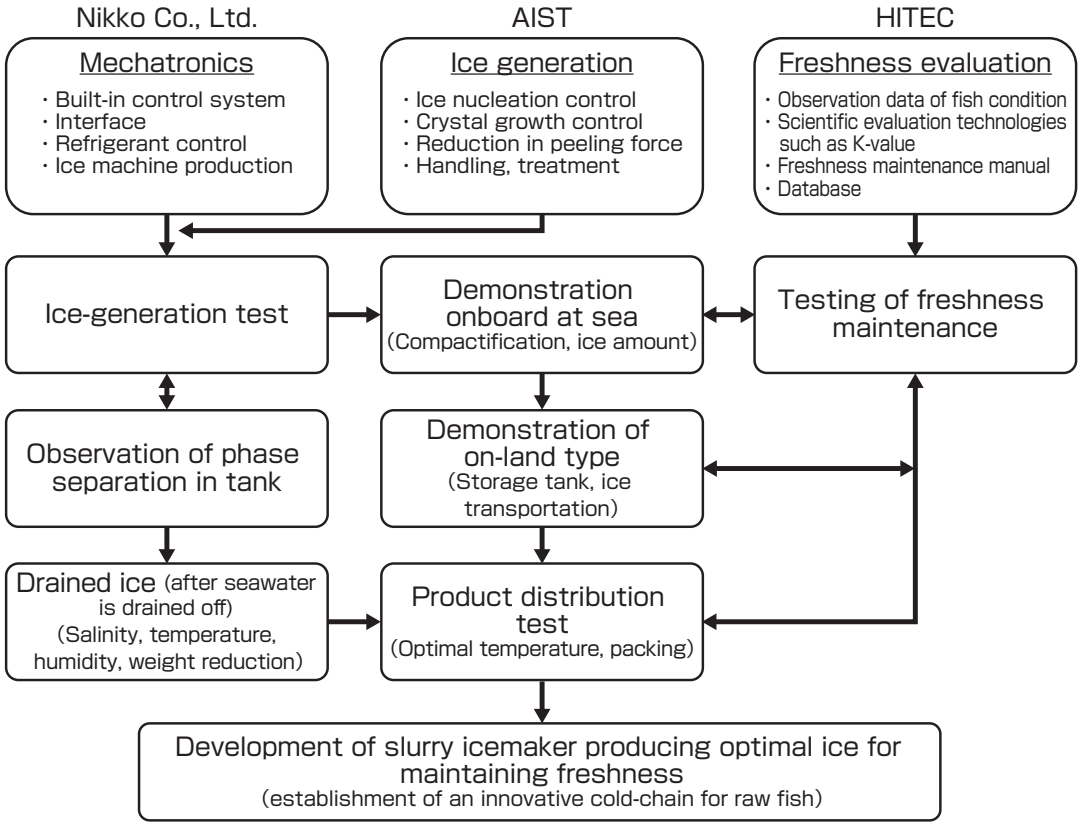


Fig. 1 Scenario of development and introduction of a compact slurry icemaker

trade-off relationship. The icemaker should fit in a narrow empty space of a fishing vessel, taking into consideration the configuration and installation direction of the icemaker to avoid waste of space, without adversely affecting the ice generation rate. Three specifications for the icemaker in this project were set: (1) produce slurry ice appropriate for maintaining freshness of raw fish, (2) have a simple configuration that reduces structural materials and reduces manufacturing costs, and (3) have a high ice generation rate. As for specification (1), the properties of slurry ice can be characterized by several parameters, such as the ice particle size, ice fraction, and temperature. The smaller the ice particle size, the higher the cooling rate of fish. In practice, the ice particle size should range between 10 and 100 μm, but must be at least smaller than 1 mm. The temperature of slurry ice should be lower than the lethal temperature of fish, although both the lethal temperature and time depend on the species of fish. Because slurry ice produced from seawater is actually a suspension in which ice particles are dispersed in seawater that mainly includes sodium chloride (NaCl) as solute, the ice temperature is below 0 °C. The temperature of slurry ice produced from seawater can be calculated based on the initial concentration of NaCl and the ice fraction. For example, when the initial NaCl concentration is 3.5 wt%, the freezing point becomes −2.2 °C, assuming $\Delta T = Km$,[Footnote)] where ΔT is the depression of the freezing point, m is the molality of the solute [mol/kg], and K is the molal freezing point depression constant (K=1.85 K·kg/mol when the solvent is water). By controlling the ice fraction, the concentration of NaCl in the unfrozen part of seawater can be adjusted, and thus the temperature of slurry ice can be controlled in accordance with the above relationship between ΔT and m.

To satisfy specification (2), it is necessary to consider the following three points so as to effectively cool the source seawater even in a simple icemaker configuration:
(a) Increase the effective surface area of the heat exchanger in the generator of slurry ice.
(b) Use materials with high thermal conductivity for the components of the heat exchanger.
(c) Increase the temperature gradient between the source seawater and the refrigerant, by decreasing the evaporation temperature of the refrigerant.
Generally, scaling up of a heat exchanger or adoption of a multi-tube type is effective in increasing the surface area of the heat exchanger. However, we adopted a double-tube heat exchanger using direct expansion of a refrigerant[Term 3] (Fig. 3), so as to simultaneously satisfy the compactness and high ice-generation rate and to produce optimized slurry ice continuously and stably. Stainless steel 316, to which existing manufacturing technologies can be applied, was selected as the heat exchanger material, considering not only its thermal conductivity but also its strength, corrosion resistance, machinability, and cost. In the double-tube heat exchanger, which satisfies specification (2), ice formation likely proceeds via ice nucleation on the inner wall surface of the inner tube, followed by ice growth. If the generator is equipped with a scraper, which rotates in the inner pipe and thus scrapes the ice from the wall surface before it grows and strongly adheres to the surface, slurry ice including fine ice particles that satisfy specification (1) can be produced. The shape of the scraper was designed so that the force required to scrape the ice would be minimized. The scraper also has a function to thin the thermal boundary layer inside the inner tube, so that the temperature gradient in the radial direction would decrease. This change in temperature distribution would help reduce the force required to scrape the ice by changing the ice growth pattern. In addition, if the ice layer on the wall is always kept thin by the scraper, the thermal resistance due to the ice layer could be reduced, and thus help satisfy specification (3).

The separation of ice from the solid wall is classified into two patterns (Fig. 4): (1) peeling off ice completely from the wall surface (perfect separation), and (2) breaking portions of ice off while leaving part of the ice on the wall surface (partial separation). The force required to scrape ice from the wall surface is determined by the lower force of the two patterns, either that for the perfect or partial separation. Which force

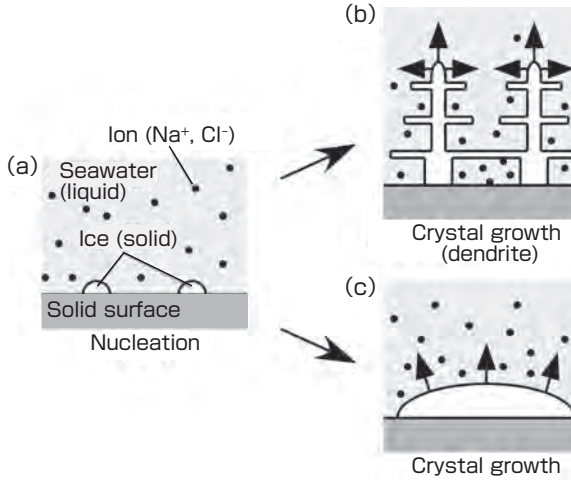


Fig. 2 Schematic of ice generation from seawater. Nucleation and growth of ice on a cooling solid surface.

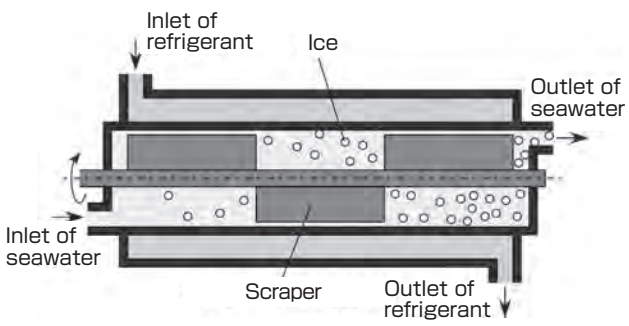


Fig. 3 Double-tube heat exchanger (generator) producing slurry ice from sea water

dominates depends on the operational conditions of the icemaker. The force required for the perfect separation can be effectively decreased by increasing the temperature of the wall surface[15] or by reducing the contact area between the wall and ice. However, increasing the wall surface temperature is undesirable because the higher the temperature of the wall surface, the lower the ice generation rate. Therefore, to decrease the force required for perfect separation, it is preferable to reduce the contact area between the wall and ice. The contact area can be effectively reduced by inhibiting ice growth along the wall surface by reducing the points of ice nucleation on the wall surface.

In contrast, the force required for partial separation can be decreased by making the ice porous, and therefore dendritic growth pattern is desirable, which is often found when the rate-determining process for ice growth is thermal diffusion or mass diffusion. As the ice growth rate is increased by decreasing the evaporation temperature of the refrigerant, the diffusion of NaCl can no longer catch up with the ice growth rate, and finally the porous structure of ice including unfrozen seawater at high NaCl concentration would be formed, which decreases the force required for partial separation.

According to the above discussion, in either perfect or partial separation, the force required to scrape ice from the wall surface can be decreased even when the evaporation temperature of the refrigerant is low, while maintaining a high ice-generation rate. Filtration of seawater to remove small insoluble particles, which induce heterogeneous ice nucleation on their surfaces, is also an efficient technique

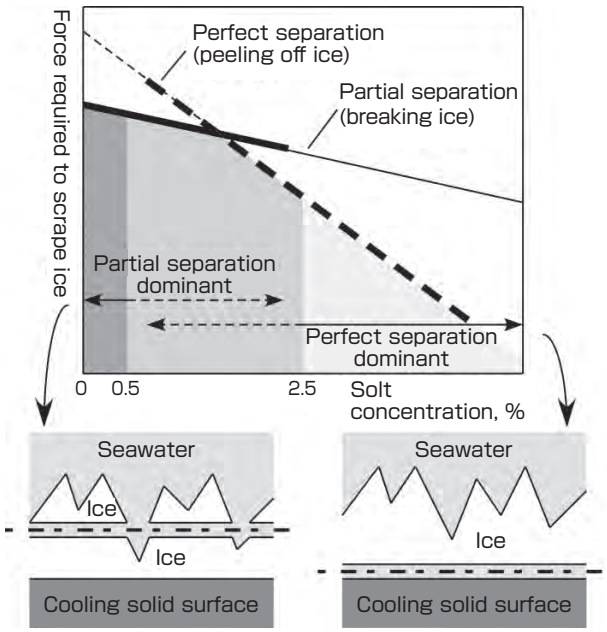


Fig. 4 Schematic of force required to separate ice from a solid surface for perfect separation and partial separation, as a function of salt concentration

to decrease the force required for ice separation. After separation of ice from the wall surface, ice particles gradually increase in size in the generator due to sintering or Ostwald ripening. However, the ice particle size can be kept almost as small as the initial size by controlling the residence time in the generator.

Based on the above design principles, we developed a prototype icemaker. Its operating conditions were then optimized through preliminary tests under various operating conditions. In general, separating ice from solid walls at temperatures below −15 °C is difficult due to the strong adhesion force. However, a generator in which ice can be continuously scraped even at such low temperatures was developed here by optimizing the operating conditions of the prototype. This generator was successfully installed in the prototype icemaker. A testing device simulating large 3-D ship motions such as pitching, rolling and yawing was developed for demonstration of the prototype icemaker. In addition, to address transport of slurry ice between fish holds in a fishing vessel, the technique for stirring slurry ice in a fish hold was optimized to prevent phase separation between the ice dominant phase and seawater phase. This knowledge on stirring was also useful for designing the stirring blades of storage tanks for slurry ice, and such stirring is required for icemakers installed on land. The prototype icemaker also allowed us to easily test oversea transport of raw fish and to examine the freshness of raw fish by scientific analyses. All these tests provided insights into the optimum conditions for maintaining freshness of raw fish and into the optimum shape and size of ice particles, depending on the species of fish. Recently, by making use of these data, we developed an icemaker that produces slurry ice that can efficiently maintain the freshness of raw fish.

Because knowledge on the technologies of control, ice making, and maintaining freshness has been accumulated efficiently by the cooperation of the three collaborating institutions through this research project, it took relatively a short period from the start of the project to the commercialization of the icemaker. After commercialization, cooperation remains among the institutions to promote the distribution of this icemaker by improving the icemaker according to various needs of the users.

4 Effect of slurry ice from seawater

The effect of the slurry ice generated by the developed icemaker is described below for maintaining the freshness of fish. A fish will not be damaged by slurry ice due to its softness. Such slurry ice made from seawater is also effective in maintaining the color and musculature of the fish itself because osmotic pressure surrounding the fish in the melting water after having been cooled is near that of the original seawater (Fig. 5). When a fish dies thus breathing stops, and

the supply of oxygen to the necessary part of the cell stops, the reproduction of ATP, which is its energy source, soon stops. While ATP decomposes as follows, the freshness of the fish degrades.

$$ATP \rightarrow ADP \rightarrow AMP \rightarrow IMP \rightarrow HxR \rightarrow Hx \quad (1)$$ [Term 2, 4–8]

When the progression of decomposition of ATP with products derived from ATP is known, the degree of freshness can be determined. Although the rate of this decomposition varies according to environmental conditions and to the species of fish, the decomposition process is approximately the same and is irreversible. Therefore, progression of the decomposition process can be used as an index of the change in freshness. In general, an often used indicator of the freshness of fish is the K-value, which is defined as follows as the percentage of the quantity of (HxR+Hx) with respect to the total of all ingredients in the decomposition product including ATP.

$$K\text{-value(mol.\%)} = (HxR+Hx) / (ATP+ADP+AMP+IMP+HxR+Hx) \times 100 \quad (2)$$

Freshness is higher when K is lower. Figure 6 is a representative comparison of the K-value when using slurry ice from seawater with immediate cooling capability and when using ordinary crushed ice (seawater + crushed freshwater ice). The K-value when slurry ice was used was lower, indicating a higher freshness of the fish. But care must be taken to not freeze the fish by "over cooling". Cells of fish bodies are damaged when ice crystals increase by freezing. Crystal growth of ice causes drip (body fluids flow out of the fish body), resulting in degradation of the quality when the fish is defrosted. After a fish dies by rapid cooling, it should be kept at a temperature higher than the freezing point (depending on the species of fish, e.g., dark meat fish:around −1.8 to −1.5 °C, white/light meat fish:around −1.5 to −0.5 °C) to prevent actual freezing of the fish. In practice, this can be done by adjusting the amount of ice for the caught fish or by maintaining a suitable temperature in the tank by heat balance calculation, for example. Moreover, to control the breeding of microbes, the temperature should be about 5 °C or less. To maintain the freshness of the fish after transferring and transporting the fish from the onboard tank into the marketing route on land, the low temperature zone should be the same as described above. A "drained ice" method can be used to realize such an environment. In this method, the seawater is drained from the slurry ice and seawater combination, and the fish is left packed in the resulting lightweight drained ice, and because its salt concentration is low, the ice temperature is higher than the freezing point. Using this method during transfer and transport extends the time of high freshness by maintaining a stable low temperature via latent heat of the ice. The advantage of utilization of the ice produced by the icemaker is its effectiveness in adjusting to onboard conditions and long-distance transport. This new technology will raise the economic value produced in the transport and distribution of marine products, the so called *advanced cold chain system*. However, to optimize this technology, the specific fish species must be taken into account to determine the optimal ice conditions such as desired temperature.

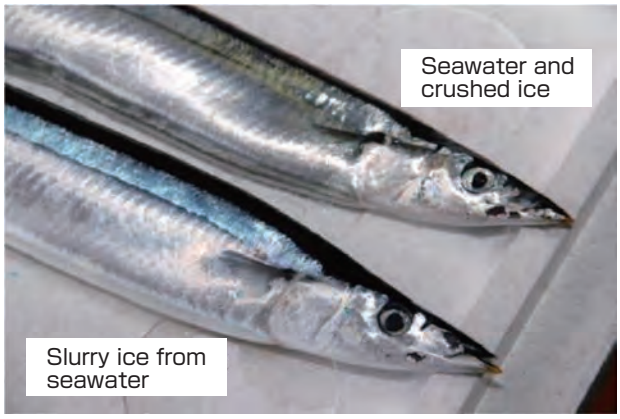


Fig. 5 Comparison of Saury caught during an onboard field test (Blue color on surface indicates high freshness)
Kokonimoatta Sansoken, No.2, p.24–25 (2014)

5 Conclusion

Usage of this compact onboard icemaker and its slurry ice described here is a new freshness management technology and system as well as a health management system. Future study is needed to determine the specifications of the ice to maintain optimum freshness for a specific fish species. "Washoku (traditional Japanese cuisine)" was added to UNESCO's Intangible Cultural Heritage list, in which "sashimi (sliced raw fish)" is part. Freshness management is crucial technology for sashimi. Japan as an advanced country

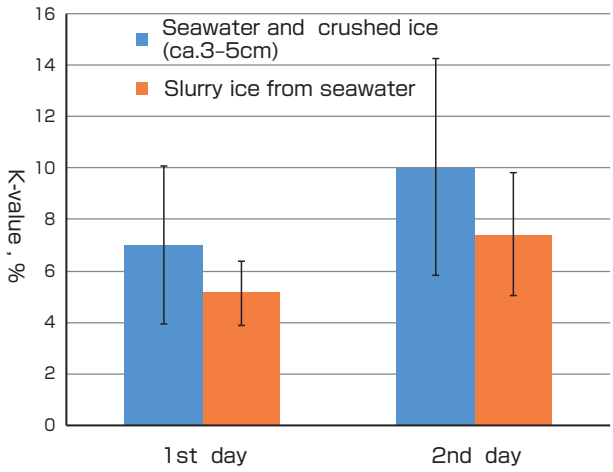


Fig. 6 Comparison of K-values for two different cooling methods
Freshly caught Saury were soaked in seawater with crushed ice (blue shading) or slurry ice from seawater (orange) and kept in a styrene foam container. The quantity of a nucleic acid related ingredient was determined for 6 tail muscles, and then used to calculate the K-value.

familiar with the highest freshness of fish is expected to lead and to globalize such freshness management technology to increase the value of marine products. In this way, utilization of sherbet-like seawater ice as slurry ice would lead to a unique business model that would add value at the highest grade level in the advanced cold chain system.

Acknowledgements

The authors are grateful to Hisakatsu Wajima, a former engineering advisor at Nikko Co., Ltd., for initially leading this project and for further helpful discussions. We also thank Toshimi Shimoyama, a former Nikko employee, who established the basics of the built-in control system of the icemaker, and thank Hitoshi Adachi, a former technical assistance at AIST, who provided advice on commercialization of the icemaker. This work was supported by Fubito Wajima, Kazuo Hirama, Shigeo Chiba and other members of Nikko. via discussions on the daily progress in the research and development.

Footnote) Strictly speaking, the formula used to estimate the freezing-point depression depends on the concentration of sodium chloride. The salinity of the surface layer of seawater ranges between 3.2 and 3.7 wt%, and the surface temperature of seawater is approximately 35 °C near the equator, −2 °C in the Arctic Ocean, and around 15 to 20 °C around Japan.

Terminologies

Term 1. HACCP: Hazard Analysis Critical Control Point. Basics of the hygiene management system recommended internationally, e.g., an inspection system required for import into the European Union and the United States.

Term 2. ATP: Adenosine triphosphate. Comprised of an adenine ring, a ribose sugar, and three phosphate groups, a nucleoside triphosphate, and coenzyme used as an energy carrier in the cells. When ATP breaks into ADP and phosphate, the breakdown of the linkage liberates energy used for functions such as muscular contraction.

Term 3. Direct expansion of refrigerant: Using a refrigerant vapour expansion/compression cycle to directly cool an item. The evaporator is located so that it is in contact with the space to be refrigerated. When the refrigerant inside this space expands (i.e., the double tube as an evaporator), it cools the space by absorbing heat from it.

Term 4. ADP: Adenosine diphosphate. A nucleotide with one less phosphoric acid than ATP.

Term 5. AMP: Adenosine monophosphate. A nucleotide with one phosphoric acid.

Term 6. IMP: Inosine monophosphate. A nucleoside monophosphate deaminated from a base of AMP, and is widely used as a flavor enhancer.

Term 7. HxR: A purine nucleoside that has hypoxanthine linked by the N9 nitrogen to the C1 carbon of ribose.

Term 8. Hx: Hypoxanthine. A purine derivative and deaminated form of adenine. It's a nucleic acid base portion of IMP and HxR, and has a bitter taste.

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Discussions with Reviewers

1 Overall

Comment (Hiroo Matsuda, AIST, Tohoku)

This article describes the development process of a new icemaker and the freshness maintenance of marine products using the seawater ice produced by this icemaker. I acknowledge that this paper is reasonably significant as a *Synthesiology* article as it seeks quality improvement of marine products by integrating several elemental technologies.

Comment (Noboru Yumoto, National Cerebral and Cardiovascular Center)

In this article, the development process of an icemaker that can supply ice stably at low cost and can be installed on a fishing boat is described with attention to the freshness maintenance of the fish using slurry ice. I judge that this paper is suitable as a *Synthesiology* article because it describes cooperative work based on a clear scenario in the industry-academia-government collaboration with each institution having their respective specialty field of mechatronics technology, ice-making technology and freshness evaluation technology to solve difficult problems in past technology development.

2 On the logic constitution and its refinement

Comment (Hiroo Matsuda)

How was the study on each elemental technology conducted? The description of the process is insufficient. There is either simply expressions quoted or just data given from previous studies. Expressions are also insufficient. To clearly describe the development process, I think that you should show the points of improvement (using figures) in the newly developed icemaker compared with the icemakers of previous technology.

Comment (Noboru Yumoto)

In the *Synthesiology* journal, the originality of the author about a scenario and the element constitution (selection and integration) are requirements of an article. Please clarify how you composed elements based on the scenario in Fig. 1 to develop a desired icemaker capable of producing slurry ice.

Answer (Hiroshi Nagaishi)

I reviewed the overall composition again and added a description of the problem and its solution. The description is now divided into background development and development underway of both the embedded software and prototype icemaker up to the point of commercialization.

The manuscript now includes description of the steps of the development process in the following order: (1) development of the control system, such as integration software, (2) design of a prototype icemaker, and (3) demonstration tests of the icemaker. In addition, information about current icemakers has been added to Table 1. Note, however, details and know-how are not disclosed because of a request from the company that carried out the collaboration work.

3 About a new freshness index

Comment (Hiroo Matsuda)

A new freshness index is suggested in this first article, but there seems to be no attempt at standardization. The K-value is utilized in the manual of maintenance freshness issued by many local governments to obtain consensus of those involved. For this proposed new index to obtain consensus, it is necessary to open the index to the public and thus enable those involved to compare its practicality.

Comment (Noboru Yumoto)

A new freshness index is suggested in the first article. I think that the validity of this index should be evaluated in a specialized journal. The *Synthesiology* journal is intended to indicate what to do to utilize an outcome of research and development in society. The validity of this new suggested index cannot be evaluated in this journal. But if standardization of the freshness management technique is achieved, the process can become a subject of this journal.

Answer (Hiroshi Nagaishi)

The freshness index described in the first manuscript is a new suggestion. In this revised manuscript, reference to this freshness index has been significantly reduced. I think that this freshness index itself cannot be patented. With the cooperation of the Intellectual Property and Standardization Promotion Division in AIST, however, I am examining a technique in the cold chain using this index.

4 Improvement of previous technology

Comment (Hiroo Matsuda)

I can understand the need to conceal the know-how. But what points were improved based on a machine of a leading company?

Answer (Hiroshi Nagaishi)

The machine developed here is based on a machine developed in advance. Although the ice-making method and the constitution of the machine are similar for both machines, the machine developed here is compactly made to fit a typical small Japanese vessel. The four main improvements are the (1) cooling surface (inner wall) of the generator, (2) number of fins of the scraper, (3) position and flexibility of the scraper, and (4) control of the ice generation conditions in the generator. As noted in the previous answer to comment (2) above, details of these improvements are not disclosed because of a request from the company. In the revised manuscript, description of the design policy has been added.

5 On freshness of fish

Comment (Hiroo Matsuda)

Are there no experimental results on freshness that the authors performed other than the K-value comparison for Pacific saury?

Answer (Hiroshi Nagaishi)

Numerous experiments and results for evaluation of freshness of fish were obtained by HITEC (by T. Yoshioka).

Here, because the effect of ice generated by the developed icemaker is considered, I believe that the K-value comparison is sufficient as an example.

Standardization of dimethyl ether (DME) fuel specifications

Mitsuharu OGUMA

[Translation from *Synthesiology*, Vol.10, No.1, p.11–23 (2017)]

Legislation and standardization are necessary and important for fuel quality control to ensure safety, security, and stability with regard to the commercialization and trading of new fuels. The author began R&D of dimethyl ether (DME) fuel utilization technology in 2001. This work involved basic research on fuel spray and combustion, applied research on the development of test vehicles, and field tests of these applications. In addition, work on standardizing DME fuel specifications commenced in 2007. In 2015, five ISO standards were published. In this paper, the standardization of DME fuel is presented, which includes a way to define limits on impurities, and the results of round-robin-tests for deterioration by impurities from the users' viewpoint.

Keywords : Dimethyl ether, DME, fuel specification, standardization, impurity

1 Introduction

Internal combustion engines are to remain the powertrain of trucks and buses for a while, as it is impossible under the present circumstances to convert them to electric vehicles. Although the rise in oil prices has stabilized, energy security is an urgent issue, and we must continue to pursue resource diversification including unused resources while keeping in mind the effect of greenhouse gas emissions.

Dimethyl ether (DME, chemical equation CH_3-O-CH_3) is a clean fuel that emits almost no particulate matter (PM) during combustion since it contains oxygen without a carbon-carbon bond. Similar to liquefied petroleum gas (LPG), this gas becomes liquid at pressurization of about 6.1 kgf/cm^2, and the cetane number is equal or higher than that of diesel fuel. When used as fuel for diesel engines, it requires no PM countermeasures, and therefore, the nitrogen oxide (NOx) reduction measure can be applied effectively by reducing the combustion temperature. Hence, it can clear the strict emission regulations without employing the advanced emission reducing catalyst system. If the synthetic gas (CO, H_2) can be obtained, it will not be necessary to rely on specific raw materials, and it can be manufactured using various raw materials including coal, oil sand, natural gas, shale gas, biomass, and others. Other than as fuel for diesel engines, it is usable as a hydrogen carrier in addition to household uses in boilers and gas turbines as alternative to LPG and city gas. The main catch copy for DME is that it is a multi-source and multi-use fuel. Currently, DME is manufactured using coal and natural gas as raw materials, but if the technology is established for manufacturing the fuel via synthetic gas from woody biomass using lumber from thinning and black liquor from paper mills and if such technology becomes economically feasible, the potential for greenhouse gas reduction in well-to-tank (from the excavation of primary energy to the filling of fuel tanks) is large, and its expectation is high as the next-generation biodiesel fuel.

In introducing the new fuel to the market, standardization of fuel quality is mandatory. As the international distribution of DME fuel including household use became highly conceivable, from 2007, DME became a subject in SC4 and SC5 of the ISO/TC28 (the technical committees for LNG and LPG). For the SC5 that discusses the sampling and measurement methods for international distribution, Japan was the secretariat (secretary: Nippon Kaiji Kentei Kyokai). For the SC4 in charge of DME quality, the secretariat was France (secretary TOTAL). Under the French secretariat, the DME quality standard was organized jointly with Japan, which was the only country in 2007 that had already defined the DME quality as industrial and power generation fuel (TS K0011, published November 2005). Japan actively dispatched experts to the ISO/TC28/SC4/WG13 [Classification and specifications of commercial dimethyl ether (DME)], WG14 [Joint project with TC28 on "Test methods for dimethyl ether (DME)"], SC5/WG3 (Procedures for measurement and calculation of refrigerated fluids), and WG4 (Sampling of refrigerated fluids). The author participated as an expert in these working groups. In July 2011, the author was appointed as convener of SC4/WG13 following the retirement of the French convener.

The discussion of DME fuel quality at the WG13 started with at which point the quality should be defined. Figure 1 shows the image of manufacturing, distribution, and use in various machines for the DME fuel. Finally, it was determined at the WG13 that the fuel quality should be defined for the base

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fuel immediately before shipment to the end user as fuel for household, industrial, and automobile use, after being shipped from the manufacturing plant and transported by tankers to primary and secondary stations. It is necessary to add lubricity improvers (LI) to automobile fuel, and in some countries, the addition of an odorant is required to enable detection of fuel gas leakage in household use. Since there are variations in country policies concerning additives, it was excluded from the definition of DME fuel quality in the WG13.

In this paper, we present the effects of impurities, the definition of contamination limit, the round-robin result of impurity analysis method, and the verification test data, that were studied from the standpoint of the fuel utilization system in the standardization of DME fuel quality.

2 Investigation of impurity contamination limit in DME fuel quality

The definition of fuel quality depends largely on compromises, including economic feasibility, of how the manufacturing side can make fuel of certain quality and what the user side demands in fuel quality for use in its system. Although DME is a multi-use fuel, when considering how much inclusion of contamination in the fuel can be tolerated from the standpoint of the utilization systems, it is necessary to correspond to the most sensitive utilization system in which the fuel will be used. Therefore, upon participating in the discussion of impurity contamination limit for DME fuel in the WG13 as an expert, the author started the experimental evaluation of the effect of impurity contamination when DME was used as fuel for diesel engines.

Figure 2 shows the points that were investigated when defining the DME fuel quality used as fuel for automobile

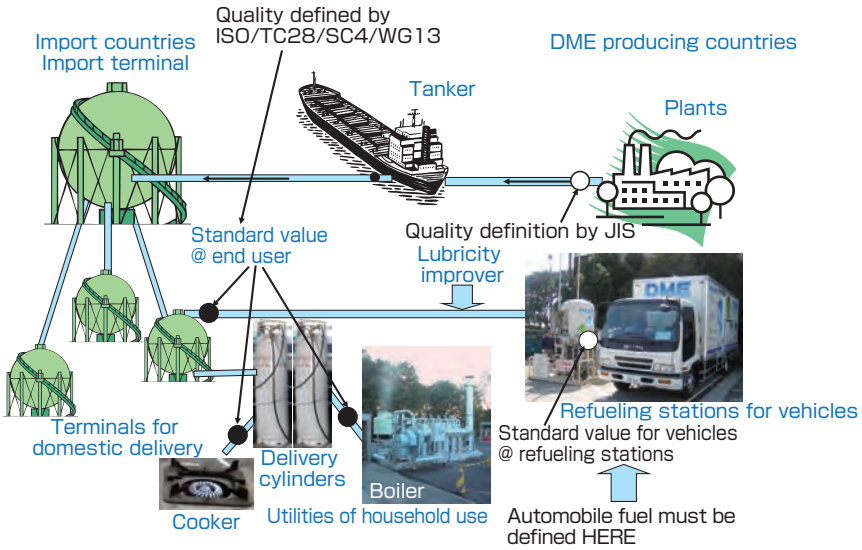

Fig. 1 The point at which DME fuel quality is defined

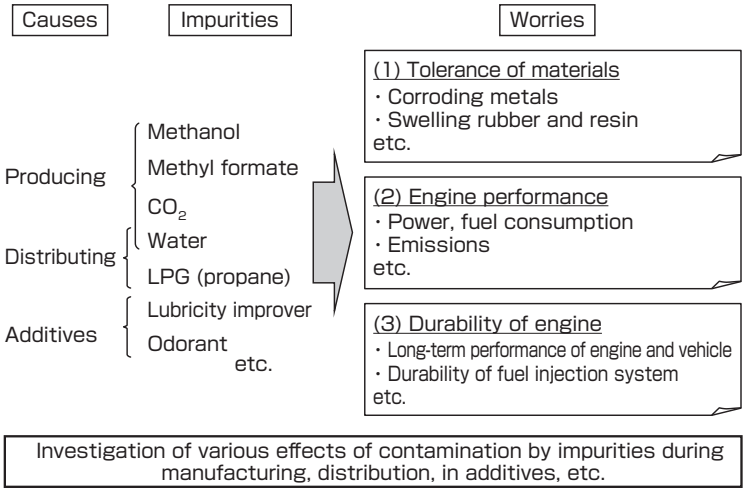

Fig. 2 Substances that may become contaminants in automobile DME fuel standardization and the concerns caused by such substances

Table 1. Test pieces for immersion test

Test materials		Test piece size	Parts*
Rubber	Modified HNBR	- Dumbbells #3 (JIS K 6251) - O-ring (P-12)	IP FT
	FFKM	- Dumbbells #3 (JIS K 6251) - O-ring (P-6)	
Metals	SG steel plate	15x15x3.2 mm with ϕ3.5 hole	FT
	Copper (C1100)	15x15x2.0 mm with ϕ3.5 hole	IP
	Brass (C3604)	15x15x2.0 mm with ϕ3.5 hole	IP
	Injector nozzle needle	-	EG
	Injector nozzle body	-	EG

*IP: Injection pump, FT: Fuel tank, EG: Engine

diesel engines. The effects that were investigated for the additives and others that could be used in automobiles and the impurities that might be introduced in the fuel manufacturing or distribution processes include the following: (1) tolerance of materials, (2) engine performance, and (3) durability of the engine systems. The verification test conducted for each points and the outline of results will be explained by presenting some data.

2.1 Effects of impurities and additives on the tolerance of materials [1]

The evaluation of effects of the impurities on the tolerance of device materials was conducted by immersion tests, where the actual materials were immersed in the DME fuel and then evaluated. The rubber and metal materials used as subjects were selected from the ones actually used in DME vehicles (Table 1).

The immersion conditions are shown in Table 2. The test pieces were immersed in pressure-tight containers, left for 1,000 h in a 80 ºC condition, and the conditions of test pieces were checked. To observe the progression, conditions at 72 h, 250 h, and 500 h were also checked. The base test fuel was DME (purity 99.9 % or higher) that is commercially distributed as a chemical product (for propellant use), and this was mixed with impurities discussed in ISO at certain mass ratio and considered as fuel DME. To obtain lubricity essential for automobile fuel, in one sample, 100 ppm of commercially available fatty acid based lubricity improver (LI) for low-sulfur diesel fuel was added, and in another sample, an excessive amount was added to raise the acid number to 0.13 mgKOH/g for the whole fuel, or until the quality standard of diesel oil mixed with 5 % biodiesel fuel (diesel fuel defined by laws concerning quality control of gasoline and other oils) was surpassed. By comparing these two, the effects of impurities and fatty acid based additives

Table 2. Test condition

Temperature [ºC]	80
Tset duration [hr.]	70, 250, 500, 1000
Test fuels	- Pure DME - Fuel DME* - Fuel DME with LI** 100 ppm - Fuel DME with LI 700 ppm

*Fuel DME: pure DME
+500 ppm of methanol
+100 ppm of water
+1.0 % of propane
+500 ppm of formic acid
+around 2 ppm of sulfur
**LI: Fatty acid based lubricity improver

Table 3. Evaluation items

Test materials	Evaluation items
Rubber	- Changing ration of tensile strength, elongation, hardness, volume and weight - Figure - Compression set (for O-ring only)
Metals	Weight changing ratio, figure

were checked. After immersion at certain time intervals, measurements and observations were done for the points shown in Table 3.

(1) Effect on rubber materials

Tetrafluoroetylene-Perfluoroalkylvinylether fluoro-rubber (FFKM) and improved hydrogenated nitrile rubber (HNBR) are rubber materials used in fuel tanks and injection pumps. According to the immersion test using these materials, there were hardly any differences between pure DME and fuel DME (no Fig.). The effect of DME that swelled the rubber material was strong, and it is thought that the impurities would have almost no effect on the rubber material with anti-DME property. Also, the effect of LI was not seen. In comparison of FFKM and improved HNBR, while there were differences in mechanical properties between the two, this was the difference of rubber material properties against DME, and it was confirmed that there was no effect by impurities or LI.

(2) Effect on metal materials

From the results of the immersion tests for metal materials used in fuel tanks, there was no conspicuous change in appearance in any condition for SG steel plates, injector nozzle needles, and the body. All parts maintained good conditions (no Fig.).

On the other hand, discoloration due to oxidation reaction was seen (Figs. 3 and 4) for copper (C1100) and brass (C3604) that are parts for the injection pump. In copper C1100, the test piece lost luster and became slightly discolored in pure DME, and nearly black discoloration occurred in fuel DME. With the addition of LI, the black discoloration was clear, indicating transformation of the surface. In brass C3604, although not as apparent as C1100, a similar tendency was seen. Since the fatty acid LI was added, it is thought that the acid number of the fuel increased and discoloration occurred by oxidation reaction. There is a possibility of fuel leakage if the corrosion by oxidation progresses in copper that is

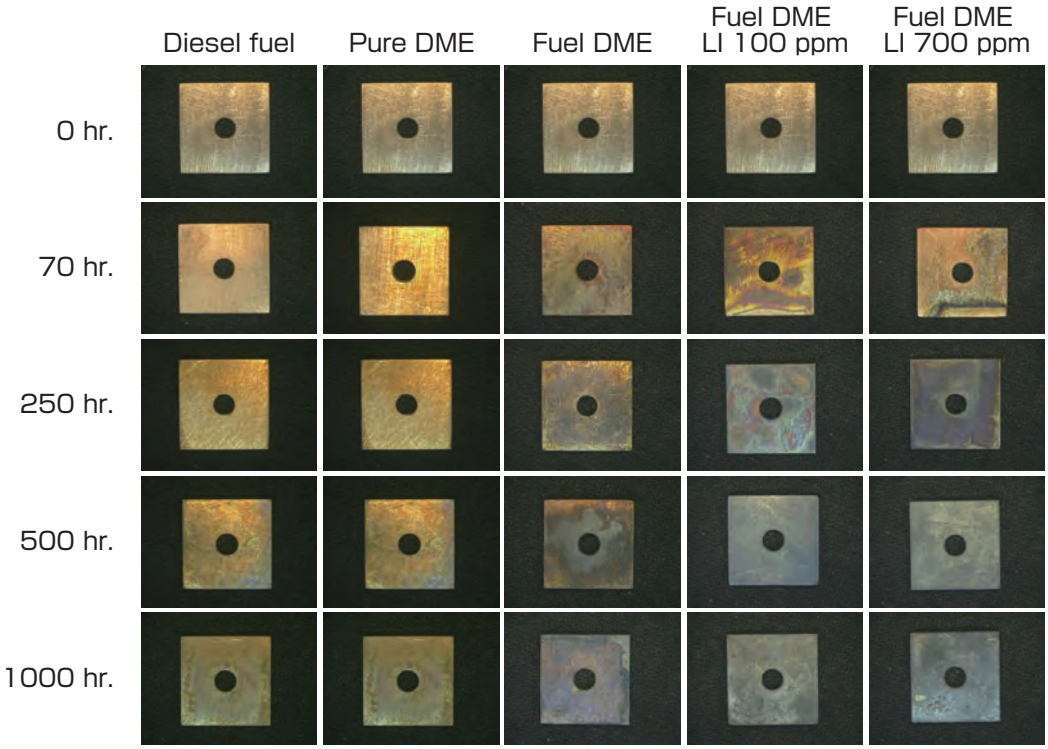

Fig. 3 Appearance of test piece after immersion test (Copper C1100)[1]

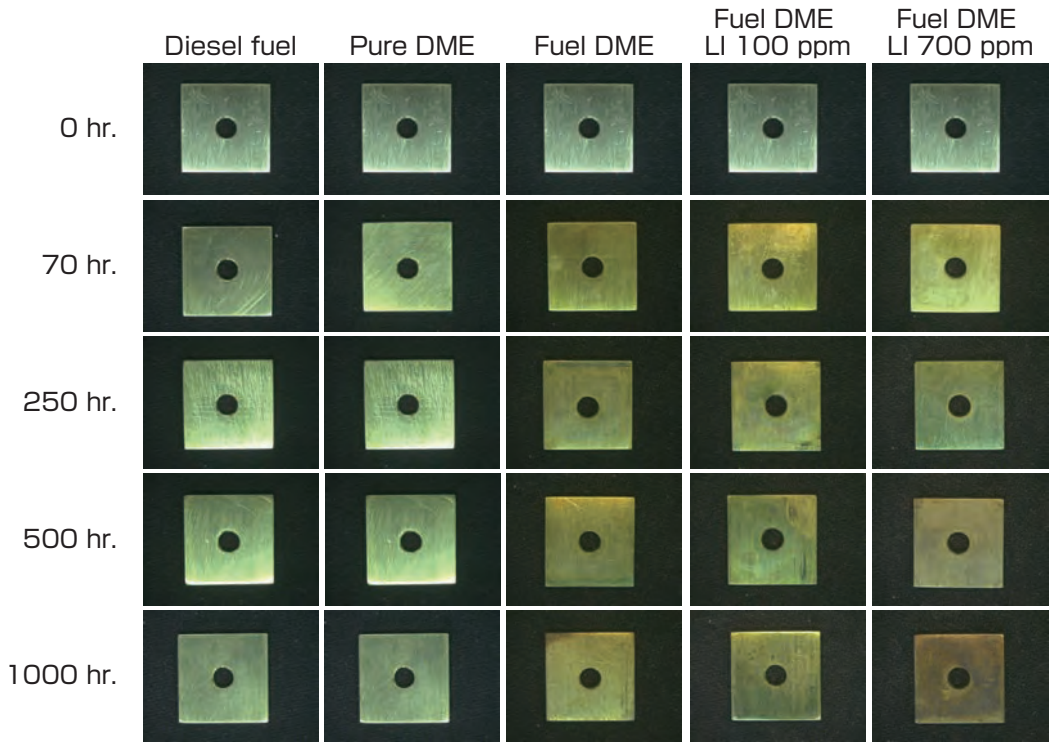

Fig. 4 Appearance of test piece after immersion test (brass C3604)[1]

Table 4. Test fuel (Effect of impurities)

Fuel name	Impurities	Lubricity improver (LI)
Reference DME	–	Fatty acid type 100 ppm
Methanol 5%	Methanol 5 %	Fatty acid type 100 ppm
Propane 5%	Non-odorant propane 5 %	Fatty acid type 100 ppm
FAME 5%	FAME 5 %	–
Water 5%	Water 5 %	Fatty acid type 100 ppm

Table 5. Test fuel (Effect of additives)

Fuel name	Impurities	Additives
Reference DME	–	LI (Fatty acid type 100 ppm)
LI 500	–	LI (Fatty acid type 500 ppm)
Pure fatty acid	–	LI (High concentration fatty acid type 100 ppm)
Odorant	–	Odorant 40 ppm (include sulfur content) LI (Fatty acid type 100 ppm)

used as a sealant for the fuel system, and care must be taken for the excessive addition of fatty acid LI. Also, fuel DME contains water of 100 ppm concentration and methyl formate of 500 ppm concentration, and it is indicated that there is a possibility that formic acid is produced from hydrolysis and increases the fuel acid number. This is thought to be one of the reasons that discoloration was seen only in fuel DME with LI additives. These test results did not show loss of weight in test pieces after the immersion test, but attention must be paid.

2.2 Effects of impurities and additives on engine performance[2][3]

Evaluations were conducted by partial load performance tests using engine dynamometer and JE05 mode tests.

The test fuel for the evaluation of impurity effect is shown in Table 4, and the test fuel for the evaluation of additive effect is shown in Table 5. For both tests, the main fuel was DME (purity of 99.9 % or higher) that is currently commercially distributed as a chemical product (for propellant use). The mixture of DME with about 100 ppm of commercially available fatty acid based LI for low-sulfur diesel fuel was used as the reference DME. The concentration of each impurity is the mass ratio of the total amount.

To investigate the effect of impurities, 5 % of the following impurities were added to the test fuel with 100 ppm of LI: methanol that may remain as residue in the DME manufacture process made by methanol dehydration; deodorized propane (no odorant additive) that may be introduced since LPG facilities may be used until the dedicated DME distribution network becomes available; water that may contaminate or be present as residue in the manufacturing and distribution processes; and fatty acid methylester (FAME, also widely called biodiesel fuel) that is reported to have functions as LI.[4] However, for FAME 5%, since there was data that lubricity becomes equivalent to diesel fuel by adding 3,000 ppm of FAME to DME,[5] no LI was added to FAME. Also, for FAME, commercially available mixed methyester was used as the model FAME. The possibility of water contamination in the market is most likely to occur as contamination by absorbed moisture in the connecting hose when the fuel is transferred between containers. Also, moisture adhesion may occur due to humidity in the filling port of vehicles and at the filling stations. DME fuel samples were taken from installed fuel tanks of medium duty DME trucks belonging to AIST and others, with which the field tests were conducted from 2004 to 2007. Moisture was measured in the fuel samples, and there were cases in which 177 ppm of water contamination was found.

To investigate the effect of additives, the following samples were evaluated: fuel with 500 ppm additive concentration of fatty acid based LI (Fuel name: LI500) used in reference DME (Fuel name: Reference DME); fuel that uses only the main ingredient fatty acid as LI (Fuel name: Pure fatty acid); and assuming that odorants are introduced as in LPG and city gas when it is used widely as fuel, fuel with 40 ppm of an LPG odorant (Fuel name: Odorant). The same LI as the Reference DME was also added to the fuel with an odorant additive.

The effects of the impurities in DME fuel and the additives to DME fuel on the engine performance and emission property were evaluated by engine tests. The tendencies are summarized in Fig. 5. The items shown in yellow and pink in the table indicate caution levels, and pink shows a higher degree of caution than yellow. The results showed that

caution was required for methanol during no-load operation where the active temperature of oxidizing catalysts had not been reached, for emissions from DME containing 5 % propane or water, and for PM number concentration by DME containing 5 % propane and 5 % FAME.

However, although the tendency was as shown in Fig. 5, it was confirmed that "the effect on the emission gas performance test results by mode operation was not that large even if DME containing 5 % impurities was accidentally used." That is, as factors that can define the purity and impurity contamination limit of DME as a fuel quality standard, rather than the effect on emission performance, there was greater effect on tolerance of engine part materials that came in contact with the DME fuel, such as fuel supply systems and fuel injection systems, as well as on the durability of the engine system and the vehicle system. Therefore, the contamination limit of impurities should be determined based on these factors.

2.3 Effects of impurities and additives on lubricity

The evaluation of the effect of impurities and additives in fuel on the durability of engine systems was substituted by the evaluation of fuel lubricity. For the lubricity evaluation of DME, multi-pressure/temperature high-frequency reciprocating rig (MPT-HFRR) adapted to liquefied gas was used.[6] This device achieved the same testing principle as the conventional HFRR device in a hermetically sealed container (Fig. 6). Table 6 shows the comparison of specs with the conventional HFRR device. The testing conditions were the same as the conditions for diesel fuel set by the Japan Petroleum Institute (JPI) standard,[7] and vapor pressure of DME at test temperature (60 ºC) was applied only to atmospheric pressure. The data management method used was that of the author *et al.*[8] in which the additional number of data was determined by the deviation from four measurements.

Please refer to a published report[8] for the relationship of the wear scar diameter (WS1.4) and the additive concentration of LI for DME with poor self-lubricity, the effect of water contamination on the wear scar diameter, and the effect of methanol contamination on the scar diameter. From these results, the following was confirmed: by adding about 100 ppm of commercially available fatty acid based LI for low-sulfur diesel fuel, the same wear scar diameter was obtained as when the commercially available diesel fuel was evaluated with the same device (that is, lubricity equal to diesel fuel was obtained); when the amount of ratio of water was increased with fixed 100 ppm concentration of the same LI, the wear scar diameter started to increase at water concentration of 300 ppm, and the scar diameter of diesel fuel was surpassed at about water concentration of 1,000 ppm (that is, lubricity equal to diesel fuel could not be achieved); and the contamination of methanol had no effect on the wear scar diameter (that is, lubricity).[8] In this paper, we add the evaluation result of the effect of coexisting methanol and water and fuel DME lubricity, and the effect of impurities on fuel lubricity is explained.

Warning index: □ << □

Impurities	Partial load constant mode test						JE05 mode test with oxidation catalyst
	Engine out			Catalyst out			
	Low → <Load> → High High → <λ> → Low			No Low → <Load> → High High → <λ> → Low			
Methanol	Formaldehyde, methanol: slight increase	THC, CO: slight increase		Formaldehyde, methanol: slight increase			
Propane	Formaldehyde: slight increase	THC: slight increase		Formaldehyde: slight increase			PM number concentration: increase
FAME		CO: increase HC: increase					CO, THC: slight increase PM number concentration: increase
Water	Ignition timing: delay THC, CO: increase Formaldehyde, Formic acid: increase			Ignition timing: delay THC, CO: increase Formaldehyde, Formic acid: increase			

Additives	Partial load constant mode test						JE05 mode test with oxidation catalyst
	Engine out			Catalyst out			
	Low → <Load> → High High → <λ> → Low			Low → <Load> → High High → <λ> → Low			
LI 500		THC (CH_4) increase			CH_4 slight increase		
Pure fatty acid							
Odorant			CO, THC increase				

Fig. 5 Summary of engine test for studying the effect of fuel property[1]

Table 6. Specification of MPT-HFRR compared to standard HFRR

	Unit	STD (JPI)	Standard HFRR*	MPT-HFRR
Fuel volume	cm^3	2±0.20	←	500
Stroke	μm	1000±30	20–2000	1000–5000
Frequency	Hz	50	10–200	10–50
Load	N	1.96 $\pm 9.81 \times 10^{-3}$	0.98–9.8	←
Surface area of fuel bath	cm^2	6±1	←	113
Fuel temperature	˚C	60±2	ambient–150	max. 100
Pressure	MPa	ambient	←	max. 10
Test duration	min	75±0.1	←	←

*Manufactured by PCS Instruments

Figure 7 shows the change in the wear scar diameter when methanol of 500 ppm concentration is mixed in pure DME and then water of 5,000 ppm concentration is gradually added. The LI remained constant at 100 ppm for the whole fuel. Figure 8 is the enlargement of the 0–1000 ppm water mixing ratio of Fig. 7. As in the water-only case in the published report, the wear scar diameter started to increase around water of 300 ppm concentration, and the scar diameter surpassed that of diesel fuel at water of 1,000 ppm concentration or higher. From this result, it can be said that when methanol and water coexist, methanol does not enhance or inhibit the effect of increasing scar diameter caused by water contamination, and the decrease of lubricity is determined only by the mixing ratio of water. The mechanism of the decreased lubricity by water is because water has some kind of effect on the breakage of boundary lubricating film that is chemically absorbed to the metal surface. The assumed cause was that the melting point decreased as the boundary lubricating film that metallic soap returned to fatty acid and reached the transformation temperature, or water might have affected the decrease of the transformation temperature.

The white dots in Fig. 7 and 8 are wear scar diameters by fuel DME that was also used in the immersion test. There are two dots in concentration of 100 ppm and 300 ppm, and along with the sample adjusted to 300 ppm by adding water to fuel DME, there are two plots for each water inclusion ratio. While the wear scar diameter increase was not significantly changed by the contamination by other impurities, the effect of impurities was not small compared to others when the water mixture fraction was small.

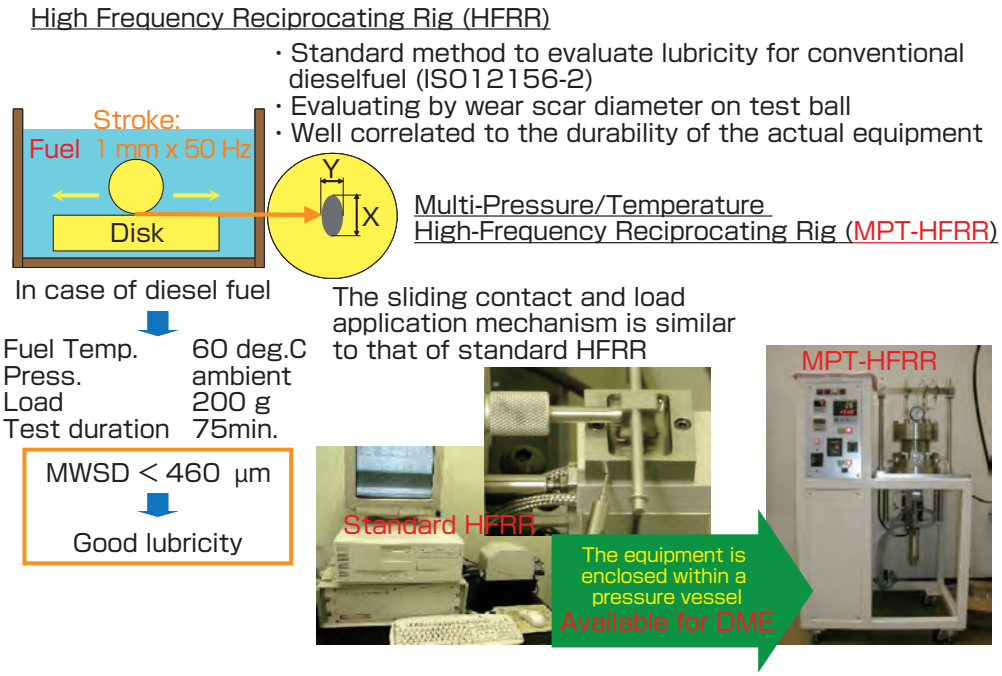

Fig. 6 Lubricity evaluation device and evaluation method

Table 7. Specification of ISO16861: 2015 (DME fuel quality) compared to ASTM D7901-14 (DME fuel quality)

Characteristic	Unit	Limit	ISO16861: 2015	ASTM D7901-14
Purity	mass %	min.	98.5	←
Methanol	mass %	max.	0.050	←
Water	mass %	max.	0.030	←
Hydrocarbons (up to C_4)	mass %	max.	1.00*	–
CO_2	mass %	max.	0.10	–
CO	mass %	max.	0.010	–
Methyl formate	mass %	max.	0.050	report
Ethyl methyl ether	mass %	max.	0.20	–
Residue after evaporation	mass %	max.	0.0070	0.05 (ml/100 ml)
Sulfur	mg/kg	max.	3.0	←
Vapor pressure @ 37.8 °C	kPa	max.	–	758
Corrosion, copper strip @ 37.8 °C		max.	–	No. 1

*In case infrastructure for LPG is diverted or converted in the DME distribution process

2.4 Definition of DME fuel quality

Table 7 shows the comparison of the specifications of ISO16861: 2015 (DME fuel quality) that was published in May 2015 after about seven years of discussion, with ASTMD7901-14 (DME fuel quality) that was published in 2014 before ISO although it was a follower to ISO. For water, 300 ppm was set as the contamination limit value considering the request from manufacturers, based on the data of the aforementioned lubricity evaluation data. For hydrocarbon (C_4 or lower), 1 % contamination was tolerated to enable diversion and conversion of LPG infrastructure at the early stages of the introduction of DME fuel to the market. For this decision, the data used as reference showed that the effect on emission gas performance was small even with 5 % contamination of LPG (represented by propane) according to the engine test. For sulfur component, it should be as close to zero as possible from the standpoint of the utilization systems such as the engine system. However, 3.0 ppm was set as the tolerance value considering the facts that sulfuric acid dehydration was still used in the manufacturing process of some DME manufacturing plants, and the low-sulfur diesel fuel that was the conventional fuel of diesel engines contained slightly less than 5 ppm of sulfur even in advanced nations. For evaporation residue, while there was extremely small possibility that high boiling point ingredients might remain in the DME manufacturing process, for the purpose of capturing contamination, the value of 70 ppm or less was employed. For other impurities, the values that took into consideration the economy of manufacturers were employed, since the effects were extremely small from the standpoint of utilization systems.

ASTM conducted exchange of information several times with the ISO/TC28/SC4/WG13 and WG14, and defined the

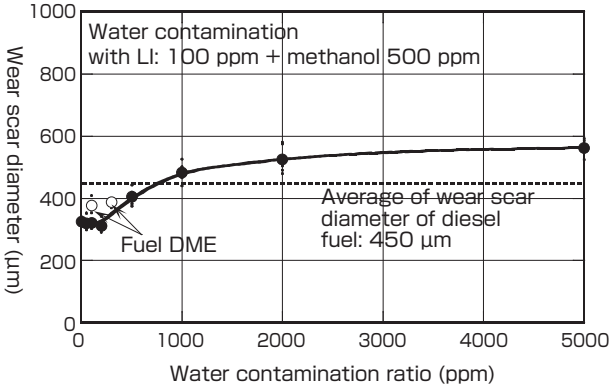

Fig. 7 Effect of water contamination on abrasion scar diameter[1]* (water 0–5,000 ppm, methanol 500 ppm)
*Data added to ref. [1]

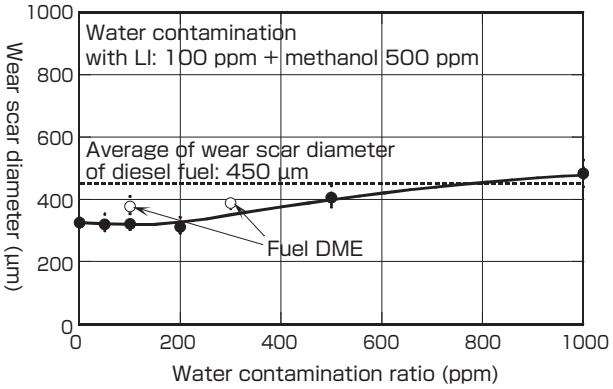

Fig. 8 Effect of water contamination on abrasion scar diameter[1]* (water 0–1,000 ppm, methanol 500 ppm)
*Data added to ref. [1]

respective items based on the discussions at ISO. Although vapor pressure and copper plate corrosion were not defined at ISO, it is thought that ASTM referred to the LPG standards.

3 Investigation of the DME fuel quality analysis method

The analysis method to determine whether the quality was satisfied was necessary when defining and standardizing the DME fuel quality in ISO16861: 2015. Since DME is a liquefied gas fuel, based on the analysis methods of mostly LPG (liquefied petroleum gas) and LNG (liquefied natural gas), the following four analysis methods were drafted, the round-robin tests were conducted, several discussions were held, and the standard methods were issued.

ISO17198: 2014, Dimethyl ether (DME) for fuels–Determination of total sulfur, ultraviolet fluorescence method (2014.11.15)
ISO17786: 2015, Dimethyl ether (DME) for fuels–Determination of evaporation residues–Mass analysis method (2015.5.1)
ISO17197: 2014, Dimethyl ether (DME) for fuels–Determination of water content–Karl Fischer titration method (2014.11.15)
ISO17196: 2014, Dimethyl ether (DME) for fuels–Determination of impurities–Gas chromatographic method (2014.11.15)

For the round-robin tests, in the case of this DME, it was necessary to consider the measurement standard of the analysis subject, that is, it was necessary to conduct precision analysis by preparing several standards for types and concentrations of impurities in the DME within the range of analysis application. However, due to the limitation of samples and time, it was conducted for one measurement standard only. For the round-robin tests, in addition to the seven laboratories in Japan, there was participation by two labs in Korea, and one lab each from Sweden, Canada, and Belgium. Eight labs participated in all analysis items although they differed in capacities to conduct certain analysis items.

The test samples were made by the author's research group by mixing the impurities in pure DME by a gravimetric method, and the labs were asked to conduct analysis with no information given about the values. In this paper, we present the results of the round-robin tests of impurity concentration by gas chromatography through which the issues became most visible.

First, Fig. 9 and Fig. 10 show the analysis results of hydrocarbon (HC) of C_4 or less and methanol, respectively, and most labs showed relatively similar analysis results for the impurity concentration mixed by a gravimetric method. The scatterings of the measurement values within the labs are shown by error bars. The precision analysis of the round-robin tests was conducted by Cochran's tests and Grubb's tests as designated by ISO5725-2, and the repeatability standard deviation and the reproducibility standard deviation were calculated. As a result, for HC of C_4 or less, the repeatability standard deviation was Sr = 0.0134 and the reproducibility standard deviation was SR = 0.0393. However, this accuracy was obtained by a 0.0952 wt.% standard test with the participation of six labs. In this experiment, one 1 % outlier each was found for Cochran and Grubb tests, but these were kept and included in the calculation. The concentration of HC of C_4 or less in the sample made by a gravimetric method was 0.100 wt.%, and this was relatively close to the general average value of 0.0952 wt.% obtained from the analysis result of the round-robin test.

For methanol, the repeatability standard deviation was Sr = 0.0025 and the reproducibility standard deviation was SR = 0.0072. This accuracy was obtained by a 0.0160 wt.% standard test with the participation of six labs. In this experiment, one 5 % outlier each was found for a Cochran's test, but these were kept and included in the calculation.

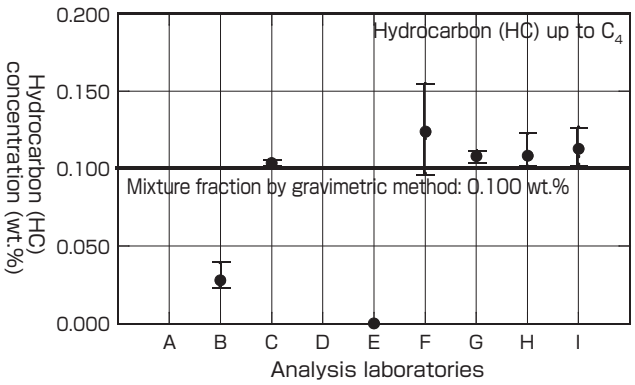

Fig. 9 Analysis result of hydrocarbon (HC) of C_4 or less in the round-robin test[9]*
*Graph based on data of ref. [9]

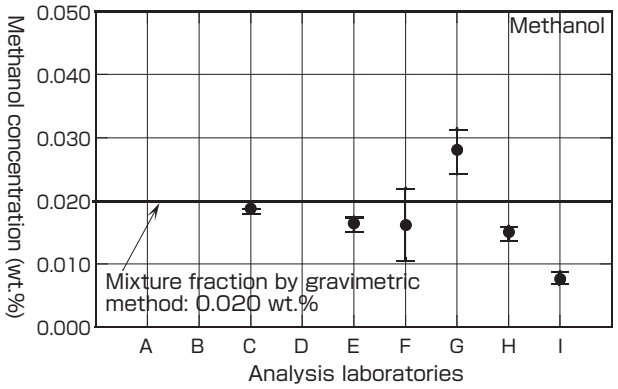

Fig. 10 Analysis result of methanol in the round-robin test[9]*
*Graph based on data of ref. [9]

The concentration of methanol in the sample made by a gravimetric method was 0.020 wt.%, and this was relatively close to the general average value of 0.016 wt.% obtained from the analysis result of the round-robin test. For the HC of C_4 or less and methanol, it is thought that the accuracy will improve through increased skill of the laboratories.

On the other hand, Fig. 11 and Fig. 12 show the analysis results of carbon monoxide (CO) and carbon dioxide (CO_2), and several labs showed highly differing analysis results compared to the impurity concentration mixed by a gravimetric method. Similarly, the scatterings of measurement values within the labs are shown by error bars, the accuracy analysis was conducted by Cochran's tests and Grubb's tests as designated by ISO5725-2, and the repeatability standard deviation and the reproducibility standard deviation were calculated. As a result, for CO, the repeatability standard deviation was Sr = 0.0006 and the reproducibility standard deviation was SR = 0.009. This accuracy was obtained by a 0.0013 wt.% standard test with the participation of five labs. In this experiment, one 1 % outlier each was found for a Cochran's test, but these were kept and included in the calculation. The concentration of CO in the sample made by a gravimetric method was 0.010 wt.%, and this was vastly disparate from the general average value of 0.0013 wt.% obtained from the analysis result of the round-robin test. However, the repeatability standard deviation and the reproducibility standard deviation were relatively small, the reproducibility was good within the lab and among the labs, and the analysis result was about one-tenth the CO concentration made by the gravimetric method.

The CO_2 showed a similar trend as CO, and the repeatability standard deviation was Sr = 0.0018 and the reproducibility standard deviation was SR = 0.0018. This accuracy was obtained by a 0.0064 wt.% standard test with the participation of six labs. In this experiment, one 1 % outlier each was found in the Cochran's test, but these were kept and included in the calculation. The concentration of CO_2 in the sample made by the gravimetric method was 0.010 wt.%, and this was vastly disparate from the general average value of 0.0064 wt.% obtained from the analysis result of the round-robin test, as in CO. However, the repeatability standard deviation and the reproducibility standard deviation were relatively small, and the reproducibility of the analysis result was good within the lab and among the labs.

The analysis of impurities by gas chromatography created so far could not be applied to CO and CO_2. Therefore, to analyze the disparity factors of the analysis results, the solubility of CO and CO_2 in DME was measured. The CO was supplied at various pressures in the container holding pure DME, the containers were shuffled to promote the mixture of DME and CO, the sample was removed from the liquid phase, and the analysis by gas chromatography was conducted. Samples were made similarly for CO_2, and the analysis was conducted. Figure 13 shows the CO solubility in DME for CO partial pressure, and Figure 14 shows the CO_2 solubility in DME regarding CO_2 partial pressure. From these data, it was confirmed that the solubility in DME of CO and CO_2 was determined by partial pressure. It became clear that, in order to accurately analyze the concentration in the samples, CO = 0.010 wt.% and CO_2 = 0.10 wt.%, made by gravimetric methods in this round-robin test by secondary polynomial approximation, it was necessary to apply backpressure of 0.0194 MPa and 0.0825 MPa or higher, respectively, during sample extraction.

As a result, since there is low possibility that CO and CO_2 are introduced in the manufacturing or distribution processes of DME, although there were issues unsolved in the analysis method, we attained issuance of ISO by describing the information of solubility in the appendix.

4 Discussion and issues

After about seven years of discussion, we were able to issue the DME fuel quality and the four types of analysis methods

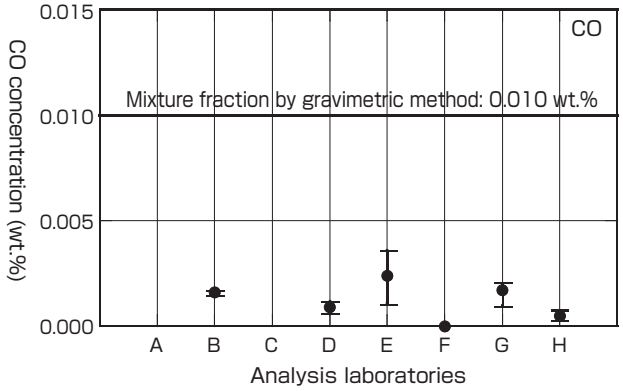

Fig. 11 Analysis result of carbon monoxide (CO) in the round-robin test[9]*

*Graph based on data of ref. [9]

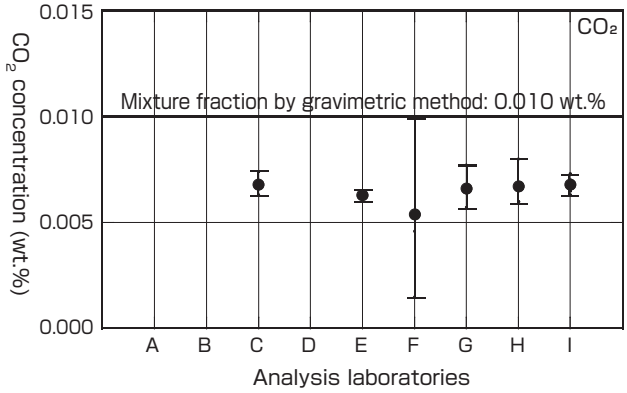

Fig. 12 Analysis result of carbon dioxide (CO_2) in the round-robin test[9]*

*Graph based on data of ref. [9]

as ISO. Here, the points encountered by the author toward the establishment of ISO will be described

(1) Efficacy of measurement data for the determination of water contamination tolerance limit

As mentioned earlier, the determination of impurity contamination limit in the DME fuel quality was made through compromise between the standpoint of manufacturers (please tolerate this much impurities) and the standpoint of users (if this much impurities are present failures may occur in the utilization systems such as engines). Since this argument was evident particularly over the determination of water contamination tolerance limit, we shall present the case here.

For the DME diesel engine that was a utilization system expected to demand the severest fuel quality, the Japanese experts including the author were aware based on the experimental data that the water contamination tended to decrease the fuel lubricity that greatly affected durability, and therefore stated that the water contamination tolerance limit should be up to 100 wt. ppm. On the other hand, the experts of a country that had several operating DME manufacturing plants demanded the tolerance of 300 wt. ppm for the manufacturing technology. This was the demand from a country with the top share in the world DME fuel market, and it was determined that it would not be positive to totally deny the demand considering the formation and expansion of the DME fuel market. Therefore, as there was some room to the level where the lubricity seriously decreased based on the experimental data, ultimately, the water contamination tolerance limit of 300 wt. ppm was accepted.

At the time, there were only three institutions, including the one to which the author belongs, that were able to evaluate the lubricity of DME fuel that was liquefied gas. The three institutions once evaluated the same sample, and from that result, the author had confidence in his and his colleagues' measurement accuracy, and was able to make decisions instantly during the working group meetings based on abundant backup data.

(2) Accumulation of experience in conducting the round-robin tests

The author and his colleagues are researchers and technicians of mechanical engineering. Although we were not planning to participate in the round-robin tests to check the accuracy of the analysis method, we started from the introduction of analysis devices as we were requested to join to secure the necessary number of participating laboratories. For manufacturing the samples by gravimetric methods, we had experience in making fuel for engine tests, and we were able to gain lots of experience along with chemical analyses. This accumulated experience became very valuable in the case where we clarified that the solubility of CO and CO_2 was affecting the analysis accuracy.

(3) Discussion and negotiation

In the discussions in international meetings, it is said that lobbying activities such as consultations and consensus building outside the meeting room are extremely important. While there are pros and cons for such activities, it is true that we witnessed such actions. On the other hand, the persuasiveness of scientific experimental data is tremendous. At the place of discussion of ISO standardization for DME fuel quality and test methods, the intentions of corporations, mainly of Europe, that were aiming to commercialize this fuel were trying to take lead of discussions. They were engaging in negotiations (consensus building led by profit and interest) based on assumptions and hypotheses, as there was lack of data on the effect of impurities on the utilization systems and impurities analysis results. There was concern that the Japanese national interest and Japanese DME industries would be affected, including the chemical manufacturers that manufactured good quality DME fuel as well as automobile manufacturers that manufactured high-performance DME automobiles. I remember that we gradually gained the lead by shifting from "negotiation" to

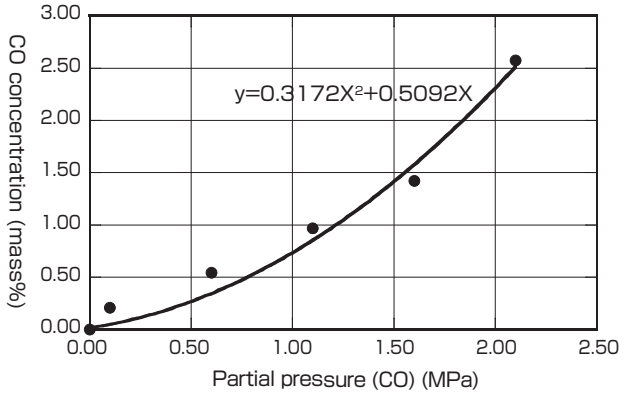

Fig. 13 Effect of CO partial pressure on CO solubility in DME[9]

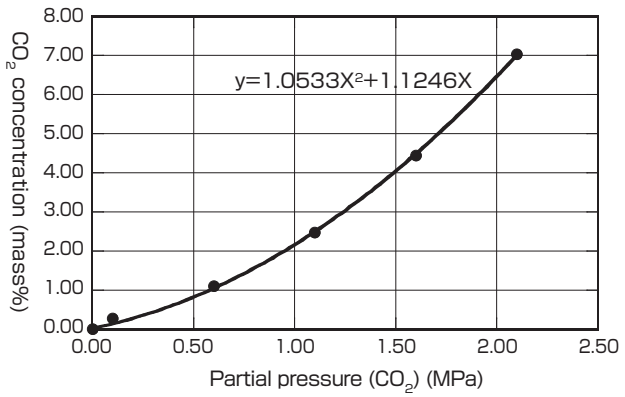

Fig. 14 Effect of CO_2 partial pressure on CO_2 solubility in DME[9]

"discussion" (technological arguments based on data) based on the abundant experimental data and analysis results.

5 Conclusion

For the standardization of DME fuel quality, we summarized the evaluation of the effect of impurities, the definition of the contamination limit, the round-robin result of the impurity analysis method, along with the investigated experimental data that were studied from the standpoint of the fuel utilization systems. The author was given a precious opportunity and was able to gain valuable experience by participating in the stage of international discussion of ISO standardization. In this process, I experienced a tense atmosphere where the national interest might be compromised unless one raised one's voice, and learned that technological argument based on data was convincing even with incomplete English. There, I experienced firsthand the importance of data. The joy in participating in international standardization activities is building networks and interacting with engineers around the world. I would like to spend effort on increasing my skills as a researcher and helping train younger researchers.

Acknowledgements

The MPT-HFRR test device was developed jointly with the Iwatani Corporation. This research includes the results of the following: the "International Standard Joint R&D Project: Standardization for Dimethyl Ether (DME) Fuel," FY 2009 Standardization R&D Commission Fund, Ministry of Economy, Trade and Industry; and the "Standardization R&D: Dimethyl Ether (DME) Fuel Standardization," FY 2010 Strategic International Standardization Promotion Project, New Energy and Industrial Technology Development Organization (NEDO). Guidance for research and standardization activity was provided by Shinichi Goto, Emeritus Researcher, Research Institute for Energy Conservation, Department of Energy and Environment, AIST. Takuro Watanabe, Senior Researcher, Research Institute for Material and Chemical Measurement Gas and Humidity Standards Group, helped us with the DME quality analysis. This standardization was accomplished through the cooperation of members of several groups, such as assistance in experiments by Kazuaki Higurashi, Technical Staff, Engine Combustion and Emission Control Group, Research Institute for Energy Conservation, AIST, and round-robin test analysis by Akiko Ohmuta, former Technical Staff, Combustion and Engine Research Team, (former) Research Center for New Fuels and Vehicle Technology, AIST. I express my gratitude to all people involved.

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of Automotive Engineers of Japan in 2012; and the Award for Contribution to International Standardization Division (Award of the Director-General of the Industrial Science and Technology Policy and Environment Bureau), Industrial Standardization Project Awards in FY 2016.

Discussions with Reviewers

1 Overall

Comment (Haruhiko Obara, AIST)

This paper is a concise summary of the research in which the author engaged for the standardization of DME fuel quality. Particularly, the author received this year's Industrial Standardization Project Awards of the Ministry of Economy, Trade and Industry, and his standardization activities are highly acclaimed both in and outside Japan. This paper also describes the international standardization activities such as the round-robin measurements, and it emphasizes the importance of international standardization activities. I believe it will be relevant to the readers outside of the field.

Comment (Hiroki Yotsumoto, AIST)

I think this paper clearly illustrates the requirements of the DME fuel as well as the efforts and hardships of international standardization.

2 Future of DME

Comment (Hiroki Yotsumoto)

In the introduction, you mention that "if the technology is established for manufacturing the fuel via synthetic gas from woody biomass using lumber from thinning and black liquor from paper mills…." How is this assumption likely to be realized? If you mention synthetic gas, all carbon resources become possible candidate raw materials, don't they? I am asking this question because I feel that the discussion may be lost on how high the feasibility of using black liquor and lumber from thinning is compared to other carbon sources.

Answer (Mitsuharu Oguma)

While the technology of manufacturing synthetic gas (CO, H_2) from black liquor or woody biomass is not that difficult, it is inferior in terms of economy compared to coal and natural gas.

I shall change the expression to the following: "if the technology is established for manufacturing the fuel via synthetic gas from woody biomass using lumber from thinning and black liquor from paper mills and if such technology becomes economically feasible…."

3 Effects of impurities and additives on metal material

Comment (Hiroki Yotsumoto)

In Fig. 3, there is discoloration in copper C1100. What kind of reaction is happening to copper? Also, how about adding the harm there may be to the material?

Answer (Mitsuharu Oguma)

The discoloration is due to oxidation reaction. Copper is used as sealant for fuel systems, and fuel leakage may occur if the corrosion by oxidation progresses. I shall add this to the paper.

4 Effects of impurities and additives on engine performance

Comment (Hiroki Yotsumoto)

It is unclear what you mean by "total emission performance," and I think you should provide an explanation. If it means the amount of environmental pollutants in the emission, I think you need to better explain the relationship with "Caution" in the explanation of Fig. 5. What do you think?

Answer (Mitsuharu Oguma)

I shall correct the applicable text to the following: "The effects of the impurities in DME fuel and the additives to DME fuel on the engine performance and emission property were evaluated by engine tests. The tendencies are summarized in Fig. 5. The items shown in yellow and pink in the table indicate the caution levels, and pink shows a higher degree of caution than yellow." Change is also made to the following: "However, although the tendency was as shown in Fig. 5, it was confirmed that 'the effect on the emission gas performance test results by mode operation was not that large even if DME containing 5 % impurities was accidentally used.'"

5 Discussions and issues in the international standardization process

Comment (Hiroki Yotsumoto)

Don't you think you should explain the specific difference between "negotiation" and "discussion" in the meetings for international standardization?

Answer (Mitsuharu Oguma)

Thank you very much for pointing this out. I shall explain as follows. Negotiation is consensus building led by profit and interest. Discussion is technological argument based on data.

A study on high-density recording with particulate tape media for data storage systems

— On the process of introducing barium-ferrite tape media to the market —

Takeshi HARASAWA* and Hitoshi NOGUCHI

[Translation from *Synthesiology*, Vol.10, No.1, p.24–32 (2017)]

Magnetic tape storage systems are widely used for archive and data backup from their characteristic of low cost and large capacity. Research on magnetic tape targets higher recording densities to meet market needs for continuously increasing storage capacity like in other storage media. Progress on increasing the recording density of magnetic tapes using conventional magnetic metal particles has slowed in the years leading up to 2010. However, progress improved in 2011 with the introduction of tape media using barium-ferrite magnetic particles. In this paper, we describe the process of going from basic research on tape media using barium-ferrite to marketplace introduction.

Keywords : Barium ferrite, particulate tape media, areal recording density, linear tape system, data storage

1 Introduction

At present, the rate of data creation is increasing explosively, because of the advancement of computers, increased communication events through established communication infrastructures, and the improved performance of devices such as sensors. The volume of data generated and duplicated internationally was 0.9, 1.8, and 4.4 ZB (10^{21} bytes) in 2009,[1] 2011,[2] and 2013,[3] respectively. This is an increase of 40 % per year, with this value being expected to reach 44 ZB in 2020. Therefore, continuous increase in recording density for data storage devices is being pursued. Among the various storage systems, magnetic tape continues to be used as a back-up for hard disk drives (HDDs) and for archives requiring long-term data storage,[4] because of the associated low system and bit costs, long lifespan, and energy-saving properties of this technology. As shown in Fig. 1, while the areal recording densities of HDDs, optical disks, and magnetic tape are increasing annually, progress regarding recording density development has recently been on the decline. Further, the rate of improvement of magnetic tape recording density began to decelerate in approximately 2010. This deceleration occurred because a limit was reached regarding the creation of the fine particles essential for achieving high-density with metal-particle (MP) magnetic material, which was the mainstream recording material at that time. Therefore, in 2011, Fujifilm Corporation introduced magnetic tape based on a barium-ferrite (BaFe) magnetic material to the market. The use of BaFe allowed further fine particle formation and succeeded in re-accelerating the rate of recording density enhancement. In this paper, we describe the process from basic research to commercialization for BaFe tape, which is a major technological innovation for the magnetic tape industry.

2 History of magnetic tape

2.1 Use of magnetic tape

Magnetic recording was first introduced in 1898, with the invention of the magnetic recording device by Valdemar Poulsen, a Danish scientist working on audio recording. Later, magnetic recording technology expanded from sound recording to the fields of image and information recording, with the rapid advancement of television, computers, and similar technology.

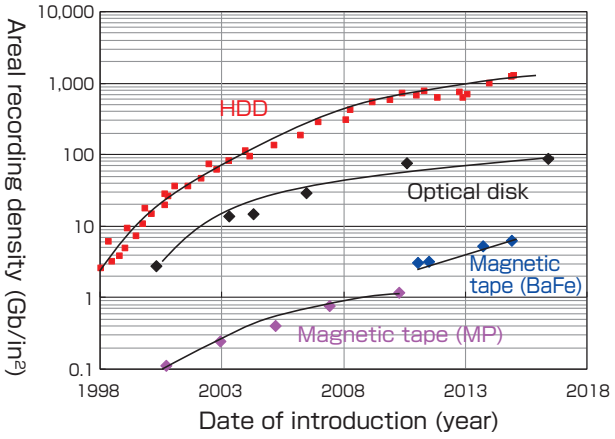

Fig. 1 Transition of the surface recording density of HDD, optical disk, and magnetic tape

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In the audiovisual field, audiocassettes and videotapes associated with technology, such as the video home system (VHS) and the 8-mm video system, were widely used until the 1990s; however, these devices disappeared with the advancement of optical disks such as compact discs (CDs) and digital versatile disks (DVDs). After the 2000s, broadcasting videotapes were replaced by new recording media such as optical disks and HDDs, with videotapes gradually reaching the end of their application lifespan.

On the other hand, in the information recording field, magnetic tapes were employed as memory devices for computers from approximately 1950 onwards. With further development of computers, magnetic tapes, which were characterized by low price and large capacity, were widely employed by data centers, major companies, government agencies, and research institutes. Recently, magnetic tapes have predominantly been employed in the information recording field rather than image recording. Currently, three representative tape systems exist: linear tape-open (LTO), which is most widely used as an open format, and International Business Machines (IBM)'s 3592 and Oracle's T10000, which are closed systems for enterprise use. The recording densities of these three systems are continuously improving. Currently, LTO, 3592, and T10000 have achieved 6,[5] 10,[6] and 8 TB tape cartridges, respectively.[7] All these systems use the BaFe magnetic material.

2.2 Transition of magnetic tape technology

At present, mainstream magnetic tape manufacturing procedures employ a coating method to achieve low cost and mass production. As shown in Fig. 2, a nonmagnetic layer that provides conductivity and surface smoothness is formed on a plastic base film. Then, a magnetic layer for data recording is formed on top of that layer, whereas a backcoat layer is formed on the backside of the base film to provide runability for the tape medium.

High-density recording for magnetic tape was achieved in accordance with the principles of magnetic recording. That is, the following three requirements were satisfied: (1) fine particles of magnetic material; (2) a thin magnetic layer; and (3) surface smoothness of the magnetic layer. Figures 3–5 show the trends for the magnetic-material particle sizes in the magnetic tapes, the magnetic layer thickness, and the average surface roughness *Ra*, which is the surface smoothness index. It is apparent that continuous progress has been made toward finer magnetic particles, thinner magnetic layers, and smoother surfaces. In particular, as is apparent for the magnetic layer thickness trend shown in Fig. 4, the technological innovation that shifted the magnetic tape layer structure from single to multilayer (where the magnetic layer was formed on a nonmagnetic layer) caused the magnetic layer thickness to shift from the order of several microns

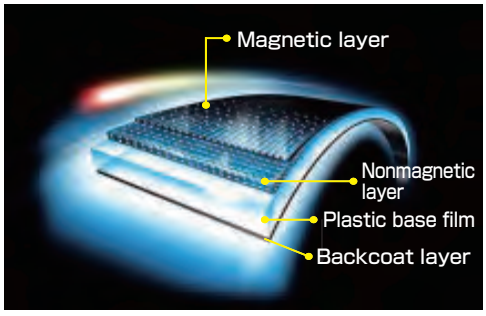


Fig. 2 Structure of the magnetic tape

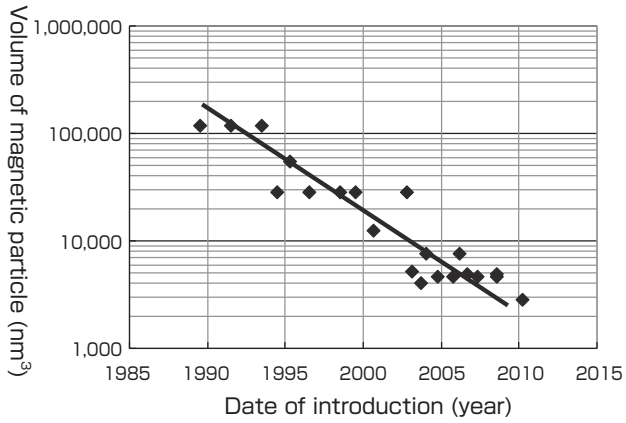


Fig. 3 Transition of the magnetic material volume

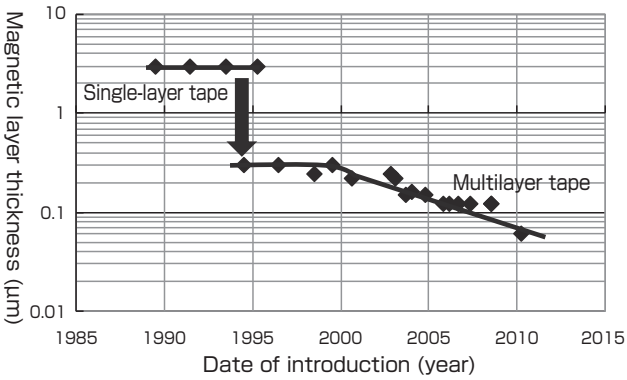


Fig. 4 Transition of the magnetic layer thickness[8][9]

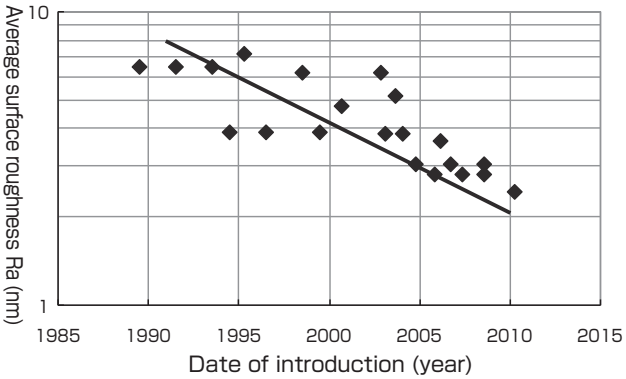
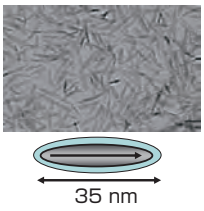
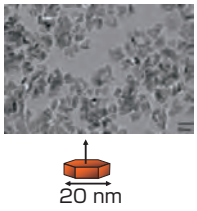


Fig. 5 Transition of the surface smoothness of tape

Table 1. Characteristics of the magnetic material

Magnetic material	MP	BaFe
TEM image and schematic diagram		
Raw material	FeCo, Required passivation layer for oxidation-protection	$BaO(Fe_2O_3)_6$, Stable because of oxide
Origin of magnetization	Shape anisotropy	Crystal anisotropy
Direction of magnetization	Longitudinal direction	Perpendicular direction
Saturation magnetization (Ms)	600–900 emu/cc	250–300 emu/cc

to submicron order; hence, the layer thinness decreased dramatically.[8][9] On the other hand, the fineness of the MPs for the magnetic materials approached a limit in the latter half of the 2000s. This was because it became difficult to maintain sufficient coercivity (a property necessary to maintain recorded signals) as the MPs became finer (Fig. 6). On the other hand, in the newly developed BaFe magnetic material, sufficient coercivity is maintained for fine particles of approximately 1000 nm^3 in size, by controlling the composition and synthesis processes. In the next section, the characteristics and issues of the BaFe magnetic material are described.

3 Characteristics and issues of BaFe magnetic material

3.1 BaFe magnetic material characteristics

The characteristics of MPs and BaFe magnetic materials are compared in Table 1. The MP magnetic material is a needle-shaped metal alloy primarily composed mainly of iron (Fe) and cobalt (Co). To inhibit oxidation, a passivation layer of at least several nanometers in thickness is necessary. This material also has magnetic-shaped anisotropy, where the origin of the magnetization comes from the needle-like shape of particle. To increase the anisotropic energy, a high axial ratio (long-axis length/short-axis length) is necessary. On the other hand, the BaFe magnetic material is an oxide with crystal magnetic anisotropy, where the anisotropic energy is determined by the crystal structure of the particle. Further, the axis of easy magnetization of the BaFe magnetic material is perpendicular to the plate surface. This material has the characteristic of a magnetization component that is perpendicular to the magnetic layer when the tape is formed.

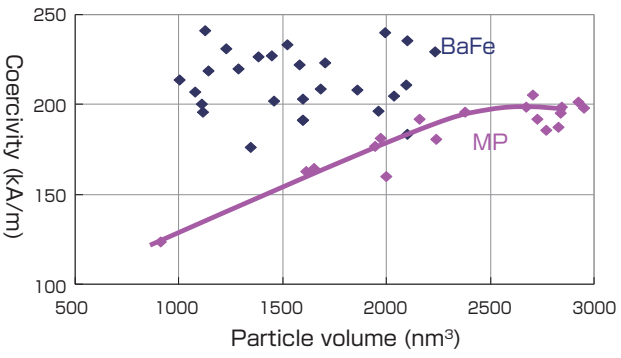


Fig. 6 Particle volume and coercivity of the magnetic material

Based on consideration of these characteristics, we determined that the BaFe magnetic material possesses the following superior properties, concluding that this material was the most likely candidate for a post-MP magnetic material.

(1) Regarding the MP magnetic material, it is difficult to increase the high axial ratio of the particles, passivation layer for oxidation-protection is required, and sufficient coercivity cannot be maintained as the particles become finer. In contrast, the BaFe magnetic material can maintain high coercivity in fine-particle form, because its anisotropic energy is determined by the crystal structure. Further, no passivation layer for oxidation protection is required.

(2) The BaFe magnetic material, which is an oxide, is extremely stable in a high-temperature and high-humidity environment, and has excellent long-term storage properties in tape form (Fig. 7).

(3) As a magnetization component exists perpendicular to the magnetic layer when the tape is formed, application of the BaFe magnetic material to the perpendicular magnetic recording achieved for HDD could be expected.

3.2 Issues related to BaFe magnetic material

As mentioned in the previous subchapter, although the BaFe magnetic material has many characteristics that are advantageous for achieving high density, the following issues required resolution during R&D in order to fully exploit its characteristics.

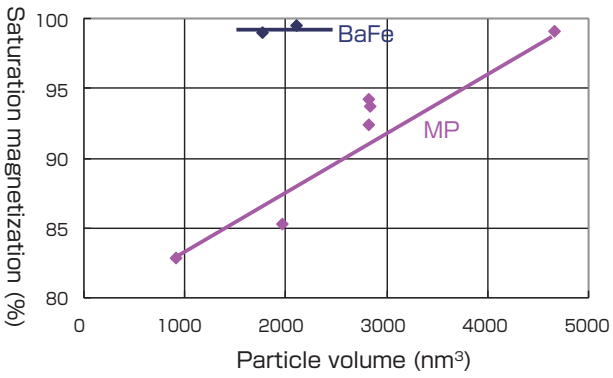


Fig. 7 Change in the saturation magnetization (Ms) for one month storage at 60 °C and 90 % RH (relative humidity)

(1) As the BaFe magnetic material is in fine-particle and plate form, the magnetic materials are likely to aggregate. Thus, it was not possible to create a uniform, smooth, and thin magnetic layer using conventional dispersion or coating technology.
(2) As the saturation magnetization quantity (Ms) of the BaFe magnetic material is small, the reproduced output is lower than that of MP tape; thus, it was necessary to supplement this output with a highly sensitive magnetic head (Fig. 8).
(3) As the BaFe tape has perpendicular magnetization components, the symmetry of the isolated transition response is extremely poor; signal processing technology was required to correct this asymmetry.
(4) No system manufacturers expressed interest in BaFe tape or were willing to conduct joint development.

4 Scenario for market entry and execution

4.1 Scenario for market entry

Research on BaFe tape media was initiated in 1992 by three engineers. After ten years, as we were unable to show results that exceeded the performance of MP tapes, the project was then interrupted several times. Finally, the project was almost terminated. However, with the development of new dispersion and coating technology in 2001, which extracted the potential of the BaFe magnetic material, as well as the introduction of evaluation technology using a high-sensitivity head that enhanced the BaFe tape performance, we could present results that dramatically exceeded the performance of MP tape. Thus, the research project was continued under stimulus (first stage).

Joint research with the IBM Corporation, a system company that possesses leading technology, was later initiated based on these results. We successfully demonstrated a previously unseen high areal recording density, through the development of drive technology that extracted the BaFe tape performance capability (second stage). Based on the results of this joint research, system development was initiated with several tape drive system manufacturers, including IBM. We conducted practical application development and mass-production technology development for the BaFe tape, and introduced various tape products to the market (third stage). Along with product development, we conducted joint research with IBM towards achieving a higher areal recording density using the BaFe tape. The results were published concurrently with the introduction of the aforementioned BaFe tape products to the market. With these announcements, BaFe technology became recognized as being capable of supporting tape systems for several future generations, and this result accelerated the growth of the products that had been introduced to the market (fourth stage). Further details of each of these steps are presented in the following subchapter.

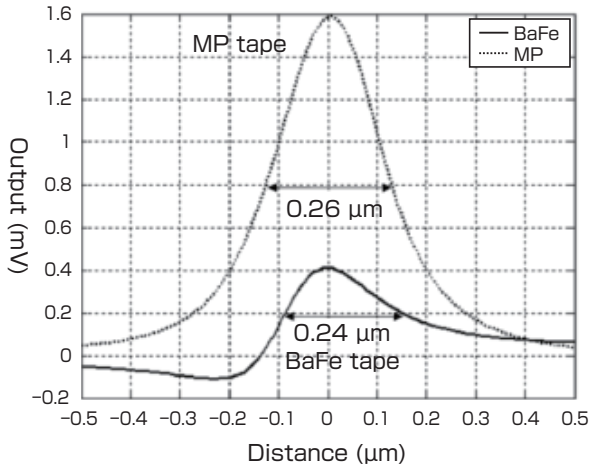


Fig. 8 Comparison of the isolated transition response of BaFe/MP

4.2 Scenario execution

4.2.1 First stage: Basic research (–2003)

Demonstration of recording performance by Fujifilm

To extract the high recording performance expected for BaFe tape by overcoming the issues listed in Subchapter 3.2, we first developed high-dispersion technology and ultra-thin layer coating technology for application to the BaFe tape. In addition, we developed evaluation technology using a high-sensitivity head.

(1)-1: High-dispersion technology

As the BaFe magnetic material is comprised of fine magnetic particles and the shape of the particle is hexagonal plate-like, the plates tend to stack upon each other and aggregate. Thus, the magnetic material must be uniformly dispersed to obtain the fine particle effect. However, when the conventional dispersion method is employed, the fine magnetic particles aggregate into blocks of several to dozens of particles, yielding a non-uniform distribution, as shown in Fig. 9. By shifting from MPs to BaFe magnetic material, we succeeded in achieving monodispersity of the fine particles of the BaFe magnetic material. This was achieved through the development of new organic materials such as dispersants and polymers, as well as the development of a new dispersion process.

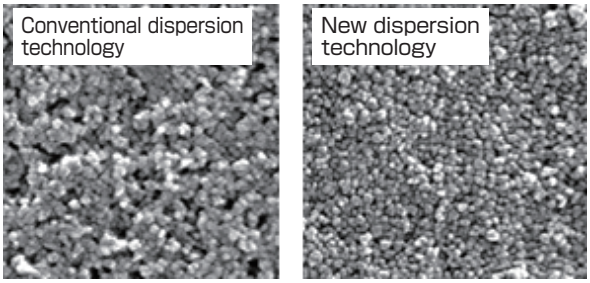


Fig. 9 SEM image of the tape surface

(1)-2: Ultra-thin layer coating technology

Although the achievement of a thin magnetic layer is essential for obtaining a high recording density for a magnetic recording medium, it was difficult to achieve a thin layer of 0.1 μm or less using the conventional coating method. As shown in Fig. 10, it was not possible to obtain a uniform thin magnetic layer using this method because of the turbulence at the interface with the nonmagnetic layer forming the lower layer. This tendency was prominent for the coating solution of the BaFe magnetic material. Therefore, we developed a new coating method and achieved a thin magnetic layer with a thickness of several tens of nanometers, a low thickness variation, and uniformity, with no mixing at the interface with the nonmagnetic lower layer.

(2) Development of evaluation system using high-sensitivity head

At this stage, no high-sensitivity magnetic head had been developed that could adequately extract the BaFe tape performance. Therefore, we decided to conduct media evaluation using the HDD head that was the leading technology at the time. Rather than the tape-evaluating device that was in our possession, we acquired an evaluation device for disks, which featured a merged anisotropic magneto resistive (AMR) head with a 1.9 μm reader-width that was used for HDD. We then conducted the evaluation using a disk-shaped BaFe medium. Figure 11 shows the modulation spectra of the MP medium used at the time and the BaFe medium to which new dispersion and coating technology was applied, at a linear recording density of 180 kfci (flux change per inch). As is apparent from these modulation spectra, the BaFe-medium output was +4 dB compared to that of the MP medium. The BaFe-medium signal-to-noise ratio (indicating the read/write performance of the medium) was +10 dB, which indicated the possibility of achieving a recording density ten times higher than that of the MP medium.[10]

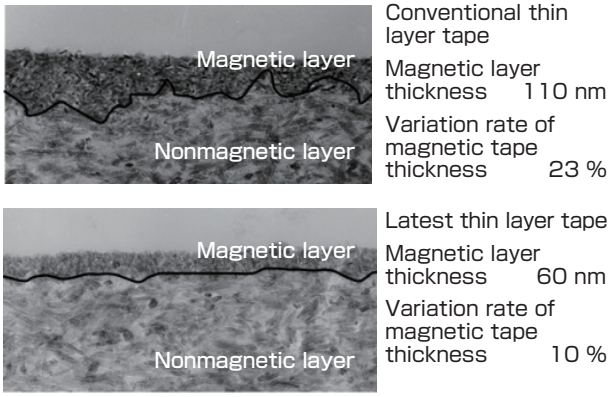


Fig. 10 TEM image of the tape cross-section

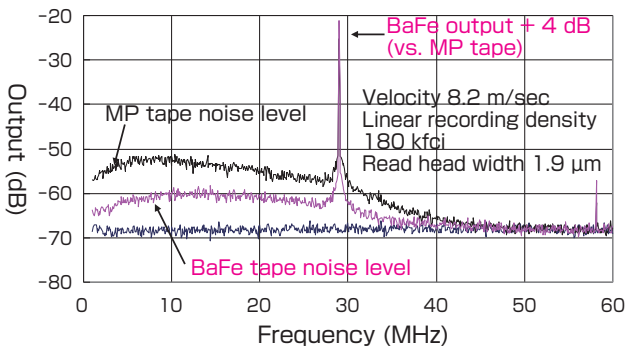


Fig. 11 Modulation spectra of the MP medium and the BaFe medium[10]

4.2.2 Second stage: Research on potential system application (–2006)

Demonstration of recording density for tape-drive system manufacturers

During the first-stage research conducted within our company, we confirmed the high recording performance of the BaFe tape for the first time. However, we were unable to convince tape-drive system manufacturers to begin tape-drive system development for BaFe tape only by the recording performance result of a magnetic tape manufacturer alone. Therefore, in the next research stage, we aimed to conduct joint research involving tape-drive system manufacturers, so as to develop new signal processing technology that would enable waveform correction of the BaFe (as discussed in Subchapter 3.2). In addition, we aimed to build relationships with system manufacturers.

IBM Corporation, the leader in the field of tape-drive system technology, was selected as our partner in this joint research. Following assessment by IBM of our evaluation data and BaFe tape prototypes, along with repeated explanation by the authors of the potential of the BaFe tape, this joint research project was initiated. The major turning point in this research was the selection of the read head. We proposed an evaluation using the giant magneto resistive (GMR) head that was standard for HDD, rather than the AMR head that was used for tape at the time; however, IBM was hesitant in selecting the GMR head, which was unknown in tape industry and was a new device at the time. We conducted evaluation using the GMR head for different systems, such as HDD and demonstrated the high recording performance of the BaFe tape.[11][12] Finally, IBM decided to employ the GMR head and, as a result of this joint research, we succeeded in demonstrating a high areal recording density of 6.7 Gb/in^2 (equivalent to a cartridge volume of 8 TB) for the BaFe tape, which was approximately 17 times the 0.4 Gb/in^2 areal recording density that was common for products in 2006.[13] The technical highlights were as follows: (1) Improved linear recording density was achieved via the combination of the magnetic tape using BaFe fine magnetic particles and the new signal processing technology known as “data dependent noise predictive maximum likelihood” (DD-NPML); (2) Improved track density was obtained using high-precision head-following technology. Through application of the new DD-NPML signal process, which was developed by IBM in this joint research project,[14] the readback waveform

distortion that was detrimentally affecting the BaFe tape performance was corrected and the target error rate was reached. Hence, IBM decided to set BaFe tape as the central focus of next-generation tape technology. At this stage, we had solved issues (3) and (4) of Subchapter 3.2, i.e., the need for waveform asymmetry correction and the selection of a partner for system development. Thus, significant progress had been made toward commercial realization of BaFe tape.

Other than the BaFe tape proposed by Fujifilm, Hitachi Maxell, Ltd., proposed an iron nitride (nano-composite advanced particles; NanoCAP) magnetic material tape,[15] whereas Sony Corporation proposed a metal evaporated (ME) tape.[16] Thus, discussions were initiated among the Information Storage Industry Consortium (INSIC), the industrial organization for tape systems, regarding the next-generation tape that would succeed MP tape.

4.2.3 Third stage: Product realization research (–2011) Establishment of mass production technology for BaFe tape

The initial issues were all solved in the first and second research stages, and BaFe tape became widely known to many system manufacturers through technological demonstrations with IBM. In addition, we provided technical descriptions and prototypes of the BaFe tape to Oracle Corporation, Japan, Hewlett-Packard Japan, Ltd., and Quantum Corporation, and advanced to the practical application stage by repeating the performance evaluation of the BaFe tape. In 2011, the BaFe tape was employed in IBM's 3592 fourth-generation drive[17] and Oracle's T10000 third-generation drive,[18] and product realization of 4 and 5 TB tape cartridges were achieved, respectively. In 2012, the BaFe tape was employed in the LTO sixth-generation drive;[19] thus, the BaFe tape had been introduced to all the targeted major tape systems at that point.

Within our company, effort was expended on developing mass production technology and practical use technology. For mass production, we developed a process that enabled stable production of high-density tape, primarily by upscaling the new dispersion and coating technology mentioned in the previous subchapter. For practical use technology, improvements were primarily made to the durability of the BaFe tape. A tape system must clear severe endurance tests, such as a whole-length run, which may reach tens of thousands of passes for a wide range of temperatures and humidity (10–32 °C, 10–80 % relative humidity (RH)). Work was performed regarding the surface profile design, lubrication, and wear. After the development of this mass production technology and practical use technology, market entry of the BaFe tape, our final goal, was realized.[20]

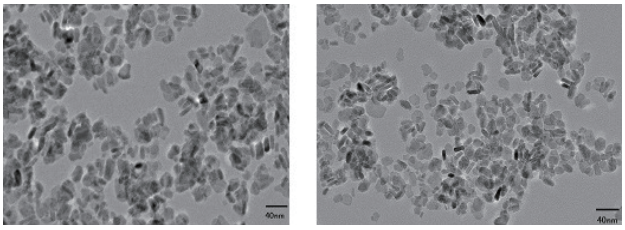
4.2.4 Fourth stage: Research on further possibilities (–2015) Demonstration of higher recording density

The long-desired market entry of the BaFe tape was achieved in 2011. However, the main point of interest for users, who were required to invest highly in accommodating system change, was the potential for future development of the BaFe technology. Therefore, we continued to demonstrate the possibilities for improving the recording density of the BaFe tape, while also implementing product realization research; these future prospects were continuously presented to the market.

In 2010, one year before the market entry of the BaFe product, we achieved a successful technical demonstration of an areal recording density of 29.5 Gb/in^2[21] (equivalent to a 35 TB cartridge volume). For the BaFe tape employed in this technical demonstration, we accomplished the following: (1) a fine particle volume for the BaFe magnetic material of 1600–2100 nm^3 (Fig. 12); (2) an average *Ra* of 0.9–2.0 nm (Fig. 13); and (3) improvement in the degree of orientation in the perpendicular directions from 0.61 to 0.86, by employing perpendicular magnetic field orientation technology for magnetic materials (Fig. 14).

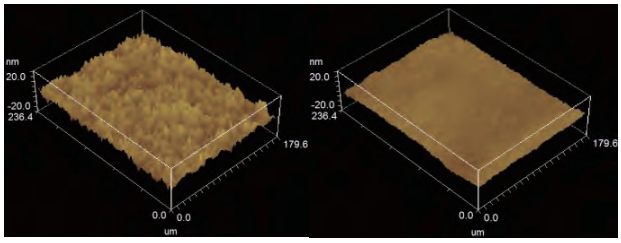
The technology used to achieve these accomplishments ((1) to (3)) were then further advanced and, in 2014, we technically verified an areal recording density of 85.9 Gb/in^2[22] (equivalent to 154 TB). This was later increased to 123 Gb/in^2[23] (equivalent to 220 TB), in 2015.

BaFe tape was introduced to the market more than 20 years after research was initiated. The potential for achieving higher recording density in the next decade has been



BaFe product		29.5 Gb/in^2 demonstration tape	
Particle volume	2100 nm^3	Particle volume	1600 nm^3
Coercivity	182 k/Am	Coercivity	203 k/Am

Fig. 12 TEM image of the BaFe magnetic material[21]



BaFe product	29.5 Gb/in^2 demonstration tape
Ra : 2.0 nm	Ra : 0.9 nm

Fig. 13 Surface profiling using the optical interferometry surface roughness meter[21]

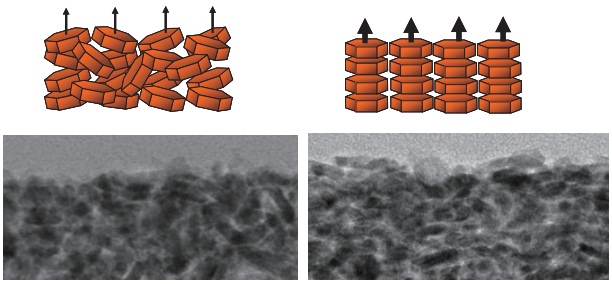
established through the technical demonstrations conducted concurrently with product development. In addition, we have established BaFe tape as the *de facto* standard to replace MP tape.

5 Summary

We successfully responded to the market demand for increased data storage capacity by accelerating the development of high-density tape systems. This was achieved using BaFe magnetic material in place of MP magnetic material, which was approaching its performance limit.

The major factors that enabled this innovation for the establishment of BaFe tape as the *de facto* standard are the following: We pursued the necessary technological development based on a belief in the potential of the BaFe tape, and we sought the involvement of other groups. Of course, the technological innovation of the BaFe magnetic material itself was important; however, the development of peripheral technology to extract the potential of the BaFe magnetic material and BaFe tape was of greater importance. The high density of the BaFe tape could not be achieved without the development of new dispersion and coating technology within our company, along with the development of a high-sensitivity magnetic head and waveform equalization technology outside our company.

A period of approximately 20 years elapsed between the beginning of the research project and product realization. During this time, we continued the research without faltering, because we understood the essential characteristics of the BaFe magnetic material (including its disadvantages), and because we also understood that these characteristics could be effectively employed to achieve high-density magnetic recording in principle. While this case study is not quite the efficient R&D that is in demand today, we hope it constitutes a study of a research process in which the innovation originated from a stance of perseverance, along with the involvement of others based on the essence and characteristics of the developed material.



BaFe product
Orientation ratio (perpendicular direction): 0.61

29.5 Gb/in^2 demonstration tape
Orientation ratio (perpendicular direction): 0.86

Fig. 14 TEM image of the cross-section of magnetic layer[21]

Finally, to respond to the continuously increasing market demand for data storage, we believe further increase to the high density of the tape system will be achieved by incorporating state-of-the-art drive technology innovation for HDD and by developing new high-density technology for tape. A timeline of the main events of this research project is presented in Fig. 15.

1st step	1992	· BaFe tape research started by three researchers.
	2001	· Succeeded in presenting high recording property of BaFe tape for the first time within company, although the research was about to be discontinued.
2nd step	2004	· Started joint research with IBM on recording density demonstration.
	2006	· Fujifilm demonstrated the potential for high recording density of BaFe tape by combination with GMR head.[11][12] · Succeeded in technical demonstration of areal recording density 6.7 Gb/in^2 (volume 8 TB equivalent) for BaFe tape jointly with IBM.[13] · Hitachi Maxell, Ltd. proposed iron nitride tape, Sony Corporation proposed metal evaporated tape, and discussion of post metal tape began in the tape industry.
3rd step & 4th step	2007	· Started providing samples and presentations of BaFe tape to various tape drive system companies.
	2010	· Succeeded in technical demonstration of recording density 29.5 Gb/in^2 (volume 35 TB equivalent) for BaFe tape jointly with IBM.[21]
	2011	· Employed as the tape for Oracle's (SUN at the time) T10000 third-generation system (volume 5 TB). This was the first successful commercialization of BaFe tape. · Succeeded in commercialization as the tape for IBM 3592 fourth-generation system (volume 4 TB).
	2012	· Succeeded in commercialization as the tape for LTO6 (volume 2.5 TB). As a result, BaFe tape was employed in all three major tape storage systems. · Oracle's T10000 fourth-generation system was released. Volume improved to 8.5 TB by using the tape (reuse) for third-generation system.
	2013	· Succeeded in technical demonstration of recording density 85.9 Gb/in^2 (volume 154 TB equivalent) for BaFe tape jointly with IBM.[22]
	2014	· Succeeded in commercialization as the tape for IBM's 3592 fifth-generation system (volume 10 TB). · Succeeded in technical demonstration of recording density 123 Gb/in^2 (volume 220 TB equivalent) for BaFe tape jointly with IBM.[23]
	2015	· Succeeded in commercialization as the tape for LTO7 (volume 6 TB).

Fig. 15 Timeline of the development of BaFe tape

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Authors

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Received MSc. from the Graduate School of Science and Technology, Niigata University, in 1991. Joined Fuji Photo Film Co., Ltd., in 1991 and was assigned to the Recording Media Research Laboratories (formerly *Jiki Zairyo Kenkyujo* or the Magnetic Material Research Laboratory). Primarily worked on the development of civilian magnetic tape, beginning the development of barium-ferrite tape in 2001. Now primarily works on technical demonstration of areal recording density.

Hitoshi Noguchi

Received MSc. from the Graduate School of Engineering, Kyoto University, in 1987. Joined Fuji Photo Film Co., Ltd. in 1987 and was assigned to the Recording Media Research Laboratories (formerly *Jiki Zairyo Kenkyujo* or Magnetic Material Research Laboratory). Began basic research on barium-ferrite media in 1992 and primarily worked on achieving high density in high-volume floppy disks, along with product development of barium-ferrite tape. Has been the director of Recording Media Research Laboratories since 2012. Received the 46th Ichimura Prize in Industry for Outstanding Achievement in 2014 and the 61st Ohkochi Memorial Prize in 2015.

Discussions with Reviewers

1 Overall

Comment (Kimihiro Ozaki, AIST)

This paper clearly presents the relationship between the demand and potential from material development to practical use and product realization, as well as the importance of external collaboration, and I think it is excellent as a research paper for *Synthesiology*. The elemental technologies and the scenario for product realization are clearly described. The fact that product realization was achieved by continuing R&D for 20 years will provide valuable insight, including topic selection and research method development, to researchers and developers of other fields. The content is easy to understand by people of other fields, and I think it is an outstanding paper.

Comment (Isao Kojima, AIST)

I reviewed this paper from the perspective of establishing the *de facto* standard in the IT (information technology) field. The activities for achieving the goal became clear through this review process and I think the authors have completed a very good paper

2 Issues of symmetry

Question & Comment (Kimihiro Ozaki)

In Subchapter 3.2 "Issues related to BaFe magnetic material," you organize the issues to be solved from (1) to (4). Of these, I understood that Issue (3) "signal processing technology to correct the symmetry of isolated transition response" was solved "by the new signal processing DD-NPML designed by IBM" as described in Section 4.2.2 "Second stage: Research on potential system application" of Subchapter 4.2 "Scenario execution." Is my understanding correct?

Is it difficult to solve the symmetry issue through materials? In this paper, it is written that it can't be done "because it possesses perpendicular magnetization components," but does the difficulty arise from the phenomenon that cannot be avoided physically due to the magnetic interaction among the fine particles? Does it arise from the magnetic property of BaFe?

Answer (Takeshi Harasawa)

Yes, issue (3) was solved using DD-NPML.

The issue of waveform symmetry essentially arises from the perpendicular magnetization component. This is because the BaFe magnetic material is plate-shaped and the axis of easy magnetization is perpendicular to the plate surface. After coating and drying of the magnetic solution, the plate-shaped BaFe maintains the magnetization component perpendicular to the tape surface, as the plates tend to align parallel to the tape surface. This is not a phenomenon particular to the magnetic property or BaFe interaction.

Further, as HDD shifted towards perpendicular recording, we considered the perpendicular magnetization component as an advantage, and succeeded in increasing the recording density by employing perpendicular orientation (an increased perpendicular magnetization component) in the 35 TB technical demonstration of 2010.

Question (Isao Kojima)

I evaluate the development of DD-NPML as one key point in resolving Issue (3) (need for signal processing technology to correct the asymmetry). Is this included in the result of the joint research with IBM? If this technology was developed only by IBM, please show how the companies other than IBM could solve this problem since setting the *de facto* standard does not mean relying on the technology of a specific company.

Answer (Takeshi Harasawa)

The development of DD-NPML itself was the work of IBM, and the result of the joint research project was the achievement of a high recording density by combining the BaFe tape and DD-NPML.

The DD-NPML contributes to increased recording density by decreasing the error rate, not just correcting the waveform, as it is a state-of-the-art signal process for a tape system. Therefore, DD-NPML is non-essential if waveform equalization only is required, and I assume that waveform equalization can be achieved using other signal processes.

In fact, HP and Quantum were concerned about waveform distortion when they initially considered the use of BaFe tape. Citing the results presented in Refs. [12] and [13], we requested that the tape-drive system manufacturers take BaFe into consideration, as waveform equalization was technologically possible. As a result, HP and Quantum developed their own signal processes that enabled waveform equalization in the course of their practical applications. This is my understanding of this topic.

3 Competing technology

Comment (Isao Kojima)

Can you describe the situation of technological competition with other companies? Although this was first-in-the-world development still holding the top share, many companies, like Hitachi Maxell, and Sony, are also at a similar technological level. It will be useful to show what impact this development by Fujifilm had on these competitors.

Answer (Takeshi Harasawa)

A summary of papers published by competitors on this technology is given below. I have added a description of the competing technology in 2006 to Section 4.2.2.

Hitachi Maxell, Ltd.

Until 2005 or 2006, this company was publishing papers on particulate tape media using iron nitride as post metal tape. No papers on iron nitride were published in 2012; however, magnetic tape produced using the sputtering method with an estimated cartridge volume of 50 TB was announced by this company.

Major papers

Iron nitride (nano-composite advanced particles; NanoCAP)

- Y. Sasaki, N. Usuki, K. Matsuo, and M. Kishimoto: Development of NanoCAP technology for high-density recording, *IEEE Trans. Magn*., 41 (10), 3241–3243 (2005).

Magnetic tape using sputtering method

- S. Matsunuma, T. Inoue, T. Watanabe, T. Doi, Y. Mashiko, S. Gomi, K. Hirata, and S. Nakagawa: Playback performance of perpendicular magnetic recording tape media for over 50 TB cartridge by facing targets sputtering method, *J. Magnetism and Magn. Matls*., 324 (3), 260–263 (2012).

Sony Corporation

This company engaged in R&D for metal evaporated tape as post metal tape, which was put to practical use in 8-mm videotapes and digital video cassettes (DVCs). Until approximately 2009, Sony Corp. published papers on the practical durability of linear tape systems and recording density improvement. In 2014, they announced a magnetic tape produced using a sputtering method rather than metal-evaporated tape with a recording density of 148 Gb/in^2 (equivalent to 180 TB volume or higher).

Major papers

Metal evaporated tape

- K. Motohashi, T. Sato, T. Samoto, N. Ikeda, T. Sato, H. Ono, and S. Onodera: Investigation of higher recording density using an improved Co-Co metal evaporated tape

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Magnetic tape using sputtering method
- J. Tachibana, T. Endo, R. Hiratsuka, S. Inoue, D. Berman, P.-O. Jubert, T. Topuria, C. Poon, and W. Imaino: Exploratory experiments in recording on sputtered magnetic tape at an areal density of 148 Gb/in^2, *IEEE Trans. Magn.*, 50 (11), Art. ID 3202806 (2014).

4 Technological background for numerical values

Question (Isao Kojima)

In the joint work with IBM, the selection of the head is one of the highlights. Since the references you cite, including References [11] and [12], are of a specialized scientific journal (*IEEE Trans. Magn.*), can you tell us (and also describe in the paper) the technological background such as the numerical values (such as AMR ratio) that supports your claim? According to Reference [11], you achieved 7 Gb/in^2. Does this mean an achievement of the absolute (rather than relative) level?

Answer (Takeshi Harasawa)

I think the supporting points are the high absolute values achieved, including the 7 Gb/in^2 (tape) value reported in Ref. [11] and the 17.5 Gb/in^2 (disk) result reported in Ref. [12].

We convinced IBM to undertake this research based on the demonstration result of the 7 Gb/in^2 recording density, which was more than ten times greater than that of the state-of-the-art system (0.4 Gb/in^2 recording density) at that time. This was achieved through the combination of the BaFe tape and GMR head.

Question (Isao Kojima)

What activities were done in getting the BaFe adopted by LTO? You described the technological presentation to HP and Quantum, but did you aim for the adoption by LTO by promoting it to such various players of LTO including IBM? Or were there direct appeals to LTO by Fujifilm?

Answer (Takeshi Harasawa)

We made appeals to HP and Quantum, which were involved in LTO development, to have the BaFe tape employed in LTO. The activities performed independently by Fujifilm in 2006 and 2007 were as follows:

(1) We developed the GMR head for several types of linear tapes with different read-width jointly with head manufacturers, announced the high performance of our BaFe tape evaluated using this head at the Information Storage Industry Consortium (INSIC), and reported this finding at technical meetings with various tape-drive system manufacturers.

(2) The technically demonstrated 6.7 Gb/in^2 tape released in 2007 was offered to these companies, and we had these tape-drive system manufacturers conduct evaluations.

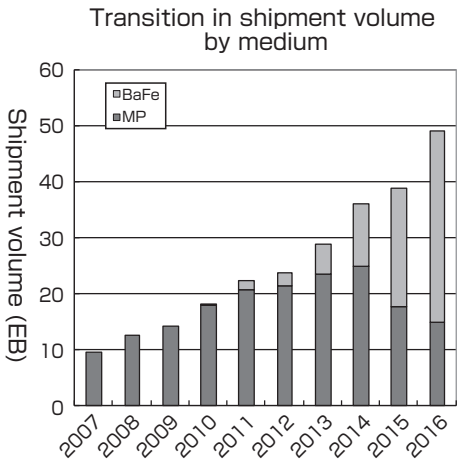
5 Successful scenario seen from market share

Question (Isao Kojima)

Considering this as a successful scenario, I think it should be eventually reflected in the market share such as shipment volume and share of tapes. I imagine that you have some data which shows the growth of the BaFE tapes and the decline of MP tape shipments, as well as the good standing of Fujifilm in the market. Is it possible to publicize these data?

Answer (Takeshi Harasawa)

The following graph shows the total shipment volume trend for linear tape [number of cartridges × volume/cartridge; the *x*-axis units are exabytes (EB)]. The switch from MP tape to BaFe tape occurred progressively and, in 2016, BaFe tape dominated the market, with an approximately 70 % market share (figures from Fujifilm).



Development of a cell microarray chip system for early and accurate malaria diagnosis

— Finding one parasite in 2 million erythrocytes for elimination of malaria —

Muneaki HASHIMOTO*, Shouki YATSUSHIRO, Shohei YAMAMURA and Masatoshi KATAOKA

[Translation from *Synthesiology*, Vol.10, No.1, p.33–40 (2017)]

Early and accurate diagnosis of malaria is needed to prevent the spread of this parasite. To this end, we developed a novel microarray chip system for the detection of malaria, and evaluated it in Africa. A chip with approximately 20,000 microchambers was developed to detect malaria parasites (hereafter called a cell chip). Leukocytes were removed by filtration columns from whole blood cells. An erythrocyte suspension containing fluorescent nuclear staining dye was dispersed onto the cell chip surface and washed, creating an erythrocyte monolayer in each microchamber that contains more than 2 million erythrocytes. Malaria parasite-infected erythrocytes are then detected using a fluorescence detector. Accurate and rapid detection of the parasites with high sensitivity was achieved by the developed system.

Keywords : Malaria, diagnosis, cell microarray chip, field work

1 Background of research

Malaria is an infectious disease transmitted by anopheles mosquitoes, and is one of the three major infectious diseases of the world. Every year, about 2 hundred million people are infected and 430 thousand people die. As a global strategy for suppressing malaria, the development of a quick and accurate diagnostic method is given as one of the most important issues.[1] Malaria diagnosis is done by microscopic observation of thin blood films stained with Giemsa stain (the Giemsa microscopy) and this has been the gold standard for over 100 years. This is an excellent method that not only detects malaria, but also enables diagnosis of the infection rate (severity). It is conducted in the following steps: (1) a drop of a patient's blood is collected, (2) a thin blood film is prepared on a slide glass, (3) the thin film is stained using the Giemsa solution, and (4) malaria parasites in the stained erythrocytes are observed through the microscope. However, accurate diagnosis cannot be made without a technician skilled in this method. Also, time is necessary (normally, about 30 min to 1 h), and it is a labor-intensive work since several thousands to several ten thousands of erythrocytes must be observed. Therefore, quick diagnosis is difficult with this conventional gold standard method, and early diagnosis of patients with low infection rates is impossible.

In the current medical settings of developing countries, rapid diagnosis tests (RDT), which employ immunochromatography as the principle and can be conducted by easy operation in a short time (20 min), are widely used. However, the detection limit of RDT is equivalent to the analysis of Giemsa microscopy, and incidences of false-positives and false-negatives are common. Therefore, it is used as a preliminary screening method prior to the definitive diagnosis by the Giemsa microscopy. It is not possible to calculate the infection rate with RDT (i.e. diagnosis of infection only), and this is one of the disadvantages. Recently, new diagnostic methods utilizing the flow cytometer and polymerase chain reaction (PCR) have been developed, but the sensitivity is insufficient for early diagnosis, and several hours are required before the results are obtained, respectively.[2]–[6] To prevent infection by early detection of malaria, development of a new diagnostic method with high sensitivity, accuracy, quickness, and easy operation is demanded. The characteristics of each diagnostic method are shown in Table 1.

We focused on the microchip technology to conduct analysis at high throughput, ultra-high sensitivity, and with ease for individual cells.[7]–[10] In this paper, we shall describe the "scenario" in which this technology was applied to develop a device that enabled highly sensitive diagnosis in an actual malaria-endemic region (Fig. 1).

2 Development of the cell chip for malaria diagnosis

In starting the development of a malaria diagnostic method, we considered it necessary to be able to measure the infection rate (what percentage of a patient's erythrocytes is infected with malaria parasites) that showed the severity of malaria, as well as the presence of infection, in order to diagnose the

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Table 1. Comparison of malaria diagnostic methods

	Giemsa microscopy	Rapid diagnosis test (RDT)	Polymerase chain reaction (PCR)	Flow cytometer	Cell chip
Detection limit	0.01 %	0.01 %	0.0005 %	0.0005 %	0.00005 %
Detection time	60 min	20 min	6 h	5 min	15 min
Difficulty level	Ultra-high	Low	High	Low	Low
Cost※	Low	Medium	High	Ultra-high	Medium–High

※Does not include cost of labor

disease at the medical settings of developing countries and then to engage in effective treatment. That was because there were possibilities of delayed early diagnosis, and increased risk of appearances of drug-resistant parasites if drugs were prescribed according to symptoms only such as fever. Therefore, we did not select improving the existing methods such as PCR and immunochromatography that merely detect the presence of infection. That is, we thought the method of counting the number of parasites was good, considering the facts that the Giemsa microscopy was considered the gold standard, was most universally used, and had high accuracy, while the PCR and immunochromatography methods had problems of producing false-positives and false-negatives. Among the existing methods, the method of counting cells included the flow cytometer method, and it was possible to calculate the infection rate. However, it was necessary to increase the number of measured cells to increase the measurement precision, and if one attempted to detect the low infection rate of one parasite in several million erythrocytes, 100 or more parasites had to be counted (refer to https://www.bc-cytometry.com/FCM/immunologyFCM_02.html). Taking into consideration the ratio of one in several million cells, it meant that over a hundred million erythrocytes had to be counted, and this would lengthen the detection time and would not lend to a quick diagnostic method. Moreover, it was too expensive for practical application if it were to be used or sold in African developing countries. In such conditions, our research group decided to utilize microfabrication technology used in the fields of MEMS and μTAS, to apply the technology of individually placing the cells on plastic substrate microarrays to erythrocytes, to arrange as many erythrocytes in a monolayer, to seek out erythrocytes infected with malaria parasites, and then to calculate the infection rate.

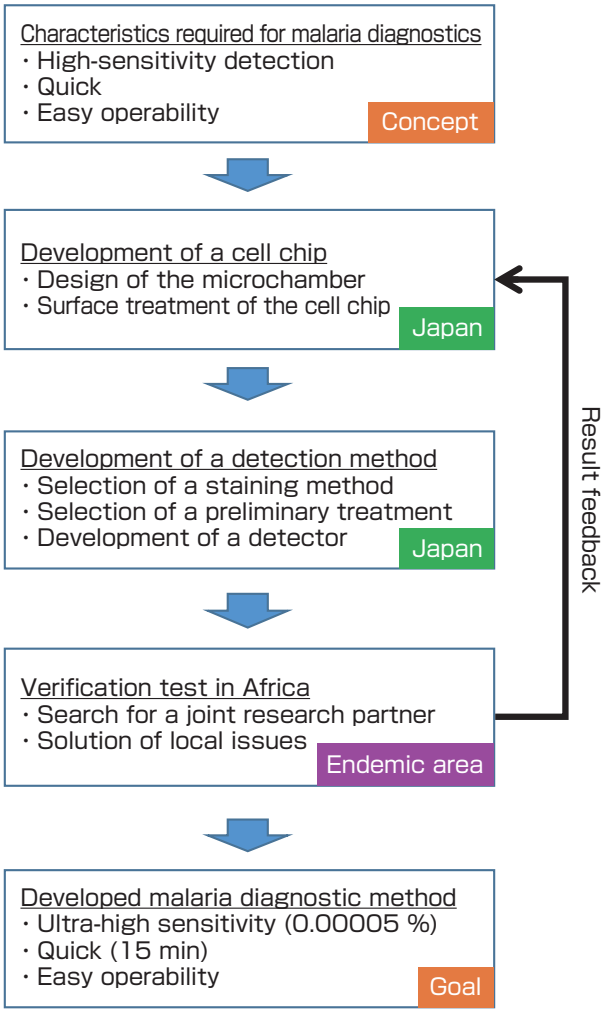

Fig. 1 Flowchart of how this research was carried out (scenario)

While details will be explained later, when erythrocytes infected with malaria parasites and their nuclei stained by fluorescence are placed on a cell chip, the erythrocytes are deposited in a monolayer in the microchamber, and it becomes possible to detect the parasites only. The goal was set of detection limit equivalent or higher than the PCR method, and we considered fabricating a polystyrene cell chip with about 20,000 microchambers. For the optimization of the cell chip, the major premise was to control the number of cells to be measured at a constant number to facilitate the calculation of the infection rate. The flow cytometer method is designed to calculate the number of measured cells by counting the erythrocytes with a detector while allowing the cells to flow along, but our goal was to control the number of cells at a certain level quickly and accurately without counting, or in other words, to accurately arrange a certain number of individual cells by advancing and developing a new design of the conventional cytobiological handling technology. When we came up with the design of cell chips

that might enable the arrangement of a certain number of cells without complicated maneuvers, we intuitively knew this method was a breakthrough. After numerous discussions with joint researchers, we thought it would be possible to arrange a large number of cells in a monolayer in each microchamber by controlling the diameter and the depth of the microchambers. We also thought that the optimal diameter of the microchambers would hold about 100 erythrocytes, and this took into consideration that if one parasite was present in the chamber, the infection rate would be 1 %, and it could also be easily used in the following culture and reaction such as in drug resistant tests. That is, to conduct a drug resistance test by a conventional method, it was necessary to use whole blood, but using this cell chip, it was only necessary to target the cell in the chamber in which the infection was found, and if there were 100 cells, there would be very few problems in conducting the test. Considering the above factors and the size of the erythrocytes, the diameter was set as 105 µm. Since we were also aiming for detection limit equivalent or higher than the PCR method, we determined the number of necessary microchambers. The final designed cell chip has 20,994 microchambers, and each individual chamber has a diameter of 105 µm and a depth of 50 µm (Fig. 2).

Since there was a possibility that infected erythrocytes would not be seen if the erythrocytes were stacked up on top of each other in the microchambers, it was necessary to arrange them in a single layer to enable analysis by microscope or a microarray scanner. When the erythrocytes were statically placed for 10 min on the cell chip, the erythrocytes gathered in the chamber by gravity. However, it was found that when the surface of the cell chip was gently washed with physiological saline, only the erythrocytes of the lowermost layer that adhered to the bottom of the chamber remained, the others were washed away, and the single-layering of erythrocytes was achieved (Fig. 3). The number of erythrocytes in each chamber was almost constant at 130 ± 6 cells, and it became possible to control the erythrocytes to a certain number without counting. This cell chip allowed analysis of about 2.7 million erythrocytes at once, and we obtained the detection limit equivalent to or higher than the PCR method.

The design of a microchamber that allowed monolayer arrangement of erythrocytes by a simple maneuver was done by trial-and-error using various shapes (such as diameters and depths). Recently, the flow of fluid within the microchamber was simulated by a computer, and it was shown that there was almost no flow at 10 µm from the bottom.[7] This provided theoretical support that the erythrocytes of the lowermost layer would not be removed by the washing procedure. In the future, we believe this simulation technology will be useful for the design and optimization of the microchambers of different sizes. Moreover, the important factor in monolayer arrangement of erythrocytes in the microchamber is the hydrophilization of the cell chip surface by oxygen plasma treatment. This treatment will increase the adhesion of erythrocytes to the chamber bottom, and monolayer formation is more easily controlled. The hydrophilization of plastic substrates is Yamamura's specialty, and he has conducted hydrophilization treatment optimized for various

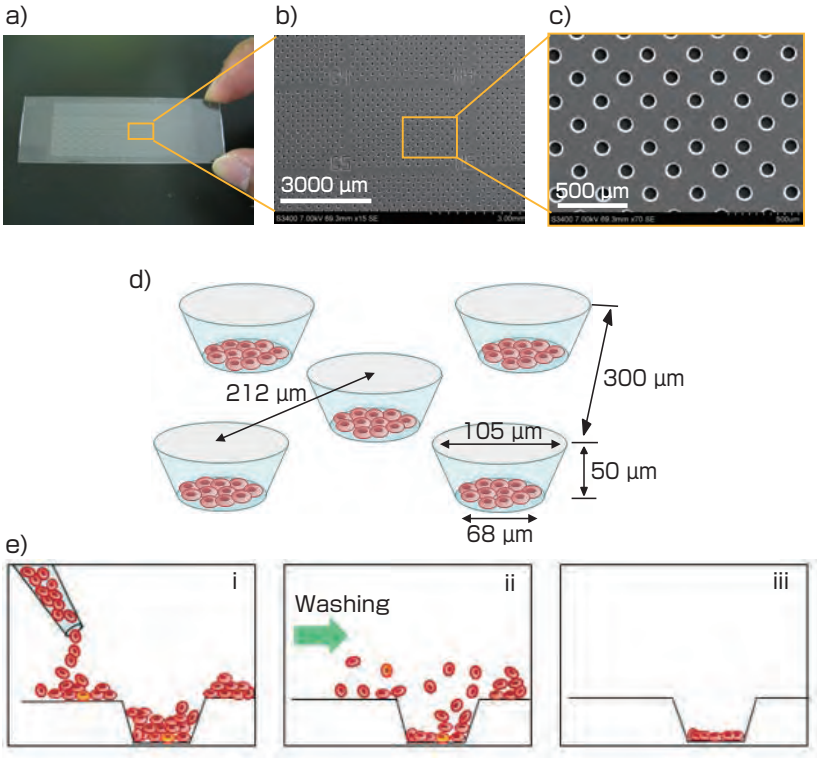

Fig. 2 Structure of the cell chip and single layering of erythrocytes in the microchamber[8]
(a) The size of the cell chip is about the same as the slide glass. (b, c) SEM image of the cell chip. The cell chip is made of polystyrene, and has 20,944 microchambers. It is formed by arrangement of 112 clusters (14 × 8) composed of 187 microchambers. (d) The microchambers have diameters of 105 µm and depths of 50 µm, and are separated from each other by 300 µm. The bottom of the chamber has a diameter of 68 µm, and is cone-shaped. This shape is appropriate for single-layering the erythrocytes. (e) The process by which the erythrocytes become arranged in a monolayer in the microchamber is shown: (i) erythrocytes are placed on the cell chip, (ii) erythrocytes that are stacked on top of each other are removed, and (iii) erythrocytes are arranged in a monolayer.

cells before this project was started. Hence, we were able to conduct optimization for erythrocytes smoothly.

3 Development of the method to detect malaria parasites on the cell chip

With the development of the cell chip, it became possible to arrange the erythrocytes in a monolayer and it became easy to keep the number of cells to be measured at a certain number. Next, we investigated the parasite detection method that was suitable for the cell chip. For parasite detection, the basic concept was to keep the maneuver as simple as possible. Therefore, it was not desirable to have methods that required imparting membrane permeability using surfactants or others, multiple washing procedures, or antibodies that specifically identified the parasites. We considered using the characteristics that erythrocytes do not have nuclei and that malaria parasites are parasitic only to erythrocytes. That is, we decided to stain the nucleic acid (DNA) that is the index of the presence of a nucleus using a fluorescent reagent, and to flag the erythrocyte with a fluorescent signal as an infected erythrocyte.

The concept of the above detection method was to keep the maneuver as simple as possible, and we set the requirements for the nucleic acid staining method as follows: parasites will be kept alive (no need for fixing), staining can be done easily and quickly (no need for washing), and detection can be done by a fluorescent method at high sensitivity. We compared almost all commercially available fluorescent stains that stained DNA and RNA (regardless of membrane permeability). As a result, we found that the SYTO reagent had excellent membrane permeability and stable staining, and we selected SYTO21 that had low background.

To apply this method to actual malaria patients, it was necessary to remove the leucocytes that were nucleated cells from the whole blood. Often, it was difficult to use the centrifuge in malaria-endemic areas due to lack of stable electric power supply. Therefore, we used several types of push columns that did not require centrifuge, and compared the amount of necessary blood and the removal rate of leucocytes. As a result, we confirmed that 99.9 % or more of leucocytes (standard WBC: 3200–9000/μL, RBC: 3.6–5.0 million/μL) could be removed from the whole blood using columns of silicon oxide nanofibers made by Panasonic Corporation. This push filter could be used for patients with sickle cell disease that was prevalent in the malaria-endemic areas.

As a detector, we used the CCD camera system (EZBLMLH01T) with a fluorescent detector from Panasonic. To complete the cell chip scanning in five minutes, it was equipped with a 480 nm semiconductor laser, objective lens, an optimized fluorescence detection filter, and an XYZ axis automatic positioning stage. To detect the parasite, it is possible to detect the fluoro-positive erythrocyte by setting the cell chip in the detector and pressing the “Analyze” button. The resolution of this device is 1.1 μm. It is possible to detect the parasite-infected erythrocyte specifically from the form of fluorescence. The malaria-infected erythrocyte was set as having 1.3 times or more and 7.5 times or less of fluoro-signal intensity compared to non-infected erythrocytes. It was programmed to detect as noise when the aspect ratio (vertical to horizontal ratio) of the fluorescent spot was 2.8 or more, or when the surface area was 4.8 μm^2 or less or 45 μm^2 or more. These figures were actually determined by comparing the blood samples of many malaria patients by microscopic observation of cell chip and Giemsa stained images, identifying the source of fluorescence (leucocyte, platelets, fragments of broken cells, etc.), and by specifically detecting malaria parasites only. The workflow from blood sampling to parasite analysis is shown in Fig. 4.

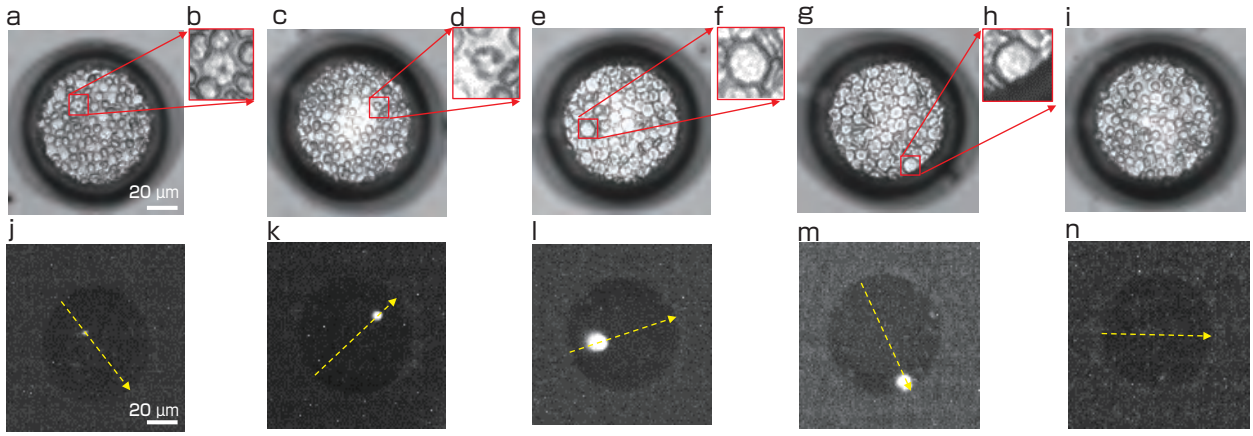

Fig. 3 Separation of the leucocytes and infected erythrocytes in the microchamber
(a,c) The bright field microscope images of infected erythrocytes in the microchamber. (b, d) Enlarged photographs of the infected erythrocytes, and (e, g) the bright field microscope images of cells in whole blood in the microchamber. (f, h) Enlarged photographs of the leucocytes, and (i) a bright field microscope image of non-infected erythrocytes in the microchamber. (j–n) Fluorescent images of cells in each chamber stained with SYTO21 and photographed with a CCD camera. It can be seen that the signal intensities and sizes differ greatly between malaria parasites and leucocytes.

4 Analysis using the cell chip in the endemic area

Gulu District, Uganda is a highly malaria-endemic area, and the research base for the verification test was established in this district. The research base was Lector Hospital, and Professor Horii of Osaka University and Professor Mita of Juntendo University were conducting field research to develop vaccine and to study drug-resistant malaria, and we were able to accompany them. We were extremely grateful since it takes time and effort to find joint research partners from scratch, including the signing of agreement for joint research. We conducted the microscopic observation to analyze the Giemsa stained image of blood samples of 41 patients who visited the hospital. We found parasites in 37 patients, and the infection rate was from 0.0039 % to 2.34 %. Analysis was done using the cell chip for the same samples, and the infection rate was 0.0033 % to 2.39 % (Fig. 5). The values of infection rates calculated by the Giemsa microscopy and the cell chips were almost the same, and showed positive correlation ($R^2 = 0.9945$).

In reality, there were major problems in obtaining the above data. First was the issue of the power source. Gulu is the second largest city in Uganda after its capital Kampala, but it was subject to frequent power outage. Since the power outage could continue for half a day or more, a high quality uninterruptible power supply system was necessary. Also, the transportation of supplies was difficult. Since about two travel days were necessary from Japan to Gulu, reagents that required refrigeration could not be used. When the machineries were checked in with the airlines, they were

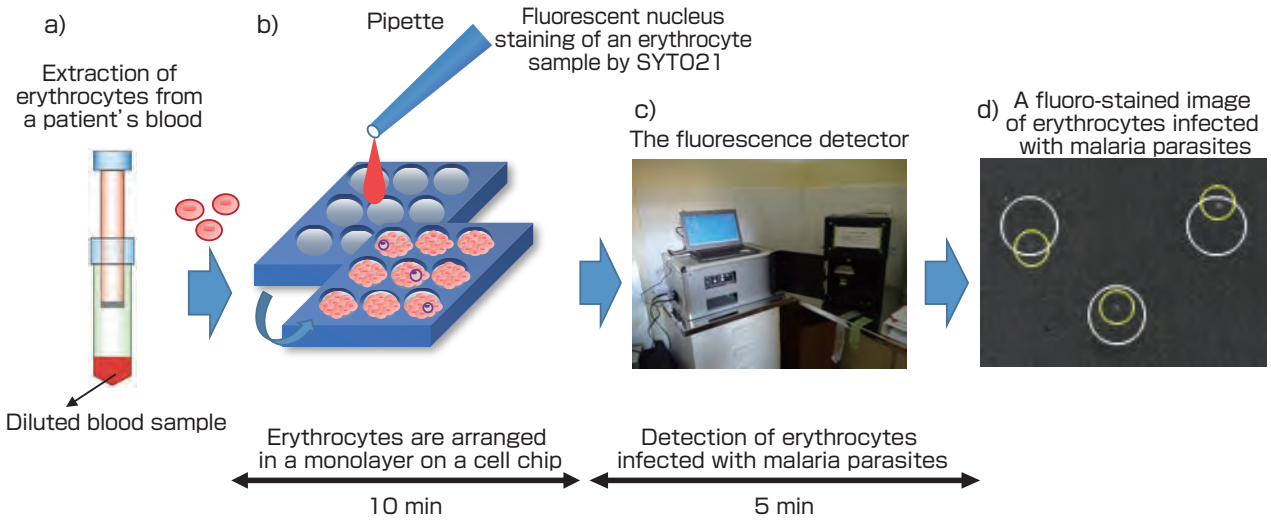

Fig. 4 Workflow of the detection of erythrocytes infected with malaria parasites, using the cell chip[8]
(a) Erythrocytes are isolated from the patient's whole blood using push columns. (b) Erythrocytes are stained with fluorescent nucleus staining reagent (SYTO21), to stain the nuclei of malaria parasites. Then, erythrocytes are placed statically on the cell chip to form a monolayer in the chamber. (c) Malaria parasites are detected by detecting the fluorescence. (d) Number of detected parasites is counted by automatic image analysis. It is possible to calculate the infection rate since the number of erythrocytes in the chamber is constant.

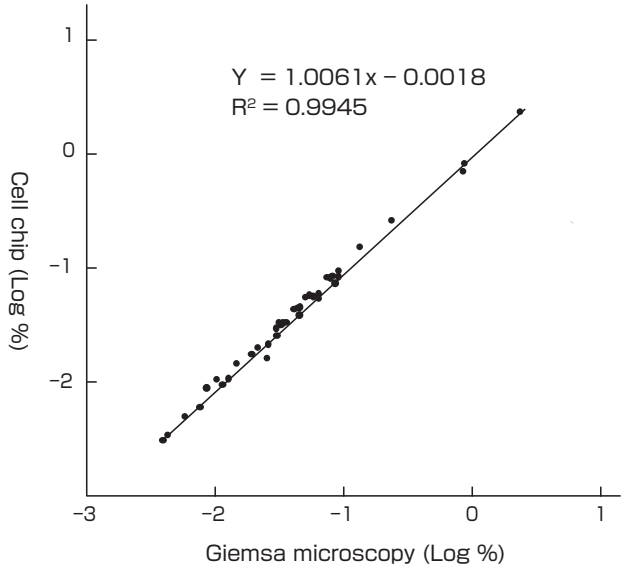

Fig. 5 Measurement of the infection rate using cell chips in the malaria-endemic area[8]
Using the cell chip (vertical axis) and Giemsa microscopy (horizontal axis), the infection rates of patients infected with malaria were calculated, and comparative analysis was conducted for Uganda

handled roughly and were broken, and there was no way of repairing them locally. We became painfully aware of the importance of selecting reagents that were stable at room temperature, downsizing the machines and devices, and obtaining reliable transportation routes.

When the above problems were solved, we thought all we had to do was to set up the experimental apparatuses and collect data. However, at the start of field research, we received almost no blood samples of patients. This was puzzling because there were many malaria patients visiting the hospital which was our joint research partner. When we asked the joint research physician, we found that the flow of action from obtainment of informed consent for research by the local physicians from the patients, the sampling of blood samples, and the transportation of samples to us was not functioning smoothly. We understood that the hospital was always full of patients and busy. It was necessary to talk to the personnel to have them understand the importance of this research. The blood samples of the 41 patients that we received after experiencing such hardship are very valuable. To publish this diagnostic system to the internationally recognized World Malaria Report, we need to obtain data of about 500 cases. From the perspective of hospitals in Japan, it may seem easy to gather 500 cases, but we face hardship in Africa.

5 Discussion and future prospects

Our cell chip technology has the potential of detecting one malaria parasite from 2.7 million erythrocytes (infection rate of 0.00005 %). In the research up to now, we have succeeded in detecting the parasites from blood of infected patients at an infection rate of 0.0033 % in the malaria-endemic area, and this surpassed the detection limit of the Giemsa microscopy (0.01 %) that is the current gold standard. That is, it is possible to detect the early stages of infection that cannot be detected by the Giemsa microscopy. The figure 0.0033 % is about two digits higher than 0.00005 %, but this is because most patients visit the hospital at advanced stages of infection. If we can obtain blood samples at early stages immediately after infection, we believe detection at a lower infection rate will be possible. On the other hand, for the problem of false-positives, out of the 41 samples, detection was not possible even by the cell chip method in four samples that was not detected by the Giemsa stain method. There were samples that turned out positive by RDT and PCR methods, and it was shown that false-positives can easily occur. For false-positives by the PCR method, cases where parasite DNA remained in the blood after treatment of malaria had been reported.[11] One of the major characteristics of the result of the cell chip is that false-positives do not occur since the malaria parasite that is actually inside the erythrocyte can be detected and visualized.

Currently, the treatment of malaria is done by artemisinin-based combination therapy (ACT). Appropriate diagnosis is important for reducing the cost and to prevent the occurrence and dispersal of drug-resistant malaria by preventing the overuse of ACT.[12] That is, the development of a high-throughput and high-sensitive diagnostic method contributes in controlling malaria, by maximizing the use of existing treatment drugs without necessitating new drug development that requires enormous amount of time and cost. The diagnostic method using the cell chip is highly sensitive, quick, and easy to handle. Moreover, the supplies needed for this diagnosis (cell chips, SYTO21, push columns, etc.) are relatively inexpensive (2 USD or less), but there are issues that the detection device is expensive (8,000 USD), bulky and inconvenient to carry, and is difficult to use in fieldwork such as at villages and elementary schools.

Prototype detectors are being developed for downsizing and cost reduction (WHO aims for 1 US or less for cost of a test per person), and we hope to create a device that can be used in wide ranging fields and is affordable in malaria-endemic regions, and we will work toward commercialization. In the future, by advanced diagnosis of malaria, we hope to enable treatment at early stages of infection without inducing drug resistance, and to contribute to the suppression of malaria. For advanced diagnosis, ultra-high sensitive detection of malaria parasites is insufficient, and the development of a “highly functional diagnostic cell chip” that can identify five parasite species and can check whether there is drug resistance is essential. Currently, we are engaging in this development.

Acknowledgements

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Discussions with Reviewers

1 Overall

Comment (Hiroaki Tao, AIST)

This paper is about the development of a device for early detection of malaria infection, which is one of the three major infectious diseases of the world. It explains the concept of a

diagnostic method to be used locally in Africa, the selection of reagents and development of a cell chip that is original technology to realize the concept, comparison with other methods such as the Giemsa stain method that is the gold standard, the problems and solutions in using the device for a verification test in Africa, and the future prospects. It will be good reference for researchers who are working on the R&D of diagnostic devices for practical use in society, and it is significant as a paper to be published in *Synthesiology*.

Comment (Noboru Yumoto, National Cerebral and Cardiovascular Center)

This paper describes the process of developing an accurate, highly sensitive, quick, and easy-to-operate diagnostic device, by utilizing the microchip technology in malaria diagnosis. To solve the issues that were difficult to solve by conventional diagnostic methods, it progresses all the way to local field testing with a clear scenario and international industry-academia-government collaboration. I think it is suitable as a paper for *Synthesiology*.

2 Clarification of the scenario

Comment (Hiroaki Tao)

I think this research was done (scenario) under the strategy of advancing the diagnostic method by repeating the four processes of "concept of diagnostic method" → "R&D of device and reagent to realize the concept" → "verification test in Africa" → "discovery of new technological development needs," and then practical use in society. I think the overall research will be better understood if you show this as a diagram at the end of Chapter 1. Why don't you talk about the simulation of fluid flow within the chamber and the hydrophilization treatment to control the adhesion of the lowermost cell? I think it will be good to state that you have theoretical support by simulation rather than designing only by experience, and that the simulation technology is useful in the optimization and design of chambers of different sizes. Also, other cell chips have been designed so that only one cell would fit in each chamber. I think you should describe the advantage of your cell chip compared to those.

Comment (Noboru Yumoto)

For research papers of *Synthesiology*, the requirement is the authors' originality for the scenario and the element constitution (selection and integration). While the current manuscript allows grasping of the overall image, it is rather difficult to understand how the authors synthesized the elements based on the scenario. I think the key to the realization of the scenario is the development of the cell chip. I think this was realized by combining the technological elements such as the selection of the substrate, design, processing, and surface treatment, and I feel there isn't much description of those elements. Also, for the field test, I think you should explain what issues were set forth by WHO and what you were trying to solve, and how you found the partners of joint research.

Answer (Muneaki Hashimoto)

• I added the flow chart of the scenario as Fig. 1.

• I added the technological elements for fabricating the cell chip as much as possible. For simulation, I added an explanation to the final paragraph of Chapter 2 citing a recently published paper, and stated that it would be useful in the future. I additionally wrote about hydrophilization treatment in the same paragraph. In this cell chip, about 100 erythrocytes are arranged in one microchamber, and if the infection rate is 1 %, one signal is detected in one chamber. Therefore, one of the aims of this cell chip is that when the infection rate is at a certain level, it is not necessary to analyze multiple chambers, and that will be its advantage. I added this point to the text. For the selection of the substrate, we actually tried several plastic materials, but the results were not much different, and therefore, it was not mentioned in this paper.

• For the issue raised by WHO, priority was cost. For the cell chip, some progress can be made on the running cost, but the problem is the expensive detection device. The one that we are developing jointly with companies have the same detection principle, but I think it will clear this cost issue. WHO states that the cost of analysis per person must be 1 USD, and I clearly wrote down this point in Paragraph 3, Chapter 5.

• On how we found our joint research partner, I added some lines to Paragraph 1, Chapter 4.

3 Comparison with existing methods

Comment (Hiroaki Tao)

I think the readers will better understand the overall picture of the current malaria diagnostic methods and the characteristic of this method, if you create a table of the principles, advantages, and disadvantages of this cell chip method along with Giemsa stain, immunochromatography, PCR, and flow cytometer methods.

Answer (Muneaki Hashimoto)

I newly created Table 1.

Editorial Policy

Synthesiology Editorial Board

Objective of the journal

The objective of *Synthesiology* is to publish papers that address the integration of scientific knowledge or how to combine individual elemental technologies and scientific findings to enable the utilization in society of research and development efforts. The authors of the papers are researchers and engineers, and the papers are documents that describe, using "scientific words", the process and the product of research which tries to introduce the results of research to society. In conventional academic journals, papers describe scientific findings and technological results as facts (i.e. factual knowledge), but in *Synthesiology*, papers are the description of "the knowledge of what ought to be done" to make use of the findings and results for society. Our aim is to establish methodology for utilizing scientific research result and to seek general principles for this activity by accumulating this knowledge in a journal form. Also, we hope that the readers of *Synthesiology* will obtain ways and directions to transfer their research results to society.

Content of paper

The content of the research paper should be the description of the result and the process of research and development aimed to be delivered to society. The paper should state the goal of research, and what values the goal will create for society (Items 1 and 2, described in the Table). Then, the process (the scenario) of how to select the elemental technologies, necessary to achieve the goal, how to integrate them, should be described. There should also be a description of what new elemental technologies are required to solve a certain social issue, and how these technologies are selected and integrated (Item 3). We expect that the contents will reveal specific knowledge only available to researchers actually involved in the research. That is, rather than describing the combination of elemental technologies as consequences, the description should include the reasons why the elemental technologies are selected, and the reasons why new methods are introduced (Item 4). For example, the reasons may be: because the manufacturing method in the laboratory was insufficient for industrial application; applicability was not broad enough to stimulate sufficient user demand rather than improved accuracy; or because there are limits due to current regulations. The academic details of the individual elemental technology should be provided by citing published papers, and only the important points can be described. There should be description of how these elemental technologies are related to each other, what are the problems that must be resolved in the integration process, and how they are solved (Item 5). Finally, there should be descriptions of how closely the goals are achieved by the products and the results obtained in research and development, and what subjects are left to be accomplished in the future (Item 6).

Subject of research and development

Since the journal aims to seek methodology for utilizing the products of research and development, there are no limitations on the field of research and development. Rather, the aim is to discover general principles regardless of field, by gathering papers on wide-ranging fields of science and technology. Therefore, it is necessary for authors to offer description that can be understood by researchers who are not specialists, but the content should be of sufficient quality that is acceptable to fellow researchers.

Research and development are not limited to those areas for which the products have already been introduced into society, but research and development conducted for the purpose of future delivery to society should also be included.

For innovations that have been introduced to society, commercial success is not a requirement. Notwithstanding there should be descriptions of the process of how the technologies are integrated taking into account the introduction to society, rather than describing merely the practical realization process.

Peer review

There shall be a peer review process for *Synthesiology*, as in other conventional academic journals. However, peer review process of *Synthesiology* is different from other journals. While conventional academic journals emphasize evidential matters such as correctness of proof or the reproducibility of results, this journal emphasizes the rationality of integration of elemental technologies, the clarity of criteria for selecting elemental technologies, and overall efficacy and adequacy (peer review criteria is described in the Table).

In general, the quality of papers published in academic journals is determined by a peer review process. The peer review of this journal evaluates whether the process and rationale necessary for introducing the product of research and development to society are described sufficiently well.

In other words, the role of the peer reviewers is to see whether the facts necessary to be known to understand the process of introducing the research finding to society are written out; peer reviewers will judge the adequacy of the description of what readers want to know as reader representatives.

In ordinary academic journals, peer reviewers are anonymous for reasons of fairness and the process is kept secret. That is because fairness is considered important in maintaining the quality in established academic journals that describe factual knowledge. On the other hand, the format, content, manner of text, and criteria have not been established for papers that describe the knowledge of "what ought to be done." Therefore, the peer review process for this journal will not be kept secret but will be open. Important discussions pertaining to the content of a paper, may arise in the process of exchanges with the peer reviewers and they will also be published. Moreover, the vision or desires of the author that cannot be included in the main text will be presented in the exchanges. The quality of the journal will be guaranteed by making the peer review process transparent and by disclosing the review process that leads to publication.

Disclosure of the peer review process is expected to indicate what points authors should focus upon when they contribute to this journal. The names of peer reviewers will be published since the papers are completed by the joint effort of the authors and reviewers in the establishment of the new paper format for *Synthesiology.*

References

As mentioned before, the description of individual elemental technology should be presented as citation of papers published in other academic journals. Also, for elemental technologies that are comprehensively combined, papers that describe advantages and disadvantages of each elemental technology can be used as references. After many papers are accumulated through this journal, authors are recommended to cite papers published in this journal that present similar procedure about the selection of elemental technologies and the introduction to society. This will contribute in establishing a general principle of methodology.

Types of articles published

Synthesiology should be composed of general overviews such as opening statements, research papers, and editorials. The Editorial Board, in principle, should commission overviews. Research papers are description of content and the process of research and development conducted by the researchers themselves, and will be published after the peer review process is complete. Editorials are expository articles for science and technology that aim to increase utilization by society, and can be any content that will be useful to readers of *Synthesiology.* Overviews and editorials will be examined by the Editorial Board as to whether their content is suitable for the journal. Entries of research papers and editorials are accepted from Japan and overseas. Manuscripts may be written in Japanese or English.

Required items and peer review criteria (January 2008)

	Item	Requirement	Peer Review Criteria
1	Research goal	Describe research goal ("product" or researcher's vision).	Research goal is described clearly.
2	Relationship of research goal and the society	Describe relationship of research goal and the society, or its value for the society.	Relationship of research goal and the society is rationally described.
3	Scenario	Describe the scenario or hypothesis to achieve research goal with "scientific words".	Scenario or hypothesis is rationally described.
4	Selection of elemental technology(ies)	Describe the elemental technology(ies) selected to achieve the research goal. Also describe why the particular elemental technology(ies) was/were selected.	Elemental technology(ies) is/are clearly described. Reason for selecting the elemental technology(ies) is rationally described.
5	Relationship and integration of elemental technologies	Describe how the selected elemental technologies are related to each other, and how the research goal was achieved by composing and integrating the elements, with "scientific words".	Mutual relationship and integration of elemental technologies are rationally described with "scientific words".
6	Evaluation of result and future development	Provide self-evaluation on the degree of achievement of research goal. Indicate future research development based on the presented research.	Degree of achievement of research goal and future research direction are objectively and rationally described.
7	Originality	Do not describe the same content published previously in other research papers.	There is no description of the same content published in other research papers.

Instructions for Authors

"Synthesiology" Editorial Board
Established December 26, 2007

Revised April 1, 2017

1 Types of articles submitted and their explanations

The articles of *Synthesiology* include the following types:
• Research papers, reports, commentaries, roundtable talks, and readers' forums
Of these, the submitted manuscripts of research papers, reports, and commentaries undergo review processes before publication. The roundtable talks are organized, prepared, and published by the Editorial Board. The readers' forums carry writings submitted by the readers, and the articles are published after the Editorial Board reviews and approves. All articles must be written so they can be readily understood by the readers from diverse research fields and technological backgrounds. The explanations of the article types are as follows.

① Research papers
A research paper rationally describes the concept and the design of R&D (this is called the scenario), whose objective is to utilize the research results in society, as well as the processes and the research results, based on the author's experiences and analyses of the R&D that was actually conducted. Although the paper requires the author's originality for its scenario and the selection and integration of elemental technologies, whether the research result has been (or is being) already implemented in society at that time is not a requirement for the submission. The submitted manuscript is reviewed by several reviewers, and the reviewers will recommend whether the manuscript should be accepted, revised, or declined. The author completes the final draft based on the discussions with the reviewers. Views may be exchanged between the reviewers and authors through direct contact (including telephone conversations, e-mails, and others), if the Editorial Board considers such exchange necessary.

② Reports
A report describes a development example of technology which has practical value as well as an example of new technology which has been put to practical use. It contains 1) the aim, 2) the process of development (the course to the goal), and 3) the outcomes. The submitted manuscript is checked by the Editorial Board. The authors will be contacted if corrections or revisions are necessary, and the authors complete the final draft based on the Board members' comments.

③ Commentaries
Commentaries describe the thoughts, statements, or trends and analyses on how to utilize or spread the results of R&D to society. Although the originality of the statements is not required, the commentaries should not be the same or similar to any articles published in the past. The submitted manuscripts will be checked by the Editorial Board. The authors will be contacted if corrections or revisions are necessary, and the authors complete the final draft based on the Board members' comments.

④ Roundtable talks
Roundtable talks are articles of the discussions or interviews that are organized by the Editorial Board. The manuscripts are written from the transcripts of statements and discussions of the roundtable participants. Supplementary comments may be added after the roundtable talks, if necessary.

⑤ Readers' forums
The readers' forums include the readers' comments or thoughts on the articles published in *Synthesiology*, or articles containing information useful to the readers in line with the intent of the journal. The forum articles may be in free format, with 1,200 Japanese characters or less. The Editorial Board will decide whether the articles will be published.

2 Qualification of contributors

There are no limitations regarding author affiliation or discipline as long as the content of the submitted article meets the editorial policy of *Synthesiology*, except authorship should be clearly stated. (It should be clearly stated that all authors have made essential contributions to the paper.)

3 Manuscripts

3.1 General

3.1.1 Articles may be submitted in Japanese or English.
Accepted articles will be published in *Synthesiology* (ISSN 1882-6229) in the language they were submitted. All articles will also be published in *Synthesiology - English edition* (ISSN 1883-0978). The English edition will be distributed throughout the world approximately four months after the original *Synthesiology* issue is published. Articles written in English will be published in English in both the original *Synthesiology* as well as the English edition. Authors who write articles for *Synthesiology* in Japanese will be asked to provide English translations for the English edition of the journal within 2 months after the original edition is published.
3.1.2 Research papers should comply with the structure and format stated below, and reports and commentaries should also comply with the same structure and format except subtitles and abstracts are unnecessary.
3.1.3 Research papers should only be original papers (new literary work).
3.1.4 Research papers should comply with various guidelines of research ethics

3.2 Structure

3.2.1 The manuscript should include a title (including subtitle), abstract, the name(s) of author(s), institution/contact, main text, and keywords (about 5 words).
3.2.2 Title, abstract, name of author(s), keywords, and institution/

contact shall be provided in Japanese and English.
3.2.3 The manuscript shall be prepared using word processors or similar devices, and printed on A4-size portrait (vertical) sheets of paper. The length of the manuscript shall be, about 6 printed pages including figures, tables, and photographs.
3.2.4 Research papers, reports, and commentaries shall have front covers and the category of the articles (research paper, report, or commentary) shall be stated clearly on the cover sheets.
3.2.5 The title should be about 10–20 Japanese characters (5–10 English words), and readily understandable for a diverse readership background. Research papers shall have subtitles of about 15–25 Japanese characters (7–15 English words) to help recognition by specialists.
3.2.6 The abstract should include the thoughts behind the integration of technological elements and the reason for their selection as well as the scenario for utilizing the research results in society.
3.2.7 The abstract should be 300 Japanese characters or less (125 English words). The Japanese abstract may be omitted in the English edition.
3.2.8 The main text should be about 9,000 Japanese characters (3,400 English words).
3.2.9 The article submitted should be accompanied by profiles of all authors, of about 200 Japanese characters (75 English words) for each author. The essential contribution of each author to the paper should also be included. Confirm that all persons who have made essential contributions to the paper are included.
3.2.10 Discussion with reviewers regarding the research paper content shall be done openly, and the Editorial Board will edit the highlights of the review process to about 3,000 Japanese characters (1,200 English words) or a maximum of 2 pages with the names of the reviewers disclosed. The edited discussion will be attached to the main body of the paper as part of the article. Regarding the reports and the commentaries, discussion with the Editorial Board members will be opened at the Board's discretion. In this case, the Editorial Board will edit the discussion to about 800 Japanese characters (less than half a page) with the names of the Board members disclosed.
3.2.11 If there are reprinted figures, graphs or citations from other papers, prior permission for citation must be obtained and should be clearly stated in the paper, and the sources should be listed in the reference list. A copy of the permission should be sent to the Publishing Secretariat. All verbatim quotations should be placed in quotation marks or marked clearly within the paper.

3.3 Format

3.3.1 The headings for chapters should be 1, 2, 3…, for subchapters, 1.1, 1.2, 1.3…, for sections, 1.1.1, 1.1.2, 1.1.3, for subsections, 1.1.1.1, 1.1.1.2, 1.1.1.3.
3.3.2 The chapters, subchapters, and sections should be enumerated. There should be one line space before each paragraph.
3.3.3 Figures, tables, and photographs should be enumerated. They should each have a title and an explanation (about 20–40 Japanese characters or 10–20 English words), and their positions in the text should be clearly indicated.
3.3.4 For figures, image files (resolution 350 dpi or higher) should be submitted. In principle, the final print will be in black and white.
3.3.5 For photographs, image files (resolution 350 dpi or higher) should be submitted. In principle, the final print will be in black and white.
3.3.6 References should be listed in order of citation in the main text.

Journal—[No.] Author(s): Title of article, Title of journal (italic), Volume(Issue), Starting page–Ending page (Year of publication).
Book—[No.] Author(s): Title of book (italic), Starting page–Ending page, Publisher, Place of Publication (Year of publication).
Website—[No.] Author(s) name (updating year): Title of web page, Name of website (may be omitted If the name of the website is the same as that of the author(s)), URL, Access date.

4 Submission

One printed copy or electronic file (Word file) of manuscript with a checklist attached should be submitted to the following address:

Synthesiology Editorial Board
c/o Public Relations Information Office, Planning Headquarters, National Institute of Advanced Industrial Science and Technology(AIST)
Tsukuba Central 1, 1-1-1 Umezono, Tsukuba 305-8560
E-mail: synthesiology-ml@aist.go.jp
The submitted article will not be returned.

5 Proofreading

Proofreading by author(s) of articles after typesetting is complete will be done once. In principle, only correction of printing errors is allowed in the proofreading stage.

6 Responsibility

The author(s) will be solely responsible for the content of the contributed article.

7 Copyright

The copyright of the articles published in "*Synthesiology*" and "*Synthesiology English edition*" shall belong to the National Institute of Advanced Industrial Science and Technology(AIST).

Inquiries:

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Letter from the editor

In this issue, we present four papers: “Development of a compact, onboard slurry icemaker to rapidly produce optimal ice for maintaining freshness of marine products,” “Standardization of dimethyl ether (DME) fuel specifications,” “A study on high-density recording with particulate tape media for data storage systems,” and “Development of a cell microarray chip system for early and accurate malaria diagnosis.” While the objectives of the papers are different, such as to deliver good quality fish to the market by maintaining freshness, to prevent global warming by promoting fossil fuel alternatives, to strengthen information infrastructure by increasing the recording density of data recording devices, or to protect the lives and health of people of developing countries by enabling early diagnosis of damaging infectious diseases, these are important studies that share the point of bringing about innovation to society through new technology. All papers describe how basic research is conducted to establish technological potential, how to fuse such technological potential to the peripheral technologies to achieve practical utilization, how to conduct verification tests, and how the results are fed back to R&D. They show the process of clarifying research scenarios, tackling R&D, and achieving social implementation.

Although science and technology play major roles in solving social problems, the R&D is not easy. In many cases, unforeseen circumstances are encountered, and much time and labor are spent in trial-and-error to break through the issues. The skill of a researcher is how to reduce the amount of trial-and-error, and it is useful to establish a clear research scenario and be mindful of such a scenario during the course of research.

In the papers published in this issue, the research is done in collaboration with other organizations, and along with how to conduct basic research, how to collaborate with which organization is considered in the research scenario. I hope you will read through the papers since useful information can be found for people who are involved in various types of R&D.

Hiroshi YOTSUMOTO, Senior Executive Editor

Aim of *Synthesiology*
— Utilizing the fruits of research for social prosperity —

There is a wide gap between scientific achievement and its utilization by society. The history of modern science is replete with results that have taken life-times to reach fruition. This disparity has been called the *valley of death*, or the *nightmare stage*. Bridging this difference requires scientists and engineers who understand the potential value to society of their achievements. Despite many previous attempts, a systematic dissemination of the links between scientific achievement and social wealth has not yet been realized.

The unique aim of the journal *Synthesiology* is its focus on the utilization of knowledge for the creation of social wealth, as distinct from the accumulated facts on which that wealth is engendered. Each published paper identifies and integrates component technologies that create value to society. The methods employed and the steps taken toward implementation are also presented.

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Highlights of the Papers in *Synthesiology*

Research papers

"Synthesiology-English edition" is a translated version of "Synthesiology," which is published quarterly, ISSN 1882-6229, by AIST. Papers or articles published in "Synthesiology-English edition" appear approximately four months after the publication of the original "Synthesiology."

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