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Synthesiology

English edition

Evaluation of earthquake occurrence from active faults

Two types of lead users in a model for the transfer of technology into households

Creating non-volatile electronics by spintronics technology

A marked improvement in the reliability of the measurement of trace moisture in gases

Development of battery-operated portable high-energy X-ray sources

2009

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MESSAGES FROM THE EDITORIAL BOARD

There has been a wide gap between science and society. The last three hundred years of the history of modern science indicates to us that many research results disappeared or took a long time to become useful to society. Due to the difficulties of bridging this gap, it has been recently called the valley of death or the nightmare stage (Note 1). Rather than passively waiting, therefore, researchers and engineers who understand the potential of the research should be active.

To bridge the gap, technology integration (i.e. *Type 2 Basic Research* – Note 2) of scientific findings for utilizing them in society, in addition to analytical research, has been one of the wheels of progress (i.e. *Full Research* – Note 3). Traditional journals, have been collecting much analytical type knowledge that is factual knowledge and establishing many scientific disciplines (i.e. *Type 1 Basic Research* – Note 4). Technology integration research activities, on the other hand, have been kept as personal know-how. They have not been formalized as universal knowledge of what ought to be done.

As there must be common theories, principles, and practices in the methodologies of technology integration, we regard it as basic research. This is the reason why we have decided to publish "*Synthesiology*", a new academic journal. *Synthesiology* is a coined word combining "synthesis" and "ology". Synthesis which has its origin in Greek means integration. Ology is a suffix attached to scientific disciplines.

Each paper in this journal will present scenarios selected for their societal value, identify elemental knowledge and/or technologies to be integrated, and describe the procedures and processes to achieve this goal. Through the publishing of papers in this journal, researchers and engineers can enhance the transformation of scientific outputs into the societal prosperity and make technical contributions to sustainable development. Efforts such as this will serve to increase the significance of research activities to society.

We look forward to your active contributions of papers on technology integration to the journal.

Addendum to Synthesiology-English edition,

"*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". The views expressed in translated version are exclusively those of the Japanese authors and editors. The Japanese authors are generally consulted regarding the translation of their papers, but are not responsible for the published English version.*

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Synthesiology Editorial Board

Note 1 : The period was named "nightmare stage" by Hiroyuki Yoshikawa, President of AIST, and historical scientist Joseph Hatvany. The "valley of death" was by Vernon Ehlers in 1998 when he was Vice Chairman of US Congress, Science and Technology Committee. Lewis Branscomb, Professor emeritus of Harvard University, called this gap as "Darwinian sea" where natural selection takes place.
Note 2 : *Type 2 Basic Research*
This is a research type where various known and new knowledge is combined and integrated in order to achieve the specific goal that has social value. It also includes research activities that develop common theories or principles in technology integration.
Note 3 : *Full Research*
This is a research type where the theme is placed within the scenario toward the future society, and where framework is developed in which researchers from wide range of research fields can participate in studying actual issues. This research is done continuously and concurrently from *Type 1 Basic Research* (Note 4) to *Product Realization Research* (Note 5), centered by *Type 2 Basic Research* (Note 2).
Note 4 : *Type 1 Basic Researc*h
This is an analytical research type where unknown phenomena are analyzed, by observation, experimentation, and theoretical calculation, to establish universal principles and theories.
Note 5 : *Product Realization Research*
This is a research where the results and knowledge from *Type 1 Basic Research* and *Type 2 Basic Research* are applied to embody use of a new technology in the society.

Synthesiology – English edition Vol.2 No.3 (Dec. 2009)

Contents

Evaluation of earthquake occurrence from active faults

— Evaluation of rupture probabilities of active faults using the Cascade Earthquake Model based on behavioral segmentation —

Toshikazu Yoshioka

[Translation from *Synthesiology*, Vol.2, No.3, p.194-200 (2009)]

In order to assess the probability of the occurrence of future large earthquakes based upon the past activities of active faults, we divided active faults into behavioral segments, and adopted the Cascade Earthquake Model, that is, a model that considers that an earthquake is sometimes caused by a single segment and sometimes caused by multiple segments. Using this model, we can evaluate the rupture probability of active faults by a uniform standard without any inconsistencies with field data. The result was published as the *Rupture Probability Map of Major Active Faults in Japan*.

Keywords : Active fault, earthquake, assessment, rupture probability, behavioral segment

1 Objective and background of the research

Japan is frequently struck by earthquakes and experiences damages almost every year. Particularly, the earthquakes that are caused by the rupture of active faults occur in the shallow part of the inland region and can cause major damages. Therefore, from the viewpoint of effective earthquake damage prevention, it is extremely important to be able to accurately predict the earthquakes that are caused by active faults. Recently, the survey of active faults has progressed throughout Japan and scores of data have been obtained. However, the predictions based on these data are not sufficient. We conducted this research because we believe that even if the information is not sufficiently accurate, the transmission of information based on the most rational and most uniform method at this point is extremely important, as a researcher and as a research institution whose mission is geological survey.

An "active fault" is a fault that has repeatedly ruptured in the past and possesses possibility of causing a major earthquake in the future. The interval of cyclic rupture is extremely long, normally from a thousand to several tens of thousands of years. Although the slip of a fault in one earthquake may be only a few meters, when the slippages are repeated in the same direction over several thousand or several million years, a displacement of several tens or hundreds of meters may accumulate. As a result, if the displacement is in the vertical direction, the upthrust side becomes a mountain range and the downthrust side becomes a plain or a basin. If the displacement is in a horizontal direction, valleys and peaks become bent. The study of active faults from such geomorphological perspective advanced rapidly in the 1960s to the first half of the 1970s. The background was the diffusion of the plate tectonics theory that states that the Japanese Archipelago is being compressed as it is pushed by the Pacific Plate. The landforms such as mountain ranges and basins of Japan were created by fault activities. The study of active faults started as an attempt to answer one of the main themes of geomorphology and geology: "Why is a mountain high?" This can be positioned as *Type 1 Basic Research*.

On the other hand, since great earthquakes are caused by the rupture of active faults, the study to predict future earthquake occurrences from the past rupture and their frequency started in the latter half of the 1970s. The method involves digging a trench across an active fault, and the past dates of fault ruptures are investigated from the slippage and the age of the geological layers (Fig. 1). This method is called trench survey, and such investigations were conducted in many places in the 1980s. With the diffusion of trench survey, various data on the past ruptures (past earthquakes) of active faults were accumulated, and they aggregated into a discipline called "paleoseismology." While conventional seismology concentrates on the observation of current earthquakes, paleoseismology offers possibility of predicting future occurrences of great earthquakes by studying the cycle of ruptures from geological history. In other words, the study of active faults evolved into its utilization by society, or *Type 2 Basic Research*.

The importance of active faults became widely known to the public in the Great Hanshin-Awaji Earthquake (Hyogoken-nanbu Earthquake) of 1995. The fact that this earthquake was caused by the rupture of active faults was widely publicized through the mass media, and the awareness for "active fault" rose sharply in society. Immediately after this earthquake, the Headquarters for Earthquake Research Promotion was

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established by the Japanese government, and the nationwide survey of active faults and the long-term assessment (probability prediction) of active faults were started. In an attempt unseen elsewhere in the world, the scale of the earthquakes that may occur in the next 30, 50, and 100 years and the probability of their occurrences were assessed for each major fault zone (98 fault zones at the time) in Japan, and the figures were publicized. This was one way for the *Product Realization* of the results of active fault research.

2 Relationship of active fault and earthquake scale

In predicting future earthquakes, it is important to predict the scale as well as the place and time of the occurrence. The size of an earthquake is proportional to the length of the active fault that ruptures during the earthquake and the amount of slip. Also, the amount of slip is proportional to the length of the active fault. Therefore, to predict the scale of the earthquake that may happen in the future, it is necessary to estimate the length of the active fault related to that earthquake.

Considering the length of an active fault, it is not easy to judge from what point to where should be considered one fault, since numerous active faults are distributed like a network throughout the Japanese Archipelago. Moreover, according to the surveys so far, there are several geological and geomorphological evidences that indicate that a slip of certain magnitude has been occurring repeatedly at the same interval from the past in a certain point of a certain active fault. However, according to the historical records of earthquakes and careful geological survey, the range of active faults that rupture during earthquakes that occur repeatedly in a region may be different for each earthquake, and the active faults may not necessary rupture within the same range.

Fig. 1 Example of trench investigation of an active fault (Nukumi Fault of the Nobi Fault zone, Ohno City, Fukui Prefecture).
This active fault is known to have ruptured in the Nobi Earthquake in 1891. The bump on the earth surface is the slip that occurred in 1891. Since the lower layer (older layer) show more pronounced horizontal displacement than the top layers, it can be seen that the slippage of the fault accumulated repeatedly.

In the Nobi Earthquake (magnitude 8.0) that occurred in 1891, the Nobi Fault System that crossed the Gifu and Fukui Prefectures ruptured, and a slip of about 6 m high occurred in Midori, Neo-mura (current Motosu City) located in the center of the fault zone. From the distribution form of the fault, it is determined that this fault zone is composed of the Nukumi Fault, Neodani Fault, Umehara Fault, and other smaller active faults (Fig. 2). It is recorded that during the earthquake of 1891, the western half of the Nukumi Fault, Neodani Fault, and Umehara Fault ruptured together and caused a slip in the earth surface[1]. However, looking at the past ruptures of this fault, it was found that the past rupture periods of Nukumi, Neodani, and Umehara faults differed[2][3]. This means that in certain periods in the past, the Nukumi and Neodani faults, and the Neodani and Umehara faults did not rupture in unison.

One problem arises. If the ranges of fault destruction when the earthquake occurs (i.e. length of the fault) are different, the amount of slip at a certain point should be different each time because the length and the amount of slip are proportional. Moreover, since the rate of an average slip of a fault (average rate of displacement) is thought to be constant over a long term, if the amount of slip differs every time, the rupture interval of the fault (interval of earthquake occurrence) must be different each time accordingly. However, as mentioned earlier, from the geomorphological and geological studies, it has been concluded that the amount of slip did not change greatly in the past except in certain points. If the amount of slip and the rupture interval of the faults differ for each earthquake, the cyclic nature of active fault rupture mentioned earlier will be negated and future prediction of earthquake occurrence becomes impossible.

The author and others looked at the Cascade Earthquake Model as a model that offers rational explanation without

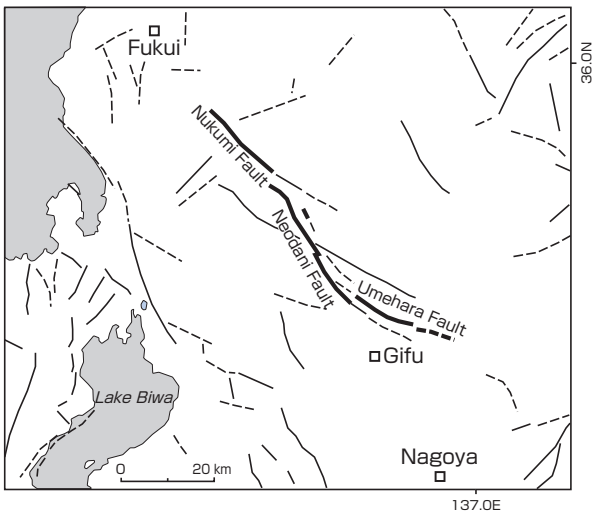

Fig. 2 The Noubi Fault zone and the distribution of surrounding active faults.
The heavy lines show the faults that ruptured in the Nobi Earthquake of 1891. Dashed lines are inferred faults.

inconsistencies. This model was employed for the active faults in California, U.S.A. as an earthquake model for a very long active fault running several hundred kilometers[4]. In this model, a continuous active fault is divided into several behavioral segments that have their own unique amount of slip and rupture intervals[5]. These segments rupture while maintaining their unique cycles and sometimes in conjunction with the neighboring segments.

The greatest characteristic of the Cascade Earthquake Model is that the length of the segment can be fixed regardless of the range value of fault rupture, by considering the large-scale fault rupture as a co-movement of the behavioral segments. By doing so, the earthquake time-varying position at a certain point can be kept constant while maintaining the proportional relationships of the fault length, the amount of displacement by earthquake, and the earthquake size. Therefore, the cyclic nature of fault rupture at a certain point can be explained extremely easily. We decided to employ the Cascade Earthquake Model as the most realistic model at this point, to achieve predictions utilizing the geomorphological and geological research results.

The schematic diagram of the Cascade Earthquake Model is shown in Fig. 3. Conventionally, active faults were given the name "X Fault" from their geographical distribution without any particular criterion. However, since the range that was given a name may not necessarily cause the next earthquake, it is impossible to estimate the earthquake scale from the length of that fault. Therefore, these active faults were categorized into behavioral segments (here four categories from A to D) as "units" that may cause earthquakes, with certain criteria such as past rupture history and distribution, regardless of the conventional fault names. By thinking that an earthquake occurs when these segments rupture individually or in conjunction with the neighboring segments, the size of the earthquake can be predicted from the length of the individual fault (behavioral segment) and its co-movement relationship with the neighboring segments.

To apply this model to the small-scale and complex faults of Japan, the data of earthquake faults with records of rupture were listed, and we prepared a trial plan for the relationship of earthquake scale and criterion for behavioral segment category[6]. Using the Kinki area as an example, the probability of future earthquake occurrence for each behavioral segment was calculated and publicized[7]. Although this calculation was only a result of one research, it was meaningful as an assessment obtained by logical deduction based on a model.

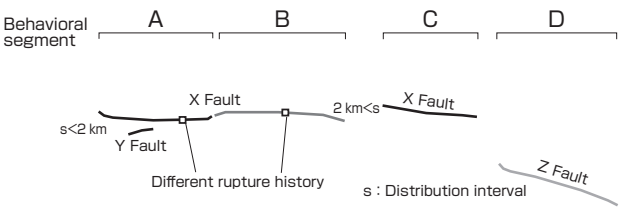

Fig. 3 Schematic diagram of the behavioral segment categories.

3 Publication of the *Rupture Probability Map of Major Active Faults in Japan*

To develop this research further and to actually make it useful in society to reduce the risk of earthquake damage, it was necessary to apply this model on a national scale. Therefore, the author and others started to compile the *Rupture Probability Map of Major Active Faults in Japan*[8].

The criteria for categorizing the behavioral segments are summarized in Fig. 4. The behavioral segments were categorized according to this criterion based on the distribution map of active faults. When covering all active faults in Japan, it was inevitable that some active faults were surveyed heavily and had plenty of data while others had hardly any specific data. In our map, we placed emphasis on obtaining uniform values for the entire country. Therefore, even in cases where no data was available, some assessment was obtained using provisional and empirical values rather than labeling them "unknown." By placing priority on national coverage, we aimed for the *Product Realization* in

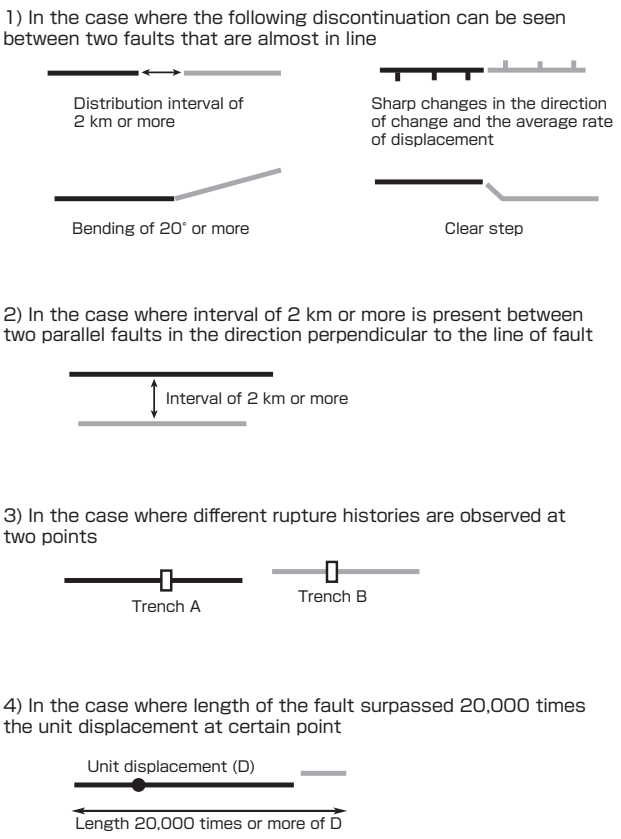

Fig. 4 Criteria for categorizing the behavioral segments used in the *Rupture Probability Map of Major Active Faults in Japan*.

this *Full Research*.

We assessed the active faults that were 20 km or more throughout Japan, and 547 behavioral segments were categorized. Of these, the parameters such as average displacement rate, amount of slip in an earthquake, and past rupture periods were estimated based on existing data for 295 behavioral segments that were 10 km or more long and had some degree of rupture level, and their future rupture probabilities were calculated. The behavioral segments grouped by color according to the rupture probability were plotted on the map of Japan (Fig. 5).

The *Rupture Probability Map of Major Active Faults in Japan* was published by the Geological Survey of Japan, AIST in September 2005. This map enabled comparison of the active faults under a uniform criterion for the entire country, although there were variations in confidence due to the fineness of data by region. As comparison became possible under a uniform criterion for the whole of Japan, it has been used for risk assessment by the insurance industry and by the regional infrastructure planners. By indicating the assessment criterion for individual behavioral segments, it could be used by users who need assessment based on different standards.

This assessment, of course, is a research result of a single research institution, and therefore is simply one prototype. It is distinctly different in character from the long-term assessment of the Headquarters for Earthquake Research Promotion of the government, which is based on an official standard determined by a committee. Therefore, the figures shown differ greatly in some places, and some people have voiced concerns that this may cause confusion in society. Also, since the results are based on estimated values using provisional and empirical values, many researchers (particularly those of geomorphology and geology) who place importance on individual data commented that faults with insufficient data should not be assessed.

However, considering the difficulty of assessing the actual earthquake occurrence, it is natural that various assessment results exist, and we thought it was important to present figures as one of the assessment results.

As a result, the *Rupture Probability Map of Major Active Faults in Japan* obtained a certain degree of acclaim from the insurance industry, and we received several inquiries about using this assessment as basic data. It can be said that it is having some social impact.

4 Limitation of prediction and future issues

Although the *Rupture Probability Map of Major Active*

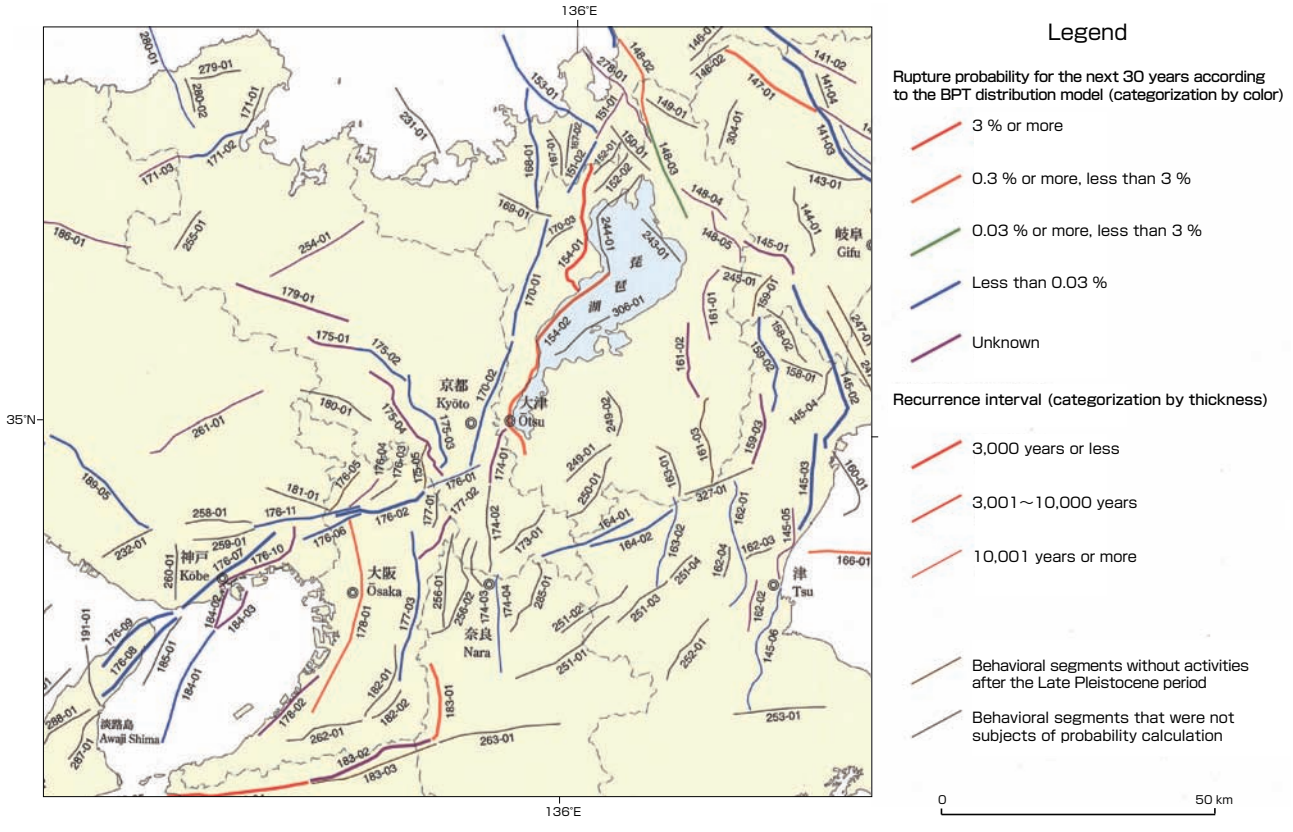

Fig. 5 *Rupture Probability Map of Major Active Faults in Japan*[8] (close up of the Kinki area).
The numbers show the behavioral segment numbers.

Faults in Japan was innovative as a nationwide assessment based on a uniform model, the assessment of co-movement among the behavioral segments was difficult, and we were unable to directly predict the scale of earthquakes that may occur in the future. Research is being continued in this aspect, but determining whether two behavioral segments ruptured at the same time in the past earthquake, or whether they ruptured with some time lag, are beyond the time resolution of geological survey. This is the limitation of this method. To solve this issue, another group in the Geological Survey is gathering the historical record of the earthquake that occurred recently and analyzing the relationship between the distribution form of faults and the propagation of ruptures using the numerical simulation of dynamic rupture[9].

Looking at some recent earthquakes that caused damage, there are cases where no major slip occurred in the active fault although the earthquake occurred along the active fault, as in the Mid-Niigata Prefecture Earthquake in 2004 (magnitude 6.8), as well as the case of Iwate-Miyagi Nairiku Earthquake in 2008, where no clear active fault was previously found although the earthquake of magnitude 7.2 occurred. Such earthquakes could not be predicted with the conventional assessment method.

Moreover, as a result of detailed survey of the active faults throughout Japan, there are data that show that the past rupture intervals of the active faults are not necessarily constant in some points. Although there are possibilities that there are flaws in the surveyed data or exceptions due to the uniqueness of the survey point, it will be necessary to review the model from the basic periodicity of the active fault ruptures.

However, the purpose may be defeated if the model becomes complicated in order to offer explanation to exceptional cases like the Iwate-Miyagi Nairiku Earthquake of 2008, and evaluation becomes impossible for the faults that do not have sufficient data for such a complicated model. Exceptions are inevitable in natural phenomena, and it is extremely important to discern what is an exception. Time range of several thousands and several tens of thousands of years is necessary to investigate the long-term prediction of earthquake occurrence, and the future issue is to build a model that is simple and universal because verification is virtually impossible.

5 Conclusion

The prediction of earthquakes that occur at active faults advanced dramatically in the past 10 years. However, we often feel the gap with what the society demands. For example, when an unpredicted earthquake occurs, society will not accept the explanation “this earthquake was unique and exceptional.” In that sense, the degree of achievement of this study is low. Another issue is that the Japanese society tends to demand an *osumitsuki* or official endorsement that may lead to blaming the government, and therefore the government tends to publicize only things that are absolutely certain.

The author feels that our social responsibility is to organize the intellectual foundation that can be used to prepare against earthquakes, by presenting the prediction of active fault ruptures as accurately and as comprehensively as possible to society. Although presenting the information with sufficient accuracy or complete comprehensiveness cannot be realized immediately, I believe that the publication of the results on a nationwide scale is significant as the first step. Whether this research methodology was effective or not will be evaluated by how the society responds.

Acknowledgements

This research is a continuation of the “Research on the Assessment for the Potential of Earthquake Occurrence by Observing Active Faults and others (FY 1994~1999),” a special research of the Agency of Industrial Science and Technology conducted at the Geological Survey, which was one of the predecessors of AIST, as well as earlier researches. The research evolved through repeated researches and discussions of numerous researchers during that time. I am grateful to all people involved, and shall emphasize that this research is the result of the collaboration with many people including my joint researcher Dr. Yasuo Awata.

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Discussion with Reviewers

1 The positioning as *Full Research* and descriptions in reference to "synthesiology"

Question and comment (Masaaki Mochimaru, Digital Human Research Center, AIST)

The positioning as *Full Research* is clearly stated in "1 Objective and background of the research." The active fault research as a study in geomorphology and geology is *Type 1 Basic Research*, while the following active fault research for earthquake prediction is *Type 2 Basic Research*. The categorization of the active faults into behavioral segments using the Cascade Earthquake Model provided a breakthrough, and investigations were done for the Kinki region. Up to this point can be considered as *Type 2 Basic Research*. The survey was conducted nationwide and the results were published and presented to society. This can be positioned as *Product Realization Research*.

However, as an academic journal, *Synthesiology*, we would like you to describe, with scientific eyes, the researcher's dream of "what kind of society you wish to realize," "why you selected" the methods that lead to practical application of basic research to achieve the dream, and whether the selected method was "effective or not" in achieving the goal. The aim of *Synthesiology* is to build the "science of synthesis" by collecting the case studies of *Full Research*. Rather than just being a collection of papers, it is important that the individual papers are written with "synthesiology" in mind.

Question and comment (Shigeko Togashi, Evaluation Division, AIST)

I recommend this paper for publication in *Synthesiology* because it practices *Full Research* by conducting rupture probability prediction as *Type 2 Basic Research* based on the geological research of active faults and by contributing to society through the publication of results that may mitigate earthquake hazards. However, there is a lack of enough explanation, which will be indicated in Discussion 2, and there are points that should be added from the *Synthesiology* perspective. Please give the definitions of the terms such as active fault, fault, and earthquake-generating fault that were mentioned in the paper.

Answer (Toshikazu Yoshioka)

I added the description on the significance of this research for preventing earthquake disaster in the introduction of the paper. Also, I added the author's thoughts in the conclusion as follows.

"The author feels that our social responsibility is to organize the intellectual foundation that can be used to prepare against earthquakes, by presenting the prediction of active fault ruptures as accurately and as comprehensively as possible to society. Although presenting the information with sufficient accuracy or complete comprehensiveness cannot be realized immediately, I believe that the publication of the results on a nationwide scale is significant as the first step. Whether this research methodology was effective or not will be evaluated by how the society responds."

The word "active fault" is used widely in general, and its definition may differ according to people and situations. Therefore, I added a clear definition. The terms like earthquake-generating faults were deleted to avoid confusion.

2 Selection of the research method and verification of its efficacy

Question and comment (Masaaki Mochimaru)

Please describe the selection of the research method and the verification of its efficacy as scientifically as possible. There are two points in this paper that are important in terms of synthesis. One is the application of Cascade Earthquake Model, and second is the nationwide survey and publication. For example, for the Cascade Earthquake Model, why did you consider using this model? Were there other models that could be used? Those are the important points in terms of synthesis. Of course, verification of the application of the Cascade Earthquake Model is also important.

Similarly, what were the reasons for selecting the method of nationwide survey and publication? Were there any other methods? What social effects did you expect by widening the area from Kinki to all of Japan? What was the effect? Were there any academic byproducts? Why did you focus on the data from a single research institute rather than obtaining consensus through a committee? What "merits" did that method bring?, and so on. It will also be good if you summarize how much of the goal (dream) you initially set was achieved, what were the limits, and how you would overcome them.

Question and comment (Shigeko Togashi)

Please describe the principle of the Cascade Earthquake Model simply, and explain specifically why you selected this model, how it was developed into a numerical simulation, and the relationship to other models.

For the limit of prediction, it is true that the accuracy of

prediction using other models had not met the demand of society, but in long-terms, the researches that address the mechanism of earthquakes are mandatory to increase the accuracy of prediction. Please state the relationship to these models objectively.

On the other hand, for the issue of contributing to society through the mitigation of earthquake hazards a simple and general model is very effective, even if it includes the case with low prediction reliability. The author has exemplified it by the publication of the *Rupture Probability Map of Major Active Faults in Japan*. The response of society should be described more clearly as a demonstration of its efficacy.

Answer (Toshikazu Yoshioka)

In the Cascade Earthquake Model, the unique regularity is not taken into account for the co-movement of multiple behavioral segments. They can act together completely randomly. However, we did investigate through numerical simulation assuming that there may be some factor in the co-movement. I added some explanation.

For the numerical simulation of dynamic rupture, the Active Fault Research Center investigated the co-movement of multiple fault rupture by dynamic rupture simulation, where various fault geometries were assumed. However, in this paper, the emphasis was placed on the use of the Cascade Model to explain the cyclic fault ruptures, and therefore, the description of dynamic rupture simulation was left to a citation of a reference.

The Cascade Model is one of the models for cyclic fault rupture, and this model does not show the rupture process of the seismic center. Therefore, it does not conflict with the dynamic rupture model that shows the fault distribution form and the propagation of rupture using a numerical simulation, the physical model of earthquake occurrence process, or other models based on the observation/analysis of stress around the active fault and the understanding of geological structure. The models can coexist. I added in the text that the research to increase the prediction accuracy should be continued separately.

On the response of society, I added the following description in the text: “As a result, the *Rupture Probability Map of Major Active Faults in Japan* obtained a certain degree of acclaim from the insurance industry, and we received several inquiries about using this assessment as basic data. It can be said that it is having some social impact.”

Two types of lead users in a model for the transfer of technology into households

— The development and diffusion of induction heating cookery —

Yuka Kubo * and Yasunori Baba

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In this paper, we propose a multi lead users model for the transfer of technology into households through an empirical analysis of the development and diffusion of induction heating (IH) cookery. Even if big firms successfully develop a technology such as IH, a new cooking paradigm based upon a special cooking device or special recipes is necessary when introducing a new technology to households. We postulate that two types of lead users play crucial roles in the task: reasoning-based lead users with technological expertise contribute to the development of functionality; sensitivity-based lead users having little expertise contribute to diffusion by making product socially trendy and authentically attractive. When introducing advanced technologies such as robots that have not been successfully diffused into households, forming flexible networks between the multi lead users and other stakeholders becomes highly indispensable.

Keywords : Advanced technology, lead user, technology diffusion, social value, induction heating cookery

1 Introduction

Today, there are many households that are introducing induction heating (IH) cooking devices and changing the heat source in the kitchen from gas to electricity. The development of IH devices was initiated by major electric appliances companies mainly for cooking stoves, in the beginning of the 1970s, and was put to practical use in the early 1990s. In the commercialized product, food is cooked by the heat of the cooking utensil placed on the stove, of which the heat is induced by electromagnetic induction in which the electricity is passed through the coil inside the stove. Initially, the price of IH was 350,000 yen, which was way too high for household diffusion, but the price dropped to less than 200,000 yen in the late 1990s. As of 2008, over 10 % of households use IH.

In analyzing the introduction of IH innovation, it is important to note that the benefit cannot be gotten with IH devices alone. It is necessary to prepare an entire system for cooking (here this will be called the cooking system), such as purchasing the cooking utensils that can be used in IH, learning the cooking skills, and developing the recipe. While the IH devices were developed actively by major corporations, in the early 1990s utensils that could be used with IH were inadequate, and the development of the components of the cooking system started only in the stage when IH began to diffuse into the households. Today, various proposals have been made for the IH cooking system, and the effort continues to design a new system where IH becomes the dominant component among all necessary components (dominant design[1]).

Traditionally, advanced technology was developed mainly by major corporations and diffused into the households through the market mechanism. However, when an advanced technology is introduced, difficult problems develop in many cases. Even if a heat source is developed, supplementary components such as utensils, skills, and recipes, as mentioned above, become necessary[2]. In addition to the development of IH by manufacturers, users who are highly interested in the development and diffusion of IH from various positions become "lead users[3]" and develop the supplementary technology to promote diffusion of the technology to households. This paper looks at this process, and surveys and analyzes how various lead users each played roles in the process of introducing the advanced technology called IH. First, the routes by which each component of the cooking system reached the household and how they spread as part of the IH cooking system are investigated by the time-series analysis of the media information on the change in the cooking method. Second, we looked at the cooking method in which "frying is done by holding the silicon spatulas in both hands" while the frying pan remains fixed. This is a method that diffused along with IH, and who developed it and how this method was developed are analyzed. Based on the above analyses, the paper proposes a model that emphasizes the roles of various lead users when advanced technology is introduced to the household. Finally, what is needed to maximize the technological potential from a social perspective when introducing advanced technology into households will be discussed.

2 Existing studies

Humans have developed various artifacts over the history to

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realize the functions needed for living. The society evaluated the produced artifacts, and selected those with excellent performance and price. In addition, certain lead users responded actively to an artifact, improved it from his/her perspective, and proposed the redesigned artifact to society[3]. There are series of researches in engineering, economics, and business administration on the evolution and mechanism of artifacts[4]-[8]. The following section describes the works by Petrosky[6] who investigated "why there are four prongs on a fork," and David[9] who analyzed "how the typewriter keyboard arrangement became QWERTY."

In the case of forks, the failure of the current products or the shape that made it difficult to use led to improvement. When cutting the food and transporting it to the mouth, people did so using their hands and teeth in prehistoric age, and a knife was used when it was invented. Later two knives were used. One knife was used to hold the food and transport it to the mouth while the other was used to cut the food. The knife for holding and transporting the food evolved into a spoon, a two-prong fork, a three-prong fork, and then the current four-prong fork, as the deficiencies of the previous products were improved. What consists as deficiency differs by person and by situation, and therefore no perfect product exists. However, the diffusion in society occurred through development of manners used when people gathered for banquets and through books on etiquette. The form of the fork evolved continuously as the process was repeated. The users played important roles in the evolution of the fork.

In case of the keyboard, the keyboard arrangement was influenced by users who typed and by schools that provided typing lessons, in addition to the manufacturers that led the development. The keyboard of the typewriter created for the first time in the late 1860s was arranged in alphabetical order ABCD from the left, and later, different manufacturers employed various arrangements. While various arrangements coexisted, users started to compete for typing speed, and contest for typing speed and typing schools that taught shorthand appeared. There, touch-typing where one memorized the keyboard and typed without looking became the standard. Since everyone protested against changes in the arrangement that they learned, ultimately, the current arrangement that was the most familiar to most people was employed. The keyboard arrangement was selected to benefit the user communities such as contests and schools, and external influence beyond the market mechanism acted in this selection process[Note 1)].

3 Framework of this survey

The IH cooking system analyzed in this paper is composed of diverse supplementary components for cooking using IH, as well as IH heat source. To investigate who developed them and how this IH cooking system was developed, the following surveys were conducted.

First, a database was built by gathering media information (books, cooking magazines, fashion magazines, and television commercial messages (TVCM)) on IH cooking, and the time-series change was analyzed[Note 2)]. Specifically, time-series analysis of the photographs of IH cooking (267 books, 57 cooking magazines, 52 fashion magazines, and 64 magazine ads) and TVCMs of IH cooking (31 cases), with subject limited to frying pan cooking, was conducted for how the new cooking method using IH was introduced by the media from 2002 to 2008[Note 3)]. The information for the media survey is listed in Appendix A.

Second, interview survey was conducted to investigate who proposed the method and how the cooking method that appeared in conjunction with the introduction of IH was developed, and for what reason it diffused. Specifically, people who contributed to the development and diffusion of the cooking method were extracted from the media information, and interviews were conducted from May to November 2008. The information for the interviews is listed in Appendix B.

4 Time-series analysis of the IH cooking method transmitted through the media

In the IH cooking transmitted through the media, the method in which frying is done by holding the silicon resin spatulas in both hands, as shown in Fig. 1 photograph, was introduced for the first time in 2004. The novelty of this cooking method was as follows. First, the silicon resin spatula was used rather than a ladle, a wooden spatula, cooking chopsticks, or a turner that were traditionally used for cooking over heat. The silicon spatula is a silicon version of the rubber spatula that had been in use. It has been imported and sold in Japan since 1999. However, at the time, the awareness of silicon spatula in Japan was extremely low. Rubber spatulas could not be used for cooking over heat, and it was mainly used for confectionery

Fig. 1 Cooking method in which silicon spatulas are held in both hands.

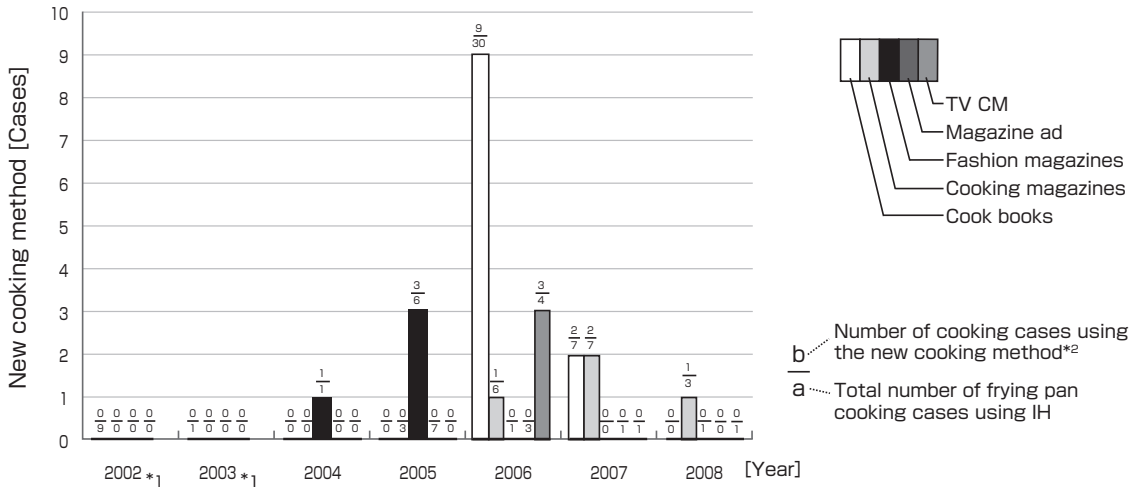
cooking. The silicon spatula could be used for cooking over heat since it is heat resistant to about 300 ºC. In IH cooking, the bottom of the pan becomes hot and frying takes place in the deep part of the pan, and spatulas must be used to scrape off the ingredients that adhere to the pan. Second, the method of using both hands to maneuver the cooking utensils was introduced as a new cooking method. When using gas, the pan had to be agitated so the ingredients would come in contact with the hot surface of the pan. In contrast, the bottom of the pan becomes hot in IH. There is no need to agitate the pan, and the tools are used to lift up and mix the ingredients. In IH cooking, the pan does not become hot if it is removed from the heat source, so it must be fixed and one does not touch the pan while cooking.

As shown in Fig. 2, the new cooking method was introduced in a fashion magazine in 2004 by cookery researcher W who studied the IH cooking method. The cooking method using silicon spatula was introduced for the first time in 2006 in books and cooking magazines, and there was increased exposure in the cooking magazines. Overall, the percentage of new cooking methods including two wooden spatulas and one silicon spatula, which were precursors of the two silicon spatula method, was about 30 percent and not overwhelming, but it was sufficient to gain social recognition. In contrast, the manufacturer's magazine ads showed strong intentions of providing correct technical information for IH against inappropriate ways of cooking, and there was no introduction of a new cooking method[Note 4)]. The information of the new cooking method was widely transmitted to prospective IH users mainly through fashion magazines where information flowed from lead users including cookery researchers, rather than companies manufacturing the IH cooking heater. For TVCMs, the all electrification sales promotion series by the Tokyo Electric Power Company (TEPCO) started in October 2004, and one of the versions aired in January 2006 featured a scene of mixing the ingredients with two wooden spatulas.

5 Development and diffusion of the cooking method in which frying is done by holding silicon spatulas in both hands

The result of the interview survey on the development of the cooking method is summarized as follows. First, the electric power company played a major role in the development of the cooking method in which silicon spatulas are held in both hands. TEPCO started cooking demonstrations by famous chefs at a cooking class held at the Ginza TEPCO Building in 1999. Measurements and recordings of the cooking were done to create the IH recipes, in hope that the professional chefs would present specialized cooking methods for the new heater. A cooking school coordinator N observed that chef Y of a famous Chinese restaurant held the wooden spatulas in both hands when he made fried rice. This frying method was positioned as the new "IH cooking method" to counter the claim that IH was unsuitable for Chinese cooking because one could not "agitate" the pan as the pan must be lifted from the heater to do so. In the TEPCO's TVCM aired on January 2006, there was a scene showing a boy making fried rice in an IH cooking school and he used two wooden spatulas to stir.

Second, a cookery researcher who studied IH cooking also played a major role. Cookery researcher W replaced the stove in her cooking school to IH in 2000, and decided to study IH cooking. She frequently exchanged information with aforementioned N about how to make fried rice with IH, and learned about the cooking method using two wooden spatulas. W immediately realized the problem where the rice adhered to the wooden spatula in this cooking method, and after experimenting to solve this issue, she found that the method of holding the silicon



*1 For TVCM, years 2002~2003 were not surveyed.
*2 New cooking methods include use of two wooden spatulas and one silicon spatula, that were used before arriving at the two silicon spatula cooking method.

Fig. 2 Changes in the new IH cooking method transmitted by the media.

spatulas in both hands worked optimally[Note 5)]. At the time, there was no cooking method using spatulas made of synthetic material for cooking over heat. However, W knew that the silicon spatula supplied as a sample by importer K was heat resistant, and she tested the silicon spatula instead of the wooden spatula and turner[Note 6)]. However, W was not satisfied with the existing product, and continued to study the silicon spatula that had optimal structure for IH cooking. As a result, the product developed jointly by Kai Corporation, a manufacturer of kitchen utensils, was announced in 2008 and was launched to the general market in April 2009.

Third, businesses that did not have knowledge of IH technology or cooking method played important roles. TEPCO promoted the "Switch the Design" Project under the concept to "propose a new lifestyle through electricity," and planned a "select shop" for IH products in the Ginza TEPCO Building. As a result of selecting a company while incorporating ideas from housewives, Flying Saucer (a division of Toko K.K.) opened a store on October 2005[Note 7)]. The company appointed S, a housewife, as the store manager and developed original cooking utensils as well as sold existing products. Food stylist C, a friend of S, proposed new ideas. Flying Saucer was already working on the development of silicon resin spatulas in 2004, and it started the seven color line-up adding black, navy, green, and brown to the red, yellow, and blue products that were already in the stores.

Next, the diffusion process by which the cooking method using the silicon spatulas diffused in society will be described. First, from the experience of developer W, her cooking method was initially not intended for the general public. The cooking method with silicon spatulas held in both hands was publicized in an article for IH fry cooking in a cooking article in a fashion magazine in 2004, as mentioned before. In the article on fry cooking in a cookbook for IH published by W in 2006, the method of holding a wooden spatula in one hand and a silicon one in the other was shown. Although two silicon spatulas were optimal, one was replaced with a wooden spatula for readers who did not own two silicon spatulas[Note 8)]. The silicon spatula for IH cooking developed by W was announced through the media for the first time in December 2008.

Second, Flying Saucer actively promoted the silicon resin spatula through the media since its launch. Rather than the cooking articles that were the subject of the time-series analysis of this research, the silicon spatulas were taken up in the goods articles of fashion magazines without much relevance to IH cooking. The focus was on the seven-color line up, and they were introduced as "colors that can be used by men in cooking" or "colors that match the interior decoration of the kitchen to create a comfortable place." The company developed an original Chinese wok for IH cooking in collaboration with TEPCO, and conducted a sales promotion by showing the wok with the silicon spatula. At the time, the silicon spatulas were not intended for IH cooking, and the combination of the spatula and the IH cooking utensil was quite coincidental. The reason the company thought of supplementing IH cooking with a silicon resin spatula was because it saw the scene where two wooden spatulas were used in the TEPCO TVCM that was aired at the time. A red silicon spatula was displayed with the wok in the show window of the Ginza store, and the booklet for the store carried a photograph of the ingredients being mixed in the wok with navy and brown spatulas in both hands. On the TEPCO's web CM, a famous chef was shown making fried rice using the wok and red silicon spatulas[Note 9)].

6 Proposal of a model for development and diffusion of the cooking system accompanying the IH technology

The progression of the cooking system accompanying IH can be divided into two stages of development and diffusion, as shown in Fig. 3. First, the cooking method where the wooden spatulas are held in both hands appeared almost spontaneously from 1999 to 2000. In the process of using the wooden spatulas, the silicon spatulas started to be used as new cooking tools. The second stage was after 2004, and the information on the new cooking system was transmitted through the media as the diffusion of IH progressed. Looking at the individuals and organizations that contributed to the development and diffusion of the cooking system, the development was led by the power company that promoted the diffusion of IH, and main roles were played by professional chef Y who cooked intuitively with wooden spatulas in both hands, cooking class coordinator N who saw that the two-spatula cooking method was unique to IH, and cookery researcher W who used silicon spatulas instead of wood. The diffusion was lead by the cookery researcher who provided IH cooking education with support from the TEPCO's sales promotion efforts for all electrification. Also, the sales promotion activities of the retailers as exemplified by Flying Saucer were transmitted to households through various media.

To consider the development and diffusion of the new cooking system, Table 1 compares the activities themselves, knowledge produced and transferred and the results of the activities with existing research for the individuals and organizations that led the development and diffusion. While the evolution of the fork was a cumulative process over a long time, the change in the IH cooking system was a discontinuous evolution brought on by advanced science and technology. As in the case of the fork, the household played a certain role as a user in the evolution of IH. Similar to the case of the keyboard, cooking tools and recipes that comprise the cooking system could be developed outside of the

company. As in the keyboard arrangement, external factors such as preferences of some users affected the development in addition to the market mechanism.

From the above investigation, the characteristics of the development and diffusion of the cooking method will be discussed. First, the development and diffusion of the cooking method that supplements IH innovation were not carried out by the major electric appliance company that was responsible for the technological development of IH. The advertisement by the electrical appliance company emphasized the point that the disadvantages of IH could be solved for consumers who were skeptical about IH, and there was no suggestion of a new cooking method utilizing the advantages of IH. The individuals who contributed to the development and diffusion were independent lead users who were not directly related to the power company or the appliance companies. They were not satisfied in simply accepting the existing cooking method like the general users, and actively offered suggestions for existing cooking utensils and methods. They were then supported by the power company, transmitted their suggestions to the household through the media such as books and magazines, and worked actively to develop and diffuse the cooking method.

Next, the motivations for the two types of lead users are considered. For lead users who developed the cooking method, the motivation was the intellectual curiosity for the IH function and the quest for a practical cooking style. They were interested in the fact that the values of electricity entered by the user and the temperature change in the pan were reproducible, and that the temperature in the pan could be maintained constant. In the course of experimenting with these functions in cooking, they found a new cooking method that took advantage of the property where the bottom of the pan became hot. The developer became curious about the IH technology itself, and approached IH from the perspective of "scientific cooking." This motivation led to the formation of an intelligent social infrastructure that stepped beyond profitability[Note 10]. The new cooking method was transmitted through the cooking articles in fashion magazines, but this was four years after the cooking method was discovered.

Table 1 Mechanism of IH innovation compared with the evolution of precursory artifacts.

	Petrosky Model [Petrosky 1992]	David Model [David,1985]	Cooking system accompanying IH technology
Example	There are four prongs in a fork	QWERTY keyboard arrangement	Cooking method in which frying is done by holding the silicon resin spatulas in both hands
Leader of development	Lead user	Manufacturer	Logical lead user
Leader of diffusion	User	User community	Sensible lead user
Driver of development	Improvement of defects in existing products	Realization of higher performance	Development of new cooking utilizing IH function
Driver of diffusion	Formation of manners (harmony with society)	Competition among users for typing ability	Match with social trend
Media environment that promotes diffusion	Books on etiquette, banquets	Typing contests, typing schools	Magazines, select shop (in the building of electric company), TVCM, Internet
Performance of diffused artifact	Certainly better than before	Not necessarily high	Provision of new social values

Looking at the motivation of the lead users who led the diffusion, the select shop exercised a sensible intuition for social trends. The seven-color silicon spatulas that they intuitively developed were highly acclaimed in the goods articles of fashion magazines in a manner that had nothing to do with cooking, such as "colors that can be used by men" or "colors that match the color of the kitchen." Such lead users were independent from the power company or the appliance companies. The fact that the period from 2004 to 2006 when the cooking method was transmitted to households through

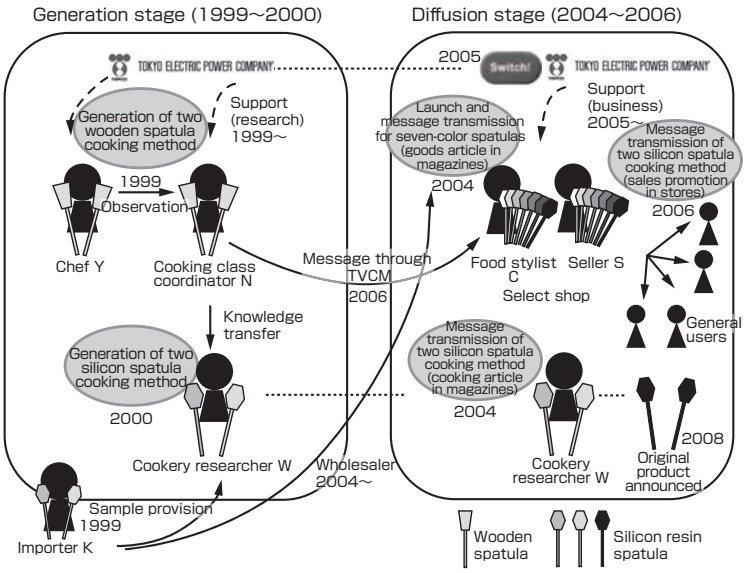


Fig. 3 Development and diffusion of cooking method in which frying is done by holding the silicon resin spatulas in both hands.

the media overlapped with the period of IH promotion by the power company was not by coincident. In fact, the select shop that served as a typical diffusion leader shared interest with the power company that was promoting all electrification. With such a background, the new cooking method was strongly pushed to households through the window of the store in the shopping district of Ginza as well as through the Internet.

From the above analysis, this paper proposes a model of the introduction of advanced technology to households through various lead users, as shown in Fig. 4. When introducing IH that was developed by a major electrical appliance company to households, the cooking method accompanying the IH played a major role. The development of the cooking method that fully brought out the technical potential of IH was done by "logical (reasoning-based) lead users" such as cookery researcher W who had specialized knowledge in the technology and cooking, and their goal was to maximize the function of IH for cooking. On the other hand, the diffusion of the new cooking method was promoted by the "sensible (sensitivity-based) lead users" such as S and C of the select shop. These users have excellent perception of the social trend. The simple combination of the advanced technology from the manufacturer and the cooking system that seek IH functionality may not necessarily result in a product that appeals to households, and may not lead to diffusion to homes. On the other hand, the cooking system proposed by sensible lead users who may lack the understanding of technology may produce attractive products that incorporate the social trend, and thereby promote the diffusion of the cooking system to households through the media. The flexible collaboration of diverse lead users contributed to the advancement of IH innovation.

7 Discussion

To introduce advanced technology to households, it is necessary to enable general users who do not understand the technology to appropriately use the product. As indicated by this study, the user cannot make fries only with the IH cooking heater developed by a company. A new cooking method must be developed by the logical lead user, and only when the supplementary components for IH cooking become available do the general users use IH at home. Moreover, the research indicates the possibility that the use of IH may spread to people who are not necessarily interested in cooking, as the users may be attracted to the element of the cooking system that reflects the social trend, such as the "color variations of the spatulas." How can we evaluate the fact that IH continues to diffuse even though the user is not conscious of the potential of the advanced technology?

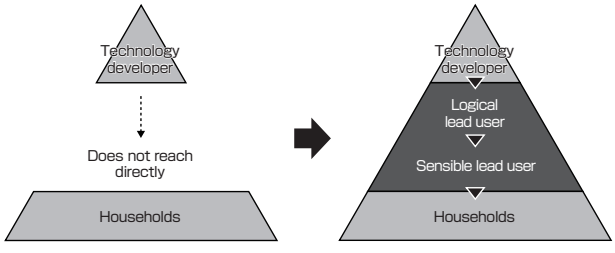


Fig. 4 Model of introduction of advanced technology to household through various lead users.

First of all, it is not easy to disseminate advanced technology to households. As shown by the case of household robots, even if advanced technology realizes wonderful functions, excellent performance alone will not ensure the introduction of a product to the households. For the potential product to be introduced to homes, it is necessary to organize the supplementary system for using the technology in households. As investigated in this paper, there are extremely diverse issues to be considered for a system for "using" advanced technology, such as how IH should be used in households, the safety of the IH cooking system, and environmental assessment (energy efficiency). The design-inspired innovation model that focuses on the product development by Apple Inc. states that the success of a product depends on the design that brings pleasure to the user[10], and the viewpoint of the user is an element that cannot be neglected. The supplementary system for using the technology at home must be designed to maximize the benefit the household user can receive in the context of social trends at that time.

The manufacturers, mainly of major corporations, develop the advanced technology itself. However, there is a limit in specialists designing the system for "using" the advanced technology. As Yoshikawa *et al* point out, even if the specialists mobilize all their knowledge of thermodynamics, surface science, and rheology pertaining to eggs, they may not necessary be able to produce good fried eggs[11]. In the case of the IH cooking system that is the subject of this research, the cooking system for "using" IH was created by fusing the cooking know-hows with advanced technology. This was accomplished by cooking specialists who sought "scientific cooking" independently from the corporations, and used their kitchens as laboratories. This indicates that by combining cooking practitioners with engineers, it may be possible to make a recipe for making good fried eggs. Also, it is important that the cooking specialist be an independent entity from the corporation. Pressure cookers and waterless cookers are now being used in general households after voluminous information had been supplied by cooking specialists. Therefore, for the advanced technology to be used in households, the autonomous participation of various lead users who specialize in "using" the technology is necessary.

Today, the importance of industry-academia collaboration that links science-technology and industry is widely

recognized. However, from the analysis of this research, to introduce advanced technology such as robots to the household, it can be seen that a social network composed of diverse lead users is needed in addition to academia and industry in order "to use" the technology. The model proposed in this research indicates the possibility that the household consumers can participate flexibly in the industrial activity using their practical knowledge. The model is expected to provide new research topics from the perspective of how to promote social innovation.

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Notes

Note 1) Recently, a powerful argument has been proposed that for the determination of keyboard arrangement, a corporate strategy to prevent entry of newcomers by the formation of a patent barrier instigated by a trust of major manufacturers played a critical role[12].

Note 2) The media information was determined as follows. For books, the books on cooking using IH cooking heater was selected visually among the books that contain "IH" in the title from the National Diet Bibliographical Database. For magazines, magazines that ranked within 5th place for circulation and were for readers 30 years old or above among "women's magazine" and "cooking and nutrition" categories were selected from the *Periodicals in Print in Japan 2008*. Magazines that contain articles on cooking using IH cooking heaters were selected visually for years 2005 and 2006 when there were highest amount of information on the IH cooking heater. For television CMs, all electrification TVCMs of TEPCO and Kansai Electric Power Company were selected.

Note 3) Even if there were several photographs, they were considered to be one data if they were for the same dish; even if it was one photograph or television CM, they were considered different data if there were different dishes. For magazines, advertisement and the text were separated; since the advertisements for cooking and fashion magazines were similar, they were jointly categorized as "advertisement."

Note 4) The IH cooking in advertisements were often for grill cooking, unlike in the books and magazines. Since IH could not be used with a grill, initially there were negative comments such as "you can't bake fish with the IH cooking heater." The grill on the IH cooking heater employs electrically heated wire rather than IH. Since the advertisements did not introduce any cooking method using new cooking tools, the figure in the graph is 0.

Note 5) W conducted experiments for a cooking method where mixing is done using two wooden spatula in her own kitchen, and observed that the rice grains got caught between the two spatulas, were squashed, and clumped together. To improve this point, she used two turners instead of wooden spatulas. However, the top edge of the turner was not perpendicular to the handle, and when held in both hands, the tips did not come together but crossed, and therefore the ingredients could not be held. She used the silicon resin spatulas instead of turners and found that mixing can be done with no problem.

Note 6) The import and sales of silicon spatulas were started for the first time in Japan by importer K who knew the high performance of silicon resin from his experience as a vacuum technology engineer. The product did not sell at all for the first three to four years, but the demand for the product rose from about 2004, and the sales for that year increased 2.7 times compared to the previous year.

Note 7) A "select shop" is a retail store that sells products of various brands selected by the sales clerk. Flying Saucer was a wholesaler of cooking utensils for professional use, but started handling products for household use in March 30, 2001, and S who was a housewife became the store manager.

Note 8) Developer W sought the silicon spatula form that was optimal for frying pan cooking using IH. The spatula designed and developed in collaboration with Kai Corporation was launched in December 2008.

Note 9) The TEPCO web contents released October 2006 include the photograph of famous Chinese chef, Mr. Ken'ichi Ching, stirring fried rice holding the original spatulas from Flying Saucer in both hands. (http://www.tepco-switch.com/others/ad/index-j.html)

Note 10) In IH, the pan itself becomes hot when the electricity is passed through the coil inside the stove, and the ingredients inside the pan are heated. Ignoring the variation of the resistance value of the metal of the pan, the power input and the heat value output of the pan will maintain a constant relationship. Also, ignoring the variation of the

quality of the ingredient, the power input and change of the ingredient will also maintain a certain relationship. In case of gas, the factors that determine the temperature inside the pan are complex[13]-[15].

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[15] K. Sugiyama *et al:* On cooking heat source – Choriyo netsugen ni tsuite gasu konro to IH hita no hikaku (kyozai kenkyu) (Comparison of gas stove and IH heater (study of educational material)), *Nihon Chori Kagakukai (Journal of Japan Society of Cookery Science),* 40 (2), 109-112 (2007) (in Japanese).

Authors

Yuka Kubo

Graduated from the Department of System Design Engineering, Keio University in 2000. Completed courses in Environmental Studies, Graduate School of Frontier Sciences, The University of Tokyo in 2002. Completed doctorate course in Human Environmental Studies, Graduate School of Frontier Sciences, The University of Tokyo in 2006. Doctor (Environmental Studies). Assigned to Assistant Professor of the Research Center of Advanced Science and Technology, The University of Tokyo in 2007. Currently studying innovations in advanced technology used in households. For this paper, worked on interview survey, media survey and analysis, and model proposal.

Yasunori Baba

Graduated from the Department of Economics, The University of Tokyo in 1977. Received Ph.D. from the University of Sussex in 1986. After working as SPRU Fellow and Senior Researcher of the National Institute of Science and Technology Policy, became Assistant Professor of the Research into Artifacts, Center for Engineering (RACE), The University of Tokyo in April 1993. Professor of the RACE in 1997. Professor of the Research Center of Advanced Science and Technology, The University of Tokyo since July 2001. Also, Professor of the Department of Advanced Interdisciplinary Studies, Graduate School of Engineering, The University of Tokyo from April 2007. In this paper, was in charge of model proposal and overall integration.

Appendix A

The surveyed books are listed in order of title, author, publisher, and publication date.

- *Seikatsu Jitsuyo Shirizu Benri Kino O Furu Katsuyo! IH Kukkingu Hita Kukkingu*; NHK Publishing; 2002.12.20.
- *Seikatsu Jitsuyo Shirizu IH Kukkingu Hita De Tsukuru Wagaya No Gochiso Reshipi*; NHK Publishing; 2003.11.20.
- *IH Reshipi Shu Katei De Dekiru Kobe Chuka Ryoriya-san No Aji*; Hyogo Prefecture Chinese Cooking Association, Hyogo Nutrition Cooking Confectionery School, and Kobe Branch of Kansai Electric Power Company; Asahiya Publishing; 2006.3.21.
- *IH Kukkingu Masuta Reshipi*; Masayo Waki; Kodansha; 2006.3.31.
- *Itsudemo Daredemo Oishiku Tsukureru! IH Kukkingu Hita Manten Reshipi*; Junko Takagi; Fuyosha; 2006.9.10.
- *IH Kukkingu Kihon No Reshipi*; Masayo Waki; Kodansha; 2007.12.1.

The surveyed magazines are listed in the order of magazine title, publisher, and publication period.

- *Okazu No Kukkingu*; TV Asahi Shuppan; Jan./Feb., 2000 ~ Oct./Nov. 2008.
- *Mrs*; Bunka Publishing; Jan. 2000 ~ Dec. 2008.

The surveyed television commercial messages are listed in order of advertiser, product name, and on-air period.

- Tokyo Electric Power Company; All Denka; 2005~2008.
- Kansai Electric Power Company; All Denka; 2005~2008.

Appendix B

The interviewed subjects are listed in order of name, company, and interview date.

- Yoshiki Ogino; All Electrification Business Promotion Division, Home Appliance Company, Panasonic Corporation; October 22, 2008.
- Hiroshi Kondo; Group Manager, Sales Planning Group, IH Cooking Heater Business Unit, Home Appliance Company, Panasonic Corporation; November 13, 2008.
- Izumi Hirota; Group Manager, Technical Group, IH Cooking Heater Business Unit, Home Appliance Company, Panasonic Corporation; November 13, 2008.
- Masaru Kita; Communication Group Manager, Sales Division, Sales Headquarter, Tokyo Electric Power Company; May 9, 2008.
- Ken'ich Morijiri; Director, Life Energy Design Center, Sales Division, Sales Headquarter, Tokyo Electric Power Company; May 11, 2008.
- Tomohiro Mizutani; Life Energy Design Center, Sales Division, Sales Headquarter, Tokyo Electric Power Company; July 15, 2008.
- Nobuko Sekikawa; IH Cooking Coordinator, Y.K. SNS Farm; July 15, 2008.
- Masayo Waki; Cookery Researcher, K.K. Trois Soeurs; June 27, 2008.
- Shuji Kato; President, K.K. Trois Soeurs; July 9, 2008.
- Hiromi Kawanishi; Y.K. Kawanishi; July 12, 2008.
- Miki Shimizu; President, Flying Saucer, Toko K.K.; May 1, 2008.
- Chizuko Shimizu; Director and Store Manager, Flying Saucer, Toko K.K.; May 19, 2008.
- Mieko Chiba; Food Stylist, Flying Saucer, Toko K.K.; July 14, 2008.

Discussion with Reviewers

1 Evidence for the discussion

Question and Comment (Motoyuki Akamatsu, Institute for Human Science and Biomedical Engineering, AIST)

The points that supplementary components is necessary for utilizing the technology, and that persons in certain positions act effectively to introduce advanced technology to households, are important for introducing the technology to society, and thus they match the scope of *Synthesiology*. However, the main thesis of the paper is somewhat unclear, and I feel there is not enough evidence.

Question and comment (Koh Naito, Center for Service Engineering, AIST)

Particularly in chapter "4 Time-series analysis…," please create a specific diffusion curve from the sales data, and draw a figure that shows how it is related to the major events.

Answer (Yuka Kubo)

I am aware of the insufficiency of evidence. I attempted to obtain cooperation from the appliance manufacturers and the cooking tool manufacturers to gather data for the sales of silicon resin spatula as well as for the actual state of IH cooking, but was unable to obtain those data. The sales data of the select shop were also undisclosed. Therefore, I conducted the analysis of household users from the information transmitted by the media. There was a huge variety of magazines, and while feeling regretful that a comprehensive analysis could not be accomplished, extraction was attempted as appropriately as possible.

This paper clarified that the cooking method unique to IH contributed to the introduction of IH to the household, and while the cooking method was diffused by sensible lead users, it was discovered by logical lead users. The logical lead users were able to discover a new cooking method because, while they were users, they were interested in the technology itself. I added revisions to express this point.

2 Sensible lead user

Question and comment (Motoyuki Akamatsu)

You mention that the cookery researcher is the logical lead user, while the select shop is the sensible lead user. The argument is clear for the cookery researcher, while it is unclear for the select shop. You wrote that the select shop is independent from the power company, but I don't think it is independent since it is located inside the TEPCO Building. Also you mention that the activity of the select shop is to respond to the social demand, but I don't think "participation of men in cooking" and "life surrounded by favorite cooking utensils" are general social demands. Also, it is written that "combining IH and silicon spatula was almost coincidental," and this means that the TEPCO CM played a significant role. From this point, didn't the power company function as a core diffuser?

Answer (Yuka Kubo)

As you commented, whether the action of the sensible lead user is a response to social demand is not clear in this analysis. It can be said that since the products born from the intuitive idea of the sensible lead user have been taken up several times in fashion magazine articles, they match the social trend as a result. I changed the description by using the word "social trend."

The select shop, which is a sensible lead user, has a store in the Ginza TEPCO Building through support of TEPCO, but TEPCO does not cooperate in development or selection of commercial goods. Although the TEPCO CM did show the cooking tools in both hands, it showed wooden spatulas, and I think it is important to note that replacing wooden spatulas with silicon ones was not an ordinary idea.

However, I do not think that it is coincidental that TEPCO supported a sensible lead user. In the process of setting up a select shop in the Ginza TEPCO Building, collaboration was done to find ideas that corporations cannot produce and to incorporate ideas from housewives. TEPCO did this intentionally to some degree. I added the point that housewives' comments were incorporated.

3 Contribution of women

Question and comment (Motoyuki Akamatsu)

The important point of the original manuscript is the "contribution by women," but in the framework of "logical lead users" and "sensible lead users," I don't think it is mandatory that the lead users are women. Of course, the two lead users were women in this case, but I think this was because the subject was cooking utensils whose primary users are women.

Question and comment (Koh Naito)

The paper discusses the importance of the role of sensible lead users who are sensible to the social demands of women. I think

this is an important point. However, there are descriptions such as "women as independent individuals" and "women who are in charge of cooking in homes," but I don't think the importance of the role of "women" is clear in the data and analysis of this paper.

Answer (Yuka Kubo)

One of the reasons the logical and sensible lead users contribute to the diffusion of technology from their positions is because the general users do not understand the content of the technology. I pointed out the contribution of women lead users because there are many technologies whose users are women who may not understand the technology. However, the roles of logical and sensible lead users who contributed to the introduction of IH to households can be played by men in non-IH cases. I removed the references that are limited to "women," and shall continue this research to demonstrate that the model applies to cases of other technologies.

4 Examples of other products

Question and comment (Motoyuki Akamatsu)

I think it will be helpful to readers if there are comparisons or discussion with other products. For example, pressure cooker may be used as another example for cooking utensils.

Answer (Yuka Kubo)

Unlike a microwave oven and a rice cooker that can be used by a user who has no skill, recipe, or tool, the user cannot benefit from an IH product alone. Similarly, for products that require supplementary components such as user's skill, recipe, and tools, various lead users are expected to contribute in diffusion, as indicated in this paper. For example, pressure cookers and waterless cookers were developed a long time ago, but information on cooking methods and recipes that optimize their functions are being transmitted by cookery researchers to this day. Recently, their users are increasing because there is a rising interest in reduced-oil cooking, and I think this is being accomplished through the contribution of lead users. I added the example of pressure cookers and waterless cookers in the discussion, and shall continue investigation of other case studies.

5 Perspective of innovation

Question and comment (Koh Naito)

On "IH innovation," in positioning it as an innovation, please describe which social value IH created when the technology appeared. I imagine that the appearance of the new cooking method affected the diffusion of IH because IH provided some new value to the user and it was not merely a technological replacement of gas.

Answer (Yuka Kubo)

The purpose of the generation and diffusion of the IH cooking system was to diffuse the value of "cooking that couldn't be done before" to society, as they were discovered when "used" by logical lead users. However, it was found that people who were not interested in cooking before or those who were simply interested in design were motivated to engage in cooking, powered by the intuitive idea of sensible lead users who contributed to the diffusion. Also, the social value of the IH cooking system is diverse including safety and environmental concerns. I added these to chapter 7.

Creating non-volatile electronics by spintronics technology

— Toward developing ultimate green IT devices —

Shinji Yuasa *, Hitoshi Kubota, Akio Fukushima, Kay Yakushiji, Taro Nagahama, Yoshishige Suzuki and Koji Ando

[Translation from *Synthesiology*, Vol.2, No.3, p.211-222 (2009)]

We have been promoting *Full Research* to develop ultimate green IT devices based on non-volatile electronics. A core technology of non-volatile electronics is a non-volatile memory possessing features such as large capacity, high-speed operation, and high endurance. To develop such an ultimate non-volatile memory, we developed a novel high-performance magnetic tunnel junction device based on magnesium oxide (MgO) and its mass-manufacturing technology. These technologies have already been commercialized for the production of the magnetic heads of ultra-high density hard disk drives (HDD). Now we are also performing intensive R & D activities for developing the ultimate non-volatile memory called Spin-RAM.

Keywords : Spintronics, magnetoresistance, hard disk drive (HDD), MRAM, Spin-RAM, non-volatile electronics, green IT

1 Introduction

1.1 The need for non-volatile electronics

Current electronic memory devices are built around a core of silicon LSI technology and are basically "volatile," which means that recorded data is erased when the power supply is cut off. That happens because the DRAM[Term 1] general-purpose high-capacity memory and SRAM[Term 2] high-speed memory used by computers and IT home electronics use volatile recording techniques, and the logic unit in the CPUs and other such electronic devices are also all volatile (Fig. 1). Generally, electronic equipment has very long input "standby time." When creating documents with personal computers, for example, the computer is idle for most of the time from one key input to the next key input, and is not doing any work at all. Even during such "standby" times, however, the power to the electronic equipment remains on and power is consumed. The volatility of electronic equipment means that the power cannot be turned off. If the electronic devices that constitute computers and IT appliances could be made "non-volatile" (i.e., memory is retained even when power is disconnected), then basic designs in which the power could be turned on or off instantly, as needed, ("quick-on") or in which the basic state is "normally off" and the power is turned on only when computation is being performed would be possible (Fig. 1). "Normally off", in particular, is a new concept that is completely the opposite of the current electronics concept of power on as the basic state and holds out the promise of ultimate green IT equipment that consumes almost no power.

To realize this dream of normally-off electronic equipment requires development of non-volatile electronics technology for the main memory (DRAM and SRAM) and for the logic unit of computers. As the first stage of research and development aiming at normally-off electronics, AIST is doing R & D to achieve the first of those two requirements, a large-capacity, high-speed and highly-reliable non-volatile computer memory. Here, high reliability mainly refers to rewrite durability. To replace the DRAM and SRAM that serve as the main memory of computers, it is necessary to attain the practically limitless write endurance of 10^{15} rewrite cycles without failure. Incidentally, the write endurance of the flash memory used in current external storage devices is only 10^4 to 10^6 rewrite cycles, and so is not usable as main memory. The ferro-electric memory devices (FeRAM), typical non-volatile memory devices that are already on the market, have a write endurance of 10^8 rewrite cycles, still inadequate to serve as computer main memory. The phase change memory (called PRAM or PCRAM) devices currently under development as non-volatile memory also have limited rewrite times. Spintronics technology (explained below), on the other hand, enables the development of non-volatile memory that has unlimited rewrite times.

The main non-volatile, large-capacity external storage device currently in use is the hard disk drive (HDD) (Fig. 1). In the future, SSD[Term 3] based on flash memory is expected to replace the HDD in small-capacity and high-end applications, but the HDD is expected to remain the primary device for large capacity and low cost storage applications that have a large market for some time into the future. However, high power

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consumption is a problem with the HDD. Reduction of the size (diameter) of the recording medium disk called "platter" is generally an important factor in reduction of HDD power consumption, and the key to that is higher recording density. At this time, HDDs that have platters that are 3.5-inches in diameter are mainstream, but those drives consume a great deal of power (typically 5 W per unit). The smaller 2.5-inch drives, on the other hand, consume only about one-fifth the power required by a 3.5-inch drive. If the current 3.5-inch HDDs can be replaced by 2.5 inch drives that have a higher recording density, the overall power consumption of HDDs could be greatly reduced. AIST is also doing R & D aimed at increasing the recording density of HDDs.

1.2 Spintronics technology for non-volatile electronics

Spintronics is a new field in which new functions are created by using both the electrical (charge) and magnetic (spin) properties of electrons (Fig. 2). Silicon-based electronic devices that use only electron charge are the foundation of information technology, but they are inadequate for implementing non-volatile memory. Magnetics, which uses only electron spin on the other hand, is good for non-volatile memory, but does not perform well in terms of logic operations or power consumption. For future technology, spintronics opens up the possibility of realizing non-volatility together with features such as high reliability, low power consumption and logic operations at the same time. Non-volatile memory that offers large capacity, high-speed and high reliability together will be the core technology for non-volatile electronics for the ultimate normally-off computers.

Spintronics uses quantum mechanical phenomena to correlate electron charge and spin. Of those phenomena, the most important is magnetoresistance. Magnetoresistance (MR) is the change in the electrical resistance of a solid or a solid-state device when a magnetic field is applied to it. The relative rate of change in electrical resistance is referred to as the magnetoresistance ratio (MR ratio). The magnetoresistance effect can be used to convert a magnetic field signal into an electrical signal, so it can be applied to sensing magnetic fields in the design of magnetic read heads for hard disk drives (HDDs). Furthermore, magnetic hysteresis characteristics of ferromagnets can be used to implement the same kind of non-volatile memory as is possible with magnetic recording.

For devices that use the magnetoresistance effect, the MR ratio at room temperature and low magnetic fields (below a few milli-tesla) serves as an index of performance, because the magnetic fields that can be generated in ordinary electronic circuits are small, several milli-tesla at most. Larger MR ratios at room temperature and low magnetic field mean that devices of higher performance can be developed. This important metric of practical application had values of only from 1 to 2 %, which was never considered as having potential for practical use. Then, A. Fert *et al.* and P. Grünberg *et al.* discovered the giant magnetoresistance effect (GMR effect) of metallic magnetic multi-layers in 1988, achieving an MR ratio at room temperature and low magnetic field of about 10 %, an order of magnitude higher than any previous value. That discovery earned Fert and Grünberg the 2007 Nobel Prize in Physics. About ten years after its discovery, GMR was applied to the magnetic read head of hard disk drives (GMR head), after which the capacity of HDDs increased rapidly (Fig. 3). Furthermore, the discovery of the GMR effect stimulated vigorous research and development on magnetoresistance around the world. It was also linked to achievement of the TMR effect (explained below) at room temperature (Fig. 3). We do not cover GMR here; for more information on that subject, see the formal paper for the 2007 Nobel Prize in Physics[1]. The TMR effect is described in more detail in the next section.

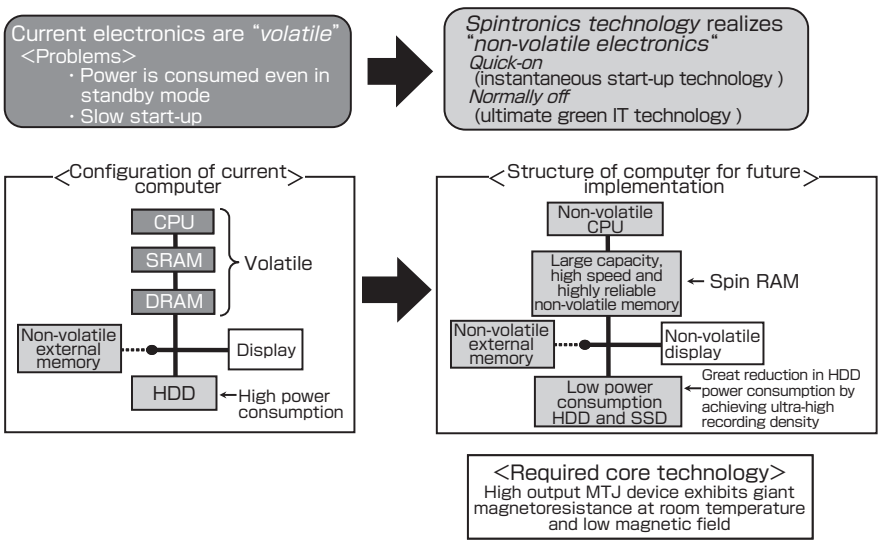

Fig. 1 Current electronics and future non-volatile electronics.

1.3 Room temperature TMR effect and its applications

A magnetic tunnel junction (MTJ) device consists of an insulation layer that is no thicker than a few nanometers (tunnel barrier) sandwiched between two ferromagnetic metal layers (ferro-magnetic electrodes) (Fig. 4). Insulators do not normally carry current, but when the insulation is less than a few nanometers thick, minute currents flow due to quantum effects, a phenomenon called the "tunneling effect." The current and electrical resistance generated by this effect are referred to as the tunnel current and the tunnel resistance. If the electrode layers are ferro-magnetic, the tunnel resistance is small in the parallel magnetization state (P state: Fig. 4(a)) and a larger current flows. In the anti-parallel magnetization state (AP state: Fig. 4(b)), on the other hand, the tunnel resistance is large and the tunneling current is small. That phenomenon is called the tunnel magnetoresistance (TMR) effect. MTJ devices can be switched between the P state and the AP state by application of a magnetic field (Fig. 4(d)), creating magnetoresistance. Also, because ferromagnets possess a magnetic hysteresis characteristic, they are bistable at zero magnetic field, having a P state or an AP state. An MTJ device can thus act as a non-volatile memory to store one bit of information.

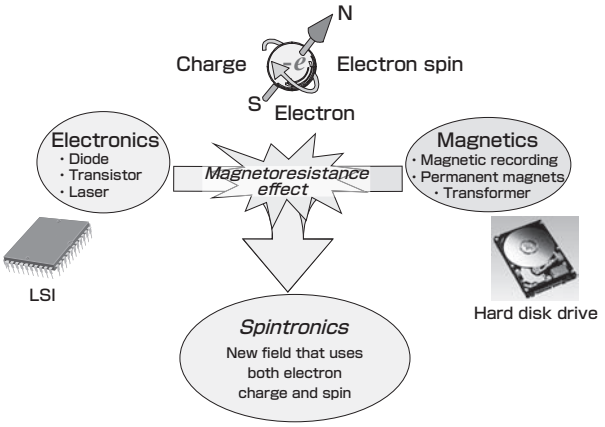

Fig. 2 Spintronics and magnetoresistance effect.

The low-temperature TMR effect has been known since the 1970s, but room-temperature magnetoresistance had not been obtained and there was little interest in the phenomenon in the following ten years. With the discovery of GMR in 1988, however, came much R & D on magnetic sensors (HDD magnetic heads, etc.) and TMR also began to attract interest again. In 1995, Miyazaki *et al.*[2] and J. Moodera *et al.*[3] used amorphous (random arrangement of atoms) aluminum oxide (Al-O) for the tunnel barrier and polycrystalline transition metals such as Fe or Co for the ferromagnetic electrodes to fabricate MTJ devices that had MR ratios of close to 20 % at room temperature and low magnetic field (Fig. 3). That was the highest room temperature MR ratio at the time, and the achievement thrust the TMR effect into the limelight. Subsequently, there was vigorous work on optimizing the method for making the Al-O tunnel barrier and the electrode material, resulting in achievement of room temperature MR ratios of over 70 % for the TMR effect.

The room temperature TMR effect was put to practical use in an HDD magnetic read head (TMR head) in 2004, about ten years after it had been implemented (Fig. 5). Combining that TMR head with a perpendicular magnetic recording medium achieved a high recording density of 100 Gbit/inch^2. Furthermore, a relatively low capacity (4 Mbit to 16 Mbit) non-volatile MRAM (Fig. 6) product based on the MTJ device was commercialized in 2006. That product drew attention as a unique non-volatile memory that featured high reliability (unlimited write endurance). The reason for the unlimited write endurance of the MRAM is that reversal of the spin direction (the rewrite operation) causes no degradation of the material at all. In addition, MRAM operation is faster than DRAM and nearly as fast as the high-speed SRAM used in the CPU. However, there remains little margin for improvement in the performance (room temperature MR ratio) of MTJ devices based on an amorphous Al-O tunnel barrier, and that was a serious problem for achieving higher

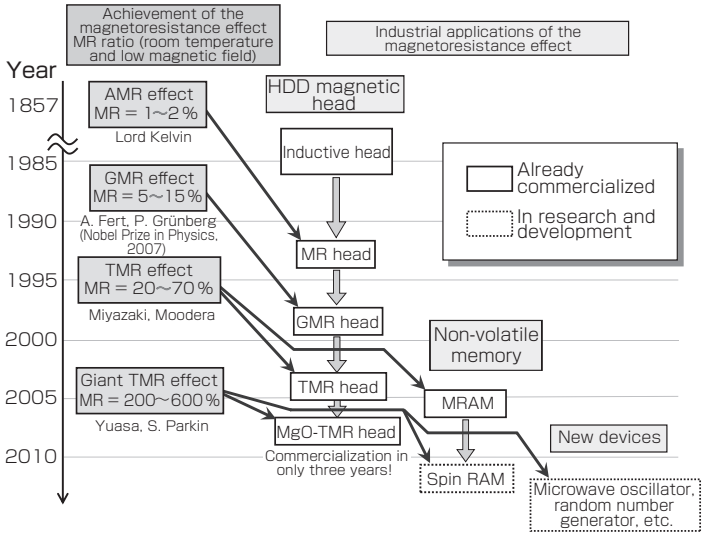

Fig. 3 History of the magnetoresistance effect and its industrial applications.

performance in hard disk drives and MRAM. With an MTJ device that uses an amorphous barrier, development of an HDD recording density higher than 200 Gbit/inch^2 and gigabit-class large-capacity MRAM was difficult (Fig. 4). To overcome that limitation and develop next-generation devices that have a higher integration scale, higher speed and less power consumption, even higher MR ratios were essential.

First-principle theoretical calculations for a single-crystal MTJ device that uses a crystalline tunnel barrier rather than the amorphous barrier that had previously blocked further progress were published around 2001, and a huge theoretical MR ratio of over 1000 % was predicted. In 2004, AIST was the first in the world to experimentally achieve a giant room temperature TMR effect in an MTJ device using a crystalline magnesium oxide (MgO) tunnel barrier, making a great step forward in applied research on the TMR effect. Beginning with the next section of this paper, we describe the stages of *Type 1 Basic Research*, *Type 2 Basic Research*, and commercialization in the R & D of a high-performance MTJ device that uses a crystalline MgO tunnel barrier, and explain how the *Full Research*[4] was conducted.

The Spintronics Group of the AIST Electronics Research Division did R & D on the two outcomes described below in the second research strategy period of AIST.

(1) A practical next-generation magnetic head for the ultra-high-density HDD

The objective is a next-generation magnetic head for ultra-high-density HDDs that have a recording density of over 200 Gbit/inch^2 to reduce power consumption by achieving an ultra-high-density, compact HDD.

(2) Basic technology for ultimate non-volatile memory (spin RAM)

The goal is basic spin RAM technology for the large-capacity, high-speed and highly-reliable ultimate non-volatile memory that will be the core technology for non-volatile electronics.

To produce these two outcomes, we target two R & D goals: (i) development of a landmark high-performance MTJ device and (ii) development of mass production technology for it.

2 Elemental technology

2.1 Theory of the magnesium oxide (MgO) tunnel barrier TMR effect

This section explains the physical theory behind the TMR effect. The good crystal lattice-matching between the (001) surface of the body-centered cubic crystal (bcc) Fe and the (001) crystal surface of magnesium oxide (MgO) allows experimental fabrication of a fully epitaxial MTJ thin film that has a high quality Fe (001)/MgO (001)/Fe (001) structure. Even combining a bcc (001) electrode layer of an alloy whose main constituents are Fe and Co and a MgO (001) tunnel barrier allows formation of high-quality single-crystal MTJ thin film in the same way. In 2001, Butler *et al.*[5] and Mathon *et al.*[6] performed first-principle theoretical calculations for a single-crystal MTJ that has an Fe (001)/MgO (001)/Fe (001) structure, showing that a giant MR ratio of over 1000 % can be expected in theory. The physical mechanism of this giant TMR effect differs from that when the conventional amorphous Al-O tunnel barrier is used as described below.

The difference in the electron tunneling process for the

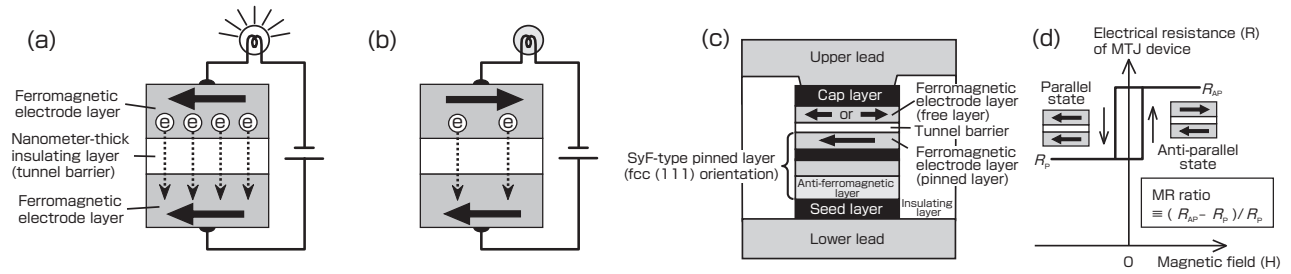

Fig. 4 Tunnel magnetoresistance (TMR) effect of magnetic tunnel junction (MTJ) device.

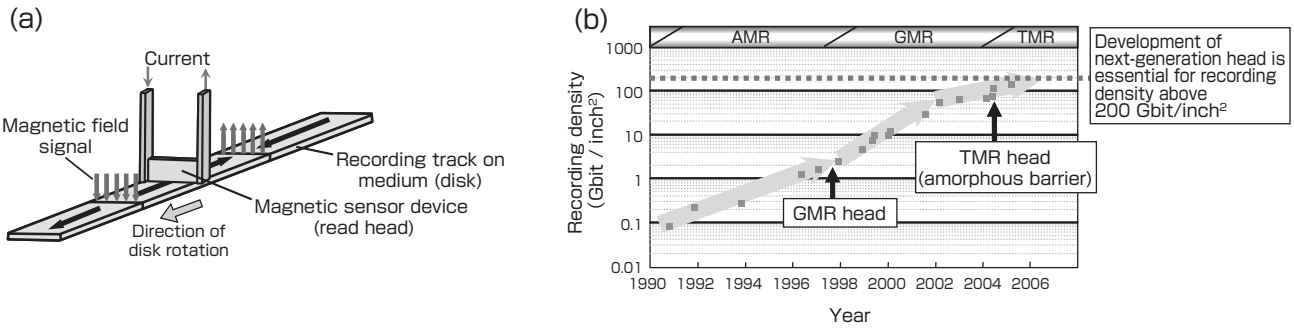

Fig. 5 (a) Hard disk drive (HDD) magnetic read head, (b) Evolution of HDD recording density and magnetic read heads

conventional amorphous Al-O tunnel barrier and the crystalline MgO(001) tunnel barrier is illustrated in Fig. 7. Conduction electron states that have various wave function symmetries (Bloch states) exist in the ferromagnetic electrode. For an amorphous Al-O tunnel barrier, the symmetry of atomic arrangement in the barrier is broken, so the various Bloch states are mixed up within the electrode and tunneling conduction occurs (Fig. 7(a)). Each Bloch state in the electrode generates a positive or negative MR ratio, depending on its orbital symmetry. With an amorphous Al-O barrier, those Bloch states are mixed and all are subject to tunneling conduction. Therefore, the mean MR ratio of the Bloch states (which is to say the MR ratio of the MTJ device) cannot attain a high value, and an MR ratio that greatly exceeds 70 % cannot be attained at room temperature.

If the tunnel barrier is crystalline MgO (001), on the other hand, an entirely different characteristic is predicted by theory. The single-crystal MTJ device tunneling process is illustrated in Fig. 7(b). The tunnel electron is often assumed to be a free electron, but the evanescent states of electrons that are in the actual band gap of the insulating tunnel barrier have a special orbital symmetry and a special band dispersion that differ greatly from free electrons. Three kinds of evanescent states exist within the MgO (001) band gap: Δ_1 (*spd* hybridized high symmetry state), Δ_5 (*pd* mixed state), and $\Delta_{2'}$ (*d* low electronical symmetry state). The attenuation rates of the density of states of these states within the tunnel barrier depend greatly on the orbital symmetry of the state. The Δ_1 evanescent states have the slowest attenuation in the tunnel barrier (i.e., the longest attenuation length). Accordingly, the tunneling current via this Δ_1 states is dominant (Fig. 7(b)). In an ideal tunneling process, only the Δ_1 Bloch states in the Fe (001) electrode can couple with the Δ_1 evanescent states in the MgO, so the dominant tunneling path is Fe-Δ_1↔ MgO-Δ_1↔Fe-Δ_1. What should be noted here is that the Δ_1 Bloch state in the Fe (001) electrode is a special electron state that can generate a very large positive MR ratio. From the fact that only electrons that have the Δ_1 symmetry selectively pass through the MgO tunnel barrier, as we see in Fig. 7(b), we can theoretically expect a huge MR ratio of over 1000 %. The theoretical prediction for such a huge MR ratio is not limited to the bcc Fe (001) electrode, but is predicted also for Fe and Co based ferromagnetic alloys that have the bcc structure.

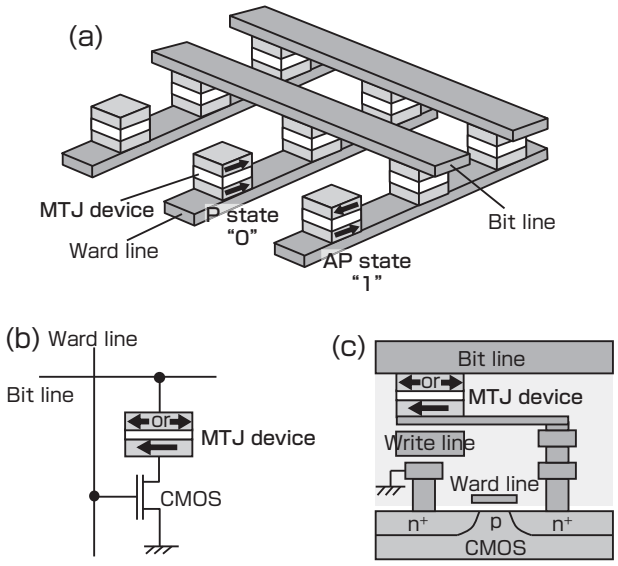

Fig. 6 Non-volatile memory MRAM (a) Schematic diagram, (b) Circuit diagram, (c) Cross-section

2.2 Achieving the MgO tunnel barrier giant TMR effect

When the theoretical prediction of the giant TMR effect for a crystalline MgO tunnel barrier appeared in 2001, there were experimental attempts to actually fabricate MTJ devices that had the single-crystal Fe (001)/MgO (001)/Fe (001) structure, mainly by public research organizations in Europe, but there was no success. Room temperature MR ratios that exceeded those of conventional amorphous Al-O tunnel barrier were not attained, so the expectations for the crystalline MgO tunnel barrier were not met and the theoretical predictions for the giant TMR effect came to be viewed with skepticism. Under those circumstances, AIST continued experimental research on crystalline MgO tunnel barriers, and succeeded in fabricating a high-quality single-crystal Fe (001)/MgO (001)/Fe (001) MTJ device using molecular beam epitaxy (MBE) in 2004 (Fig. 8)[7][8]. That single-crystal MgO-MTJ device was used to achieve the world's first room temperature MR ratio that exceeded that of an amorphous Al-O barrier in the beginning of 2004 (Fig. 9, 1))[7]. That paper also verified high reproducibility, excellent voltage characteristics and other aspects of practicality, and was thus a historical turning point that brought the crystalline MgO tunnel barrier back into the limelight. After that, AIST achieved an even higher room temperature MR ratio of 180 % by further improving the quality of the crystalline MgO tunnel barrier in the latter half of 2004 (Fig. 9, 2))[8]. On the other hand, at about the same time that AIST produced those results, Parkin *et al.* of IBM fabricated an MTJ device that used a preferred-oriented polycrystalline (textured) MgO (001) tunnel barrier with

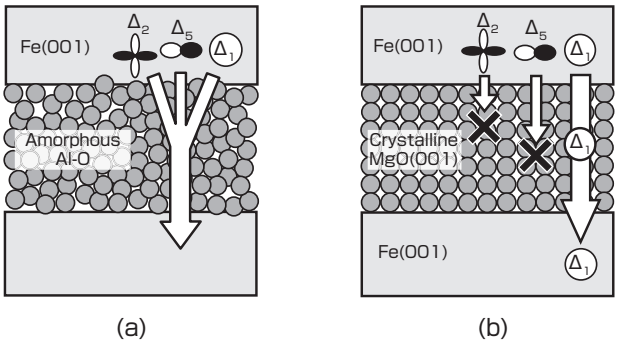

Fig. 7 Electron tunneling transport.
(a) Amorphous Al−O tunnel barrier, (b) crystalline MgO(001) tunnel barrier

a preferential orientation of (001) crystal plane formed by sputtering. That device attained a room temperature MR ratio of 220 % (Fig. 9, 3))[9]. Viewed microscopically, the textured MTJ device has basically the same structure as the single-crystal MTJ device, so it can be considered to manifest the giant TMR effect by the same mechanism. To distinguish it from the conventional TMR effect, we call this kind of very large TMR effect exhibited by the crystalline MgO tunnel barrier the giant TMR effect (nomenclature of the authors). The results shown in Fig. 9 1), 2), and 3) (references[7]-[9]) are also presented in the 2007 Nobel Prize in Physics paper[1] and recognized worldwide as historical papers.

AIST is conducting various types of *Type 1 Basic Research* using a high-quality single-crystal MgO tunnel barrier and has succeeded in observing new phenomena not seen with an amorphous Al-O tunnel barrier, such as the oscillation phenomenon of the TMR effect relative to the thickness of the MgO barrier[8][10], interlayer exchange coupling mediated by the tunnel electron[11], and a complex spin-dependent tunneling spectra[12] in addition to the giant TMR effect. Progress in understanding the physical mechanism of these phenomena should lead to further development of the physics of the tunneling effect.

2.3 Mass production technology for crystalline MgO-MTJ devices

As described above, AIST achieved the landmark *Type 1 Basic Research* result of the giant TMR effect in 2004, but at that time the skeptical outlook on industrial applications for the crystalline MgO-MTJ device was still dominant. The main reason is that neither the single-crystal MgO-MTJ device developed by AIST nor the textured MgO-MTJ device developed by IBM had a device structure suitable for application in practical devices. For application to HDD magnetic heads or MRAM, the lower structure, the "SyF type pin layer" (Fig. 4(c)), is necessary (details are omitted here). However, the basic structure of the SyF-type pin layer is (111) oriented face-centered cubic (fcc) crystal, which has three-fold rotational in-plane symmetry, and it is not possible to grow a MgO (001) layer of different crystal symmetry (an in-plane four-fold rotational symmetry structure) over it.

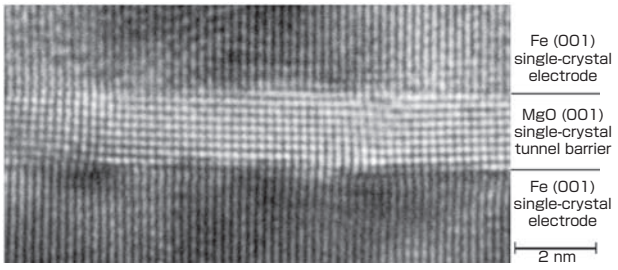

Fig. 8 Single-crystal Fe/MgO/Fe-MTJ device cross-sectional transmission electron microscope (TEM) image [8].

That fundamental problem in crystal growth was a serious problem for fabrication of a MgO-MTJ device over this "practical lower structure".

At first, AIST took the viewpoint of developing a new lower structure that had in-plane four-fold rotational symmetry and tried to sell device manufacturers the idea of MgO-MTJ device technology. However, the reaction from device manufacturers was, "The reliability of the lower structure is directly related to product (HDD and MRAM) reliability. The highly reliable SyF-type pin layer lower structure is the result of ten years or so of research and development, so the development of a new lower structure now is not possible (there is no margin for it)." To be sure, commercialization depends on satisfying many requirements. For example, even for new technology that has outstanding capabilities, just one fatal flaw would prevent commercialization and bring death to the technology, as the word in "valley of death" implies. While we know that to be the greatest difficulty to overcome in *Full Research* in our minds, the reality of the matter comes when we actually experience it. At the time, there were two straightforward solutions: (i) brute-force development of a new lower structure that has in-plane four-fold rotational symmetry structure and (ii) development of a new tunnel barrier that can be formed above a SyF-type pin layer that has in-plane three-fold rotational symmetry. Either of those solutions, however, would require at least five to ten years of development. Furthermore, the requirement of device manufacturers for "mature technology at a level that can be immediately developed and introduced to the production line" meant that it was practically impossible for AIST to independently develop such a solution. Faced with that situation, AIST began joint development with the production system manufacturer Canon ANELVA Corporation and achieved the "landmark solution" described below.

Although we might think of production system manufacturers

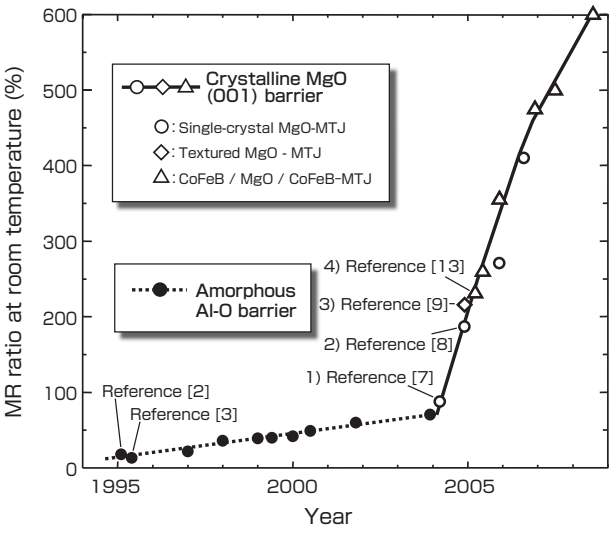

Fig.9 History of room temperature MR ratio improvement.

as simply builders of production systems, current production system manufacturers play the important role of developing new materials and new device production process technology in addition to building the systems. In this R & D, we aimed for a solution to the difficult problem described above by integrating AIST's superior material and device technology seeds and the excellent production process technology and equipment possessed by the production system manufacturer (Fig. 10). In particular, the production sputtering system of that company is a world-standard system in the HDD industry, so if mass production technology can be developed using it, that technology can be rapidly transferred to the production lines of device manufacturers.

In the joint research with the Canon ANELVA Corporation, we discovered that if amorphous CoFeB alloy is used for the lower ferromagnetic electrode, a high-quality oriented polycrystalline (textured) MgO (001) tunnel barrier layer can be grown over it at room temperature (Fig. 11(a))[13]. A CoFeB/MgO/CoFeB structure MTJ device that uses this very special manner of crystal growth can be fabricated over any base layer because the lower electrode layer is amorphous. Moreover, it is formed by room temperature sputtering, so it is ideal in terms of production process compatibility and production efficiency. When that CoFeB/MgO/CoFeB-MTJ device is heat-treated at 250 °C or above, crystallization of the amorphous CoFeB layer begins from the interfaces (Fig. 11), and a bcc CoFeB(001) structure that has good lattice matching with the MgO (001) layer forms[14]. The device structure shown in Fig. 11(b) is basically the same as the single-crystal MgO-MTJ device and the oriented polycrystalline MgO-MTJ device, so giant TMR effect manifests by the same physical mechanism (Fig. 9, 4))[13]. This thin film fabrication process is an original method that counters the common-sense understanding that crystal growth proceeds upwards from the base layer.

That CoFeB/MgO/CoFeB MTJ device has now become the mainstream technology in spintronics applications, and various types of vigorous R & D that use it are proceeding on a worldwide scale. That work has so far achieved a room temperature giant MR ratio of 600 %. Incidentally, success in developing the CoFeB/MgO/CoFeB-MTJ device was achieved in barely one year from the beginning of the joint research with Canon ANELVA. That achievement was largely dependent on the "good lineage" of the crystalline MgO tunnel barrier, but it also verified that the combination of AIST and a production system manufacturer engaging in *Type 2 Basic Research* was a highly suitable approach.

3 Creating Outcomes

3.1 Commercialization of the ultra-high-density MgO-TMR HDD head

The CoFeB/MgO/CoFeB MTJ device developed jointly by AIST and the Canon ANELVA Corporation and that company's production system were quickly introduced to HDD manufacturers' product development lines and energetically used in mass production of HDD magnetic heads (Fig. 10). The result of vigorous product development was the successful commercialization of the second-generation TMR head (MgO-TMR head) using the MgO-MTJ device by various HDD manufacturers in 2007. The MgO-TMR head had greatly higher performance than the

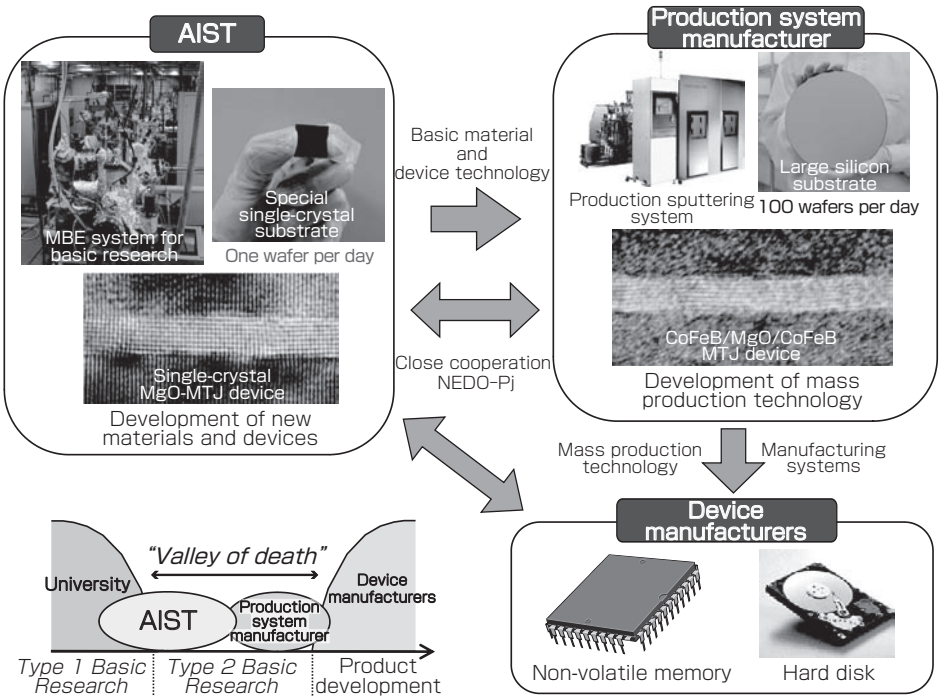

Fig.10 *Full Research* on MgO-MTJ device.

first-generation TMR head, and when combined with the latest perpendicular magnetic recording medium, realized an ultra-high-density HDD with a recording density above 250 Gbit/inch^2 (twice the previous value)[15]. The future potential for development extends up to the 1 Tbit/inch^2 next-generation HDD. By achieving such high HDD recording density, the 3.5 inch drives that are the main devices in use can be replaced with 2.5 inch drives that have adequately high capacity. As a result, 2.5 inch drives are expected to come into mainstream use, even in the market for large-capacity HDD. As described in section 1.1, 2.5 inch drives consume only one-fifth power of the 3.5 inch drive, so replacing 3.5 inch drives with 2.5 inch drives will greatly reduce overall HDD power consumption. The HDD industry has a huge market that compares in scale with DRAM and CPUs (about 3 trillion yen per year). The magnetic head is the most expensive component in a disk drive, and the market value for the magnetic head alone is a huge six hundred billion yen per year. The fact that nearly all of the drives currently being produced are equipped with MgO-TMR heads is the best indication of the social impact of these research results.

3.2 Ultimate non-volatile memory "spin-RAM" R & D

With the objective of implementing the "spin RAM" ultimate non-volatile memory that provides large capacity, high speed and high reliability and will serve as the core technology of non-volatile electronics, AIST is working together with Toshiba Corporation and other companies on the NEDO Spintronics Non-volatile Devices Project. Spin-RAM is an MRAM that uses a new physical phenomenon called "spin torque switching" in the data write technique, achieving a higher capacity than the conventional magnetic field write MRAM. To realize a gigabit-class large capacity spin-RAM requires both increased read out by the giant TMR effect of the MgO-MTJ device and a low-power write technique that uses spin torque switching. Spin torque is the torque generated by the transfer of spin angular momentum from the conduction electrons to the local magnetic moment of the magnetic layer when a current flows in the MTJ device. That spin torque can be used to reverse the direction of spin of the ferro-magnetic electrode (i.e., write). Spin torque switching has been implemented before using GMR devices and Al-O barrier MTJ devices, but the current density needed for switching is very high, so achieving a practical effect was considered difficult. AIST implemented spin torque switching in a MgO-MTJ device for the first time in the world in 2005[16][17]. Furthermore, AIST and Osaka University are collaborating to develop a method for quantitative estimation of spin torque[18][19] and have succeeded in verifying a high output microwave oscillator that uses spin torque[20]. Also, Sony, Tohoku University and many others have been doing vigorous research and development on attaining spin torque switching at lower current.

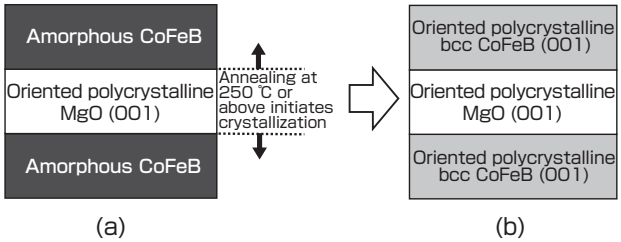

Fig.11 Structure of the CoFeB/MgO/CoFeB MTJ device.

Currently, AIST is moving forward with development of the ultimate spin RAM that uses a perpendicularly magnetized MgO-MTJ device that combines newly developed perpendicularly magnetized electrodes and a crystalline MgO tunnel barrier in collaboration with Toshiba Corporation and with the support of the NEDO project. Because this project involves industry, the government and academia and is currently in progress, there are many confidential aspects, so we must omit the details of research and development here, but we have already verified low current and high speed spin torque write operations and excellent data retention characteristics. We are continuing this R & D with the near-term objective of an ultimate non-volatile memory that uses this perpendicularly magnetized MgO-MTJ device and with the long-term objective of implementing ultimate green IT devices through normally-off computer.

4 Conclusion

We have described here our impressions based on our own experience of *Full Research* achievements. The *Full Research* scenario put forth by AIST is ultimately a conceptualism, and the actual specific methods for executing it must rely on a groping search by the individual persons engaged in R & D in the laboratories. Although conceptualisms are also important, they are not immediately useful in the R & D labs where difficult and pressing problems are being dealt with. Actually linking the results of basic research to commercialization, involves the problem of matching research with social needs and problems of dealing with complex elements such as the difference in interests between organizations and interpersonal relations in addition to the technological problems. Particularly the difference of interests between organizations may create a deadlock, even when the matter is left to the upper levels of the organizations, and there is no progress. In the end, the people in the labs must work within organizations to solve problems, so a relationship of trust among the people doing the R & D in the laboratory is important. That is to say, it is ultimately an interpersonal problem.

Speaking of the technical problems, the "valley of death" that lies between basic research and product development is wider and deeper than is imagined, and it is probably impossible for AIST to cross it alone. Particularly in mature industries

such as electronics, there is an advanced separation of labor such as among manufacturing systems manufacturers, raw materials producers, device manufacturers, and foundries. Therefore, cooperation with appropriate partners at each stage of R & D is essential. The tag-team approach used by AIST and the manufacturing system manufacturer in this case can be called a model case for *Type 2 Basic Research*. The importance of production system manufacturers is generally still not fully recognized, and we feel that AIST should actively send out that message through examples of success such as this.

As a final matter, we attempt to analyze the factors of success relating to commercialization in a mere three years from obtaining the results of *Type 1 Basic Research* in the R & D reported here. We can say that the key to success is measured by "the potential of technology seeds." When *Type 1 Basic Research* produces some remarkable capability, it is thrust into the limelight and draws great praise. In product development, however, all of ten or more important item tests must be passed, and even one failure can be a fatal defect that makes commercialization impossible. Even if it is a "landmark new technology" that gets published in a well-known scientific journal, there is most often some fatal defect, and in nearly all cases, the valley of death cannot be crossed. Although only the rare technology seeds that have true potential can cross the "valley of death", and even for those cases the stage of practicality cannot be reached without many collaborators and endorsers brought together from the industrial world. The key here is how to bring together capable collaborators and endorsers. Our feeling is that if the technology seed has strong potential and suitable results are announced with appropriate timing, "People will naturally gather together." Technology seeds that have strong potential draw capable people. In industry, most people are conservative, and views of new technology are most often skeptical and critical, but there are certainly also developers and managers who can see the potential and appropriately evaluate new technology. If technology seeds fail to bring together collaborators even after a number of announcements and industry is completely unmoved, it is best to first consider whether one's own technology seeds might have weak potential before putting the blame on the conservatism of industry.

Note

This research was done in part with support from the Japan Science and Technology Agency (JST) and also from the New Energy and Industrial Technology Development Organization (NEDO) Nanotech Challenge Project and the NEDO Spintronics Non-volatile Devices Project. Parts of this work were done in joint research with Canon ANELVA Corporation and Toshiba Corporation.

Terminology

Term 1. DRAM: A type of large capacity memory used in computers. Information is stored by charging capacitors. When the power is cut off, the capacitor is discharged and the stored information is lost (volatile memory).

Term 2. SRAM: A type of volatile memory used for CPU cache memory, etc. that uses the bistable state of a flip-flop circuit to store data. It is fast and highly reliable, and also compatible with logic circuits, but it is not suited to increased integration scales and power consumption is high.

Term 3. SSD: An external storage device that uses flash memory as the recording medium. Unlike hard disk drives, it has no moving parts. SSD is an acronym for Solid State Drive. Compared to hard disk drives, it consumes little power and is resistant to physical shock. The cost per unit capacity, however, is an order of magnitude higher than current hard disk drives.

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Discussion with Reviewers

1 Achieving the breakthrough

Question and comment (Naoto Kobayashi, Center for Research Strategy, Waseda University)

I think this R & D is a rare example where the research results, through the two major breakthroughs of " 1) the giant TMR effect achieved using MgO" and " 2) the realization of CoFeB/MgO/CoFeB structure" are linked to the outcome of development of a product that is actually sold in the market in a short period of time. From this paper we can understand the most part why these respective breakthroughs could be achieved, but could we have an explanation of the process of selecting those processes and materials (including the reasons for excluding other materials and processes, etc., and, if necessary, the research and development system). Also, if there was an effective serendipity or other such factors, please also describe them.

Answer (Shinji Yuasa)

Half of the success factors were a matter of achieving set goals, and the other half involved good luck (serendipity). Theoretically, there are a number of promising crystalline tunnel barrier materials other than MgO. The first thing to consider is the problem of which theoretical prediction is correct (which prediction should we bet on). Furthermore, to reach a practical stage means the satisfaction of various requirements such as (i) fabricating thin films of nanometer order thickness without opening up pinholes, (ii) prevention of reaction and atomic diffusion at the interface with the metal electrode material, (iii) crystallization in low temperature layer formation, (iv) a sufficiently high breakdown voltage, and (v) product-level reliability. Based on the experience and knowledge we have accumulated and as the result of considering what material system would be the most promising from many angles, we came to the conclusion that using anything other than crystalline MgO would present difficulties even before we began any experimentation. However, that did not at all mean that there would be no problems if we chose to go with MgO. The most troubling problem before beginning the research and development was production process compatibility, but that problem was solved by the development of the CoFeB/MgO/CoFeB structure in joint research with Canon ANELVA Corporation. Serendipity was large among the success factors in forming the CoFeB/MgO/CoFeB structure, I believe, even with the active work done by the excellent engineers of the Canon ANELVA Corporation. Also, the inability to judge ultimate reliability at the product level until the final stage of research and development is reached is a problem, but we had the feeling from the initial stage of research and development that the reliability of MgO was high. That was not a conclusion based on theoretical consideration, but an intuition that can only be felt by people working in the lab.

2 Reduction of power consumption

Question and comment (Naoto Kobayashi)

You have described that the development of non-volatile memory is linked to the realization of ultimate green IT devices. While practical non-volatile memory will greatly reduce the power needed to operate memories, I think that no overall reduction in power consumption can be expected unless the non-volatile CPU is realized as described in Fig. 1. Thus I have two questions: 1) As far as we know now, what are the approximate relative proportions of the power consumption decrease that can be achieved by implementing the spin RAM and that achievable by implementing the future non-volatile CPU? and 2) I believe that spin FETs is essential to implementation of future non-volatile CPUs. What do you think of the prospects for that field of research and development?

Answer (Shinji Yuasa)

1) Not much reduction in power consumption can be expected from simply replacing DRAM and SRAM with MRAM and spin RAM. As you point out, a radical reduction in power consumption would require achieving non-volatility for both memory and the CPU together as a set. While non-volatile DRAM and SRAM serve as the first stage, the merits of that in itself include (i) an opening up of the near-future SRAM and DRAM scaling limits and (ii) higher integration scale and lower cost achieved by on-chip system LSI memory consisting of spin RAM only.

2) While switching devices that have a non-volatile memory function such as spin FETs are ideal, one opinion that non-volatile logic circuits can also be designed by combining existing memory devices (MTJ devices and ferro-electric memory devices, etc.) and CMOS. In any event, achieving normally-off equipment with a non-volatile CPU is a grand plan that will require considerable time and investment, and so is expected to need R & D on a 20-year scale.

Question and comment (Kazuhito Ohmaki, Department of Information and Computer Sciences, Toyo University)

Particularly in the Introduction, TMR is proposed as a key technology for normally-off computers. With today's computers, it may look like the computer is idle between key inputs, but in the meantime, the computer is busy monitoring communication lines,

scanning display devices, and occasionally computing encryption algorithms and other such tasks, and thus not necessarily idle. I think you need to strengthen the explanation of the architecture for normally-off computers a little.

Answer (Shinji Yuasa)

My desktop personal computer is connected to a liquid crystal monitor, two hard disk drives, a DVD drive, LAN cables, and USB memory. I have Windows running on it, and under Windows I run word processing software, an Internet browser, a mail program, presentation authoring software, and a spreadsheet program all at the same time. Still, monitoring the CPU use shows that the use rate is normally only 1 to 4 %, and rarely exceeds 5 %. In other words, even though various kinds of processing go on in the background, it's still true that most of the time the CPU is in the standby state. Current electronics that are based on volatility are designed to reduce power consumption by lowering the CPU clock frequency or the power supply voltage during times of low load, but those methods are naturally limited. We think that, in the long term, green IT technology implemented with normally-off technology is necessary.

3 Switching speed

Question and comment (Kazuhito Ohmaki)

I'm not an expert in this, but my feeling is that switching speed will be a problem when using magnetic operations. When aiming for energy conservation by using magnetism, as with the TMR device, I wonder about the prospects for switching speed compared to current silicon technology for implementing normally-off computers. Would you comment on that?

Answer (Shinji Yuasa)

The magnetic switching speed is essentially fast, and can be faster than a few nanoseconds. Non-volatile memory that operates about as fast as the fast memory SRAM that is currently used in CPUs is feasible. It does not, however, represent a landmark speed increase over current silicon technology. In other words, while we can expect about the same operating speed as with current technology, the objective for non-volatility is landmark low power consumption. We added a brief explanation concerning operating speed to section 1.3.

4 Theoretical background

Question and comment (Naoto Kobayashi)

You have explained that what provided the opportunity for the first breakthrough (giant TMR using MgO) was the first-principle computation by Butler and Mathon, but I would like to have some explanation of the research background of their dealing with MgO. Was there any theoretical contribution from Japan regarding this?

Answer (Shinji Yuasa)

It's not that Butler and Mathon were the first to predict the giant TMR effect, but that they were the first to choose the Fe/MgO/Fe structure as a representative example that strict first-principle computation was possible. With the conventional amorphous Al-O tunnel barrier, the unordered amorphous structure did not allow first-principle computation. The Fe/MgO/Fe structure, on the other hand, features good crystal lattice matching and the possibility of experimental implementation, and I heard that led them to consider the Fe/MgO/Fe structure. Thus, theoretical prediction of the giant TMR effect as a result of performing the Fe/MgO/Fe theoretical calculations could be called a kind of serendipity. In 2001, there were, unfortunately, no researchers who had performed those theoretical calculations in Japan. The computation itself is not particularly difficult, and I even remember it being called "a too-obvious theoretical calculation." However, this was a "Columbus's egg" kind of thing, and I think that to actually perform an obvious computation and present the results to experimenters is a praiseworthy achievement.

5 A critical eye on technology seeds

Question and comment (Naoto Kobayashi)

The statement in the conclusion that "the potential of technology seeds" is very important is highly interesting. 1) It is very clear that this R & D was a technology seed of very high potential, but I wonder if that might be considered "serendipitous". Or perhaps only the technology seeds that reach the final goal can be said to effectively have had potential. If that is not so, how should we cultivate the feeling or sense (Leo Esaki's taste?) for looking critically at the potential in advance? 2) I think that whether the technology seed has potential or not is, not knowable until technological development has proceeded to some extent. Getting across the valley of death requires passing over a number of hurdles, but how can we judge the potential of technology seeds in those respective stages? In other words, how should we decide whether to continue technological development or abandon it when taking the "critical view"?

Answer (Shinji Yuasa)

Let me respond to questions 1) and 2) together. As is also described briefly in discussion 1, of the various requirements for practicality, whether or not product-level reliability, yield, and other such requirements can be satisfied cannot be known until the final stage of *Full Research*. Accordingly, we can also say that whether it is good technology that has potential or not cannot really be known until commercialization is attempted. Nevertheless, concerning the opposite judgment that this technology has weak potential, we believe that it is possible to make a decision before beginning R & D or in the first stage. Out of the many technology seeds that appear, I, myself, try to quickly terminate work on weak technology in an early stage. I think that recognizing technology as strong or weak requires a sense that allows phenomena to be analyzed logically and from many viewpoints in a broad field of view. This is probably the opposite of the ability to drill down deeply into a particular phenomenon. While both of those capabilities are essential to conducting *Full Research*, it is probably too much to expect both from a single researcher. I believe that researchers who are able to drill down into a single phenomenon are in the majority, and those capable of analyzing phenomena from diverse viewpoints in a wide field of view are a smaller group. If researchers lack a sense that allows them to judge the relative potential of technology, the research and development manager should be able to compensate for it. I do not know how we can cultivate a sense for judging the potential of technology seeds, but I should hope that those who become research and development managers would have such a sense.

A marked improvement in the reliability of the measurement of trace moisture in gases

— Establishment of metrological traceability and a performance evaluation of trace moisture analyzers —

Hisashi Abe

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The reliability of the measurement of trace moisture in a gas was improved markedly owing to the establishment of a trace moisture standard at AIST. As a result, problems with conventional methods of measuring trace moisture in gases have been revealed. This article presents the scenario that we adopted to improve the reliability of the measurement of trace moisture, development of a national trace moisture standard, provision of metrological traceability to a national standard for industrial measurements, and problems of the commercially available trace moisture analyzers as revealed through comparisons with the national standard. This improvement makes it possible to properly evaluate the quality of high-purity gases used in industry.

Keywords : Trace moisture, primary measurement standard, humidity, metrological traceability, reliability

1 Introduction

Water is an important substance without which mankind cannot survive. However, it also causes problems such as residual impurity in scientific experiments and in manufacturing processes where a high-vacuum condition or a high-purity gas is required. This is because water is present in the atmosphere in large quantities and therefore can intrude readily everywhere, and can adsorb quickly onto the surfaces of many materials. Because of these characteristics, water is considered to be an extremely troublesome impurity that is difficult to remove. Therefore, in any scientific experiment or manufacturing process, if a high-vacuum condition or a high-purity gas is required, it is necessary to address the effect of residual moisture at some point. In fact, in the field of semiconductor manufacturing, the high purity of the gas used in manufacturing is essential with the rapid progress in the integration and refinement of devices, and the control of trace-level residual moisture in the gas has become a major issue. For example, in the case of the high-purity nitrogen gas used to prevent contamination of the wafer surface, the control of residual moisture to several nmol/mol (ppb) or less in amount-of-substance fraction (mole fraction) is considered necessary[1], corresponding to a frost point [Term 1] of -100 ºC or less at atmospheric pressure. As another example, in the case of a gallium nitride (GaN) light-emitting diode, a significant decrease in light-emitting efficiency was observed in the presence of trace residual moisture of 1 μmol/mol (ppm) or less[2] in an NH_3 gas used in the film growth by metal-organic vapor-phase epitaxy (MOVPE). In any case, a highly reliable moisture measurement is essential to control the trace level of moisture and to correctly assess the effect of residual moisture.

Figure 1 shows the humidity range for which the national standard has been established in Japan. Several types of measuring instruments[3] for measuring the moisture content in gas (humidity) at various ranges are available commercially and are used in many production sites and scientific experiments. However, in the range of trace moisture of 1 μmol/mol (ppm) or less, the measured value of humidity may vary with the instrument used even if the measurement is performed under the same conditions. When such issues arise, they are usually resolved by calibrating the measuring instruments on the basis of the national standard, but there has been no national standard in the trace moisture region until recently, and the calibration of instruments is not straightforward. Hence, when the calibration is essential, the users or measuring instrument companies have had to attempt the calibration by some method that they devised on their own. However, the mismatch of values indicated by the instrument occurred frequently even when the calibration was performed in this way, and the measurements of trace moisture have not been particularly reliable.

To settle this issue, the National Institute of Advanced Industrial Science and Technology (AIST) established the national standard for trace moisture (trace moisture standard); the standard with the highest metrological quality called the primary measurement standard, [Term 2] which is directly traceable to the International System of Units (SI), was developed. Furthermore, a system for providing the trace moisture standard (calibration service) was organized, as illustrated in Fig. 2. In the process of developing the trace moisture standard, it was found that a new trace moisture analyzer based on cavity ring-down spectroscopy (CRDS) had

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excellent capability for measuring trace moisture. Because of these three reasons, namely, the national standard for trace moisture was established, the calibration service system traceable to the SI was organized, and a high-performance measuring instrument became available commercially, the reliability of trace moisture measurement has very recently increased markedly. Simultaneously, the problems concerning the methods conventionally used for measuring trace moisture have been revealed.

This paper describes the scenario adopted by AIST for increasing the reliability of trace moisture measurement, research activities toward achieving this objective, and the recent changes in the reliability of trace moisture measurement brought about by the results of this research.

2 Scenario and objective of increasing the reliability of trace moisture measurement

To maintain the high reliability of trace moisture measurement in the production sites and scientific experiments, the following are necessary: (1) establishment of a national standard for trace moisture as the basis of the scale of the trace moisture analyzer, (2) organization of a calibration service for measuring instruments, and (3) a high-performance measuring instrument that can be used for the trace moisture measurement in the production sites or scientific experiments. The relationships among these three factors are shown in Fig. 3. The issues that must be resolved to realize (1)-(3) are shown in the second column from the left, and are connected by arrows to (1)-(3). These issues must be solved by national metrology institutes, calibration service providers, and measuring instrument manufacturers. In the figure, the arrows connect the issues that each party must tackle. In the relationships among (1)-(3), it is important to organize the calibration service system traceable to the SI, and it is extremely effective to demonstrate the measurement capability of the instrument on the basis of an experiment in which the traceability to the national standard is maintained. Therefore, we considered (1) as the priority among the three. Because (1) was an issue that must be addressed by national metrology institutes, AIST initiated research to establish the trace moisture standard in a gas (nitrogen) in 2001. The goal for the lower limit of trace moisture was set as 14 nmol/mol (ppb), corresponding to a frost point of -100 ºC at atmospheric pressure, and the upper limit was set as 1 µmol/mol (ppm), corresponding to -75 ºC, considering the lower limit of the frost point achievable using the current frost point generator at AIST. The measurement uncertainty was set as a relative standard uncertainty of 11 %, corresponding to 0.5 ºC around the frost point of -100 ºC. This figure surpasses the standard uncertainty of 0.5 ºC at a frost point of -95 ºC that was set as a goal by the National Physical Laboratory (NPL) of UK at that time[4]. For (2), we planned, for some time immediately after the establishment of the standard, that the calibration service would be provided directly by AIST to individual users upon request using the national standard, and that calibration service providers would be recruited at the same time. For (3), we planned, after the establishment of the national standard, that the evaluation of the measuring instrument would be performed using the national standard to find instruments demonstrating high performance. Research activities performed in line with this scenario are described below.

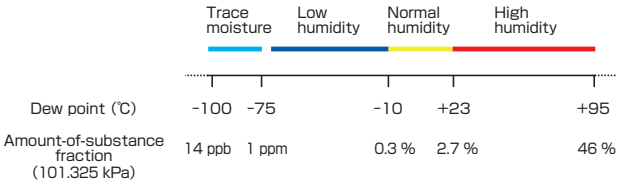

Fig. 1 Current state of calibration service for moisture in gas (humidity) in Japan.
The calibration service for the frost point range between -75 ºC and -70 ºC is currently in preparation.

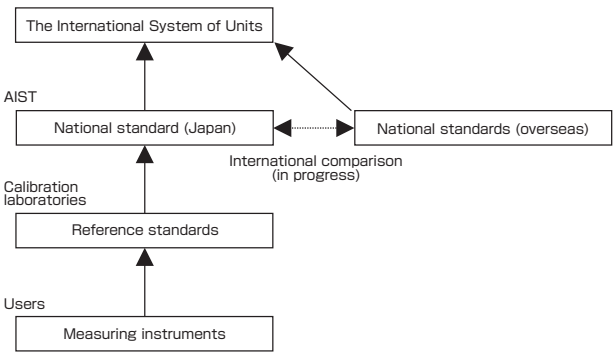

Fig. 2 Traceability system for trace moisture measurement.

3 Establishment of the national standard for trace moisture

The national standard for trace moisture can be realized with a generator that generates gas containing a certain amount of moisture with a known standard value (trace moisture generator), and then performing the uncertainty evaluation of the standard value. When establishing a national standard,

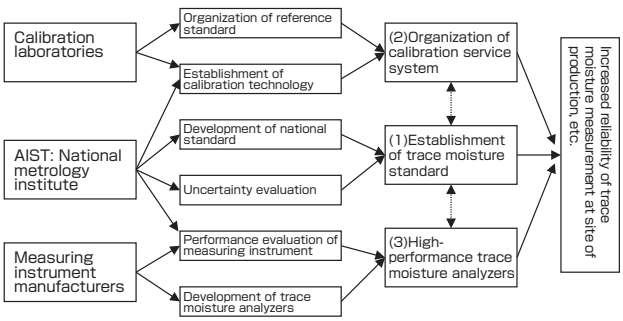

Fig. 3 Scenario for elemental technologies and integration to increase the reliability of trace moisture measurement.

the traceability to the SI may be guaranteed by calibrating the standard using a national standard of the same quantity established in another country, and many developing countries often select this method. In contrast, many advanced countries develop a standard called a primary measurement standard that is directly traceable to the SI. The method adopted depends on the technology, cost, and level of the domestic industry. Because there are advanced semiconductor and process industries in Japan, AIST decided to develop its own primary measurement standard.

Another very important task is selecting the generation method for the trace moisture generator. Here, AIST selected a unique method, different from those of national metrology institutes of other countries. The reasons were because we had a specific plan for this unique method, we considered it possible to achieve the world's highest precision if this unique method was successful, and the reliability of the standard could be increased by maintaining multiple national standards using different methods around the world, rather than relying on a single method.

3.1 Selection of the generation method

There are several methods of generating trace moisture, and AIST selected a method called the diffusion tube method. Figure 4 shows a schematic diagram of the trace moisture generator using the diffusion tube method[5]. A diffusion cell is composed of a small water vessel and a diffusion tube made of metal such as stainless steel, and this is placed in the generation chamber under controlled temperature and pressure. The water vessel of the cell contains water, and water vapor with pressure that responds to a given temperature is generated inside the vessel. The vapor passes through the diffusion tube and travels to the generation chamber. Trace moisture is generated by mixing the vapor with flow-controlled dry gas in the chamber. The amount-of-substance fraction of water in the gas is determined by measurement of the mass of moisture that evaporates per unit time (evaporation rate) and the mass of dry gas that flows per unit time (flow rate).

Table 1 Comparison of diffusion tube method and frost point generation method.

Generation method	Diffusion tube	Frost point
Achievement and check of saturation	Not necessary	Necessary
Vapor pressure equation and enhancement factor	Not necessary	Necessary
Measurement of extremely small mass loss rate	Necessary	Not necessary
Zero gas	Necessary	Not necessary
Past performance as a primary standard	No	Yes

National metrology institutes other than AIST commonly employ a method called the frost point method as the core of their low-humidity and trace moisture generation system. Figure 5 shows a schematic diagram of the frost point generator. A gas is passed through a saturation chamber that is saturated with water vapor evaporated from ice at a constant temperature, and humid gas is generated by mixing the vapor and gas. When saturation is complete and the state of equilibrium is achieved, humid gas with a frost point equivalent to the temperature of ice is generated. Trace moisture can be generated using low-temperature ice (-75 ºC or less). The amount-of-substance fraction of water in the generated gas is determined by measuring the ice temperature, the pressure inside the generation chamber, as well as the vapor pressure formula of ice and the enhancement factor of ice [Term 3] [6][7]. Although the principle of this method is simple, it is highly reliable for generating humidity.

Table 1 gives a comparison of the advantages and disadvantages of the two methods. One of the advantages of the diffusion tube method is that it does not require the achievement and confirmation of saturation, which are necessary in the frost point method. In the frost point method, complete saturation in the saturation chamber is necessary, but the state of equilibrium is difficult to achieve in the low-temperature range because the amount of vapor

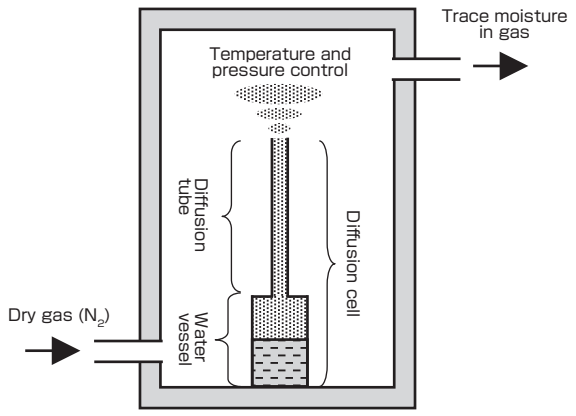

Fig. 4 Schematic diagram of diffusion-tube trace-moisture generator.

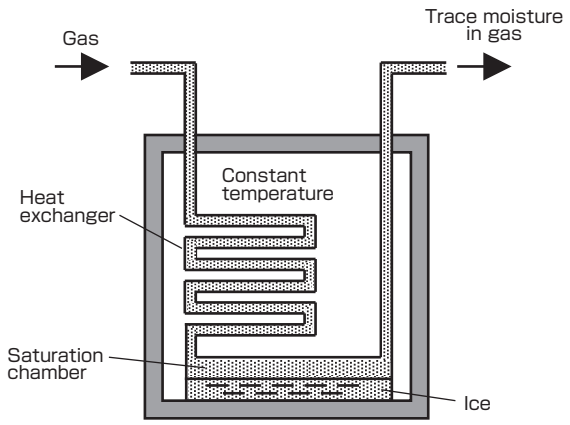

Fig. 5 Schematic diagram of frost point generator.

from ice decreases with the decrease in temperature, and it becomes more sensitive to external thermal stimuli as the vapor pressure becomes more susceptible to temperature change. Moreover, even when saturation is achieved, it is not easy to confirm the state of equilibrium, and therefore, a large degree of uncertainty must be taken into account. These issues can be avoided in the diffusion tube method. For determining the amount-of-substance fraction of water in the generated gas, the frost point method requires the vapor pressure formula and enhancement factor. In contrast, the diffusion tube method does not suffer from this problem, because this method can be used to determine the amount-of-substance fraction of water in the gas from the measurements of the evaporation rate and flow rate, and therefore, it is a method clearly traceable to the SI to generate a humid gas with a known amount-of-substance fraction of water. Because Sonntag's equation,[7][8] which is often used as the vapor pressure formula for ice, is only effective down to -100 ºC, the amount-of-substance fraction of water cannot be determined when Sonntag's equation is used for temperatures below -100 ºC in the frost point method. Moreover, there have been no reports of experiments using the enhancement factor below -35 ºC, and it has been pointed out that the extrapolation of the formula is unreliable below this temperature[9]. This is not an issue for the diffusion tube method, which neither requires the vapor pressure formula nor the enhancement factor.

However, there are disadvantages in the diffusion tube method, one of which is the need to measure a very small evaporation rate. For the generation of trace moisture of 14 nmol/mol (ppb) using the diffusion tube method, the evaporation rate becomes extremely small, approximately 14 μg/h (≈780 nmol/h) when the dry gas is nitrogen and the flow rate is 20 L/min (at 0 ºC and 101.325 kPa); the key is how this evaporation rate can be measured with as small uncertainty as possible. There is also the problem of zero gas. In the diffusion tube method, the moisture remaining in the dry gas may generate uncertainty, and it is necessary to prepare a dry gas called zero gas where the moisture content is lowered to as close to zero as possible. Another problem is that there have been no previous reports in which the diffusion tube method was established as a primary measurement standard of trace moisture. This meant that little of the knowledge, experience, or technology needed for the standard establishment has been accumulated, and it was necessary to design the instrument and develop the analysis method of the uncertainty almost from scratch.

After carefully reviewing the advantages and disadvantages of the diffusion tube and frost point methods, AIST selected the diffusion tube method, as already mentioned. There were four primary reasons for selecting this method, as follows. First, the realization and confirmation of saturation required in the frost point method were considered problematic. Investigation of the development of the trace moisture standard started in around 1999 at AIST (formerly, National Research Laboratory of Metrology, Agency of Industrial Science and Technology), but some national metrology institutes in other countries had already started developing the trace moisture standard on the basis of the frost point method. However, to the best of our knowledge, there was no published research at that time that reported the uncertainty of the trace moisture standard around the frost point of -100 ºC on the basis of measurement traceable to the SI and that described the analysis method of uncertainty evaluation. We considered that this was attributable to the difficulty in the realization and confirmation of perfect saturation around -100 ºC. Second, the direction of future trace moisture measurement was considered. According to the International Technology Roadmap for Semiconductors 2000[10], the level at which trace moisture must be controlled had already been reported to be equivalent to the frost point of -100 ºC or less, and it stated that controlling residual moisture in the lower frost point range will be required in the future. We considered that the reliability of the vapor pressure formula and enhancement factor would become issues when the frost point method is used in this range. The third reason was the development of a new balance called the magnetic suspension balance[11]. This balance became commercially available in the early 1990s, and many research studies using this balance were published from the middle to late 1990s. By 2000, it was demonstrated that this balance has excellent performance and usability, and we considered that the difficulty in measuring the very small evaporation rate in the diffusion tube method could be overcome through the use of the magnetic suspension balance. Finally, as metrologists, we find the clear traceability to the SI of the diffusion tube method very fascinating.

3.2 Development of the primary measurement standard

The amount-of-substance fraction of the trace moisture in the gas generated by the trace moisture generator using the diffusion tube method is given as

$$x_{\mathrm{w}} = \frac{N + N_{\mathrm{b}} + Fx_{\mathrm{b}}}{N + N_{\mathrm{b}} + F} \approx \frac{N}{F} + \frac{N_{\mathrm{b}}}{F} + x_{\mathrm{b}} \qquad (1)$$

where

x_{w}: Amount-of-substance fraction of trace moisture in generated gas [mol/mol]
N: Amount-of-substance of moisture generated as vapor from diffusion tube per unit time [mol/h]
N_{b}: Amount-of-substance of moisture that moves by adsorption/desorption per unit time [mol/h]
F: Flow rate of dry gas per unit time [mol/h]
x_{b}: Amount-of-substance fraction of residual moisture in dry gas (zero gas) [mol/mol]

The strategy of developing the primary measurement standard by the diffusion tube method can be described using Eq. (1) as follows. The measurement method for each of the physical quantities on the right side of Eq. (1) is established. A generator is developed to fulfill the conditions $x_w >> x_b$ and $N >> N_b$ at $x_w \sim 14$ nmol/mol and $N \sim 780$ nmol/h, and Eq. (1) can be approximated as

$$x_w \approx \frac{N}{F} \qquad (2)$$

N and F are stabilized (their variabilities are reduced). A technology is developed to measure N and F with small uncertainty, and the standard value x_w can be determined using Eq. (2).

However, the actual development was not performed in this order. There were several technologies, both major and minor, that had to be developed, and when advances in one technology were achieved, other technologies had to be revised. The development did not progress in one direction only.

3.2.1 Measurement of evaporation rate

N in Eq. (2) can be obtained by measuring the evaporation rate [g/h] and by dividing it by the molar mass of water (18.02 g/mol). As mentioned in section 3.1, we planned to use a magnetic suspension balance for the measurement of the evaporation rate. The magnetic suspension balance is structured so that the diffusion cell placed inside the generation chamber is suspended on an exterior electronic balance by a magnetic force. By using this balance, the mass-loss rate of the diffusion cell owing to water evaporation (evaporation rate) can be measured continuously without interrupting the generation of water vapor. The magnetic suspension balance at AIST was specially customized for the development of the trace moisture standard; the inner surface of the balance in contact with the gas is electropolished because it is part of the generation chamber; the balance can measure a change in mass of the diffusion cell of approximately 12 g at 1 µg resolution; the indication of the balance can be calibrated using external weights traceable to the SI, and so forth. Figure 7(a) shows an example of a measurement using the magnetic suspension balance. It can be seen that a small evaporation rate of 5.1 µg/h can be measured with sufficient accuracy. Figure 7(b) shows an example of a measurement without using the magnetic suspension balance. The generation chamber was exposed to the atmosphere at each mass measurement of the diffusion cell, and the diffusion cell inside the generation chamber was measured by suspending it using a wire connected to the hook of the electronic balance placed above the generation chamber. The diffusion cell used was the same as that shown in Fig. 7(a). For further details of the experiment, refer to Ref. [12]. In this case, an evaporation rate of 6.6 µg/h was measured, and it appeared that this method would suffice except for the fact that it was time-consuming. In fact, the diffusion tube method is often used as a simple trace moisture standard, after the evaporation rate has been determined in advance using gravimetry without using a magnetic suspension balance in a manner similar to that described above. This evaporation rate was used as a constant value under the same experimental conditions, in particular, temperature and pressure. However, from the comparison with values calculated theoretically, a problem was found in the experiment illustrated in Fig. 7(b). Assuming ideal diffusion in the diffusion tube method, the evaporation rate can be calculated from the inner diameter and length of the diffusion tube, the pressure in the generation chamber, and the temperature of the water inside the cell. The details of the calculation method are provided in Ref. [5]. In the case of the experiment illustrated in Fig. 7(b), the theoretical value was 4.1 µg/h, which differed slightly from the measured value. This difference could not be explained on the basis of the uncertainty of pressure, temperature, or other factors. In the experiment using the magnetic suspension balance (Fig. 7(a)), the theoretical value was 5.1 µg/h, which was consistent with the measured value. To confirm that this was not a coincidence, we performed experiments using diffusion tubes with different inner diameters and different pressures and temperatures in the generation chamber, and it was found that the theoretical and measured values were consistent

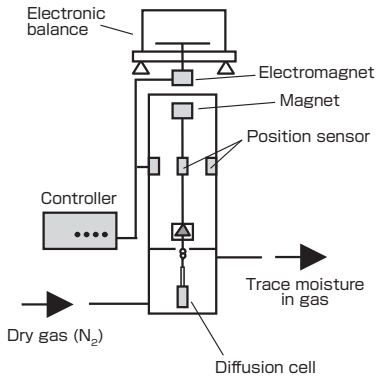

Fig. 6 Schematic diagram of mass loss measurement of diffusion cell using magnetic suspension balance.

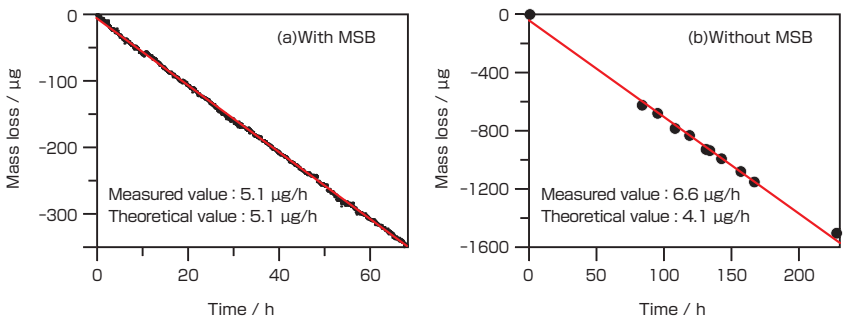

Fig. 7 Data of mass loss measurement of diffusion cell.
MSB stands for magnetic suspension balance.

with each other within the uncertainty[5]. From this result, it was demonstrated that the magnetic suspension balance was extremely effective for reliably measuring an evaporation rate at the 10 µg/h level. Another finding from this result was that the evaporation in the generator could be explained on the basis of the diffusion phenomenon. This indicates that N in Eq. (2) can be sufficiently stabilized by controlling the temperature and pressure. This was also confirmed experimentally, and a stability within a relative standard uncertainty of 0.6 % was realized by precise temperature and pressure control[5]. It is interesting to note that the technology developed for the measurement of N led to the technology required for the stabilization of N.

3.2.2 Measurements of adsorbed and desorbed moisture and residual moisture in zero gas

Measurements of N_b and x_b were necessary to develop a trace moisture generator that fulfills the conditions $x_w >> x_b$ and $N >> N_b$ of Eq. (1) even for x_w~ 14 nmol/mol (ppb) and N~ 780 nmol/h, and a high-performance trace moisture analyzer was necessary for this purpose. To address this issue, we initially considered using the vacuum ultraviolet fluorescent trace moisture analyzer (VUV)[13] developed at the National Research Laboratory of Metrology and the atmospheric pressure ionization mass spectrometer (APIMS) [3] that was introduced in 2000. VUV is an instrument that detects the fluorescence emitted from OH radicals in an electronic excited state produced by the photolysis of water molecules using vacuum ultraviolet absorption. APIMS is an instrument that detects a trace substance (in this case, trace moisture) in a gas by ionizing the sample gas using a corona discharge under atmospheric pressure, and then introducing the gas into a mass spectrometer by differential evacuation. Both VUV and APIMS are sufficiently sensitive in the trace moisture region, but the instrument itself is not scaled, and it is necessary to create a calibration curve using a standard. We considered creating the calibration curve by a standard addition method using trace moisture generated by the diffusion tube generator that was being developed. That is, assuming that N_b and x_b of Eq. (1) maintain constant values, x_w is varied by changing N and F, the indications of VUV or APIMS are recorded, and the calibration curve is created from the relationship of the differences between the N and F values and the indications. However, in the trace moisture range, because a long time was necessary for dry purging (the removal of residual moisture) after exchanging the diffusion cell (to change N), the baseline drift and sensitivity drift of VUV or APIMS during the experiment period could not be ignored because of their non-negligible magnitudes compared with the signal intensity. Moreover, it was unknown how much dry-purging time was necessary to ensure that N_b was the same as that before the exchange, and it was also unclear whether N_b could be considered to be approximately constant after dry purging. Therefore, as the research progressed, we found that it was difficult to create a reliable calibration curve using this method in the trace moisture region.

In the trace moisture region, because we expected that the uncertainty due to adsorption/desorption moisture and residual moisture in zero gas would strongly dominate in the total uncertainty, we considered the measurement of N_b and x_b to be particularly important in developing the generator. As the research progressed, we began to consider that it was problematic to use only VUV and APIMS.

Around that time (about 2002), a new trace moisture analyzer based on the cavity ring-down laser spectroscopy (CRDS)[14]-[17] became commercially available. CRDS is a method where a laser pulse is coupled into an optical cavity composed of two high-reflectivity mirrors, and the detection sensitivity is increased through a long optical path length generated by bouncing the laser pulse back and forth between the mirrors. The sensitivity of this method of absorption spectroscopy has increased markedly in recent years. Because x_w can be calculated directly from the absorption cross section

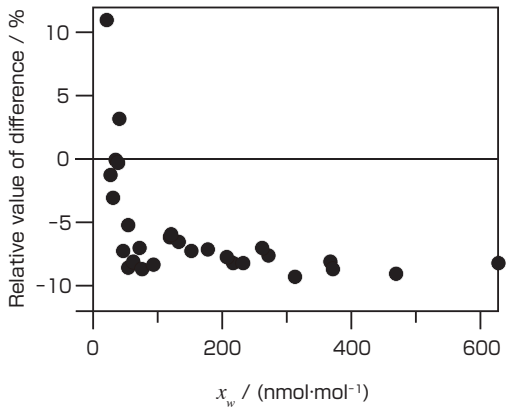

Fig. 8 Difference between indication of CRDS trace moisture analyzer and standard value (relative value).
The dots represent [(indication – standard value) ÷ standard value] × 100.

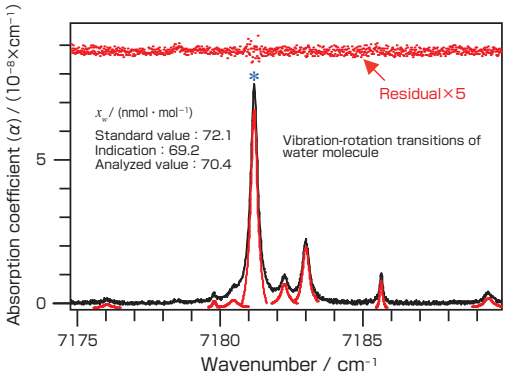

Fig. 9 Near-infrared spectrum of water.
All absorption lines in the figure are assigned to the vibration-rotation transitions of water molecule. The amount-of-substance fraction of water is determined using the $2_{02} \leftarrow 3_{03}$ transition of the $\nu_1+\nu_3$ band that is the strongest in the figure (marked with asterisk).

of the water molecule and measurement data obtained using the CRDS trace moisture analyzer, trace moisture can be measured without a calibration curve. Because this characteristic was considered an advantage over VUV or APIMS, we immediately started to gather information on the instrument performance and introduced it in early 2003. According to the calibration certificate issued by the instrument manufacturer, the accuracy of the indications of the CRDS trace moisture analyzer was confirmed in the range of 0 nmol/mol (ppb) to 1000 nmol/mol (ppb) using a reference standard traceable to NIST of the United States. To check the accuracy of the indications independently, a trace moisture gas with a known value (standard value) generated by the AIST trace moisture generator was measured using the CRDS trace moisture analyzer (indication), and the indications and standard values were compared[5]. Figure 8 shows the differences between the indications and standard values as relative values. In addition, absorption spectra were measured using a built-in function of the CRDS trace moisture analyzer, and x_w was calculated from the analysis of the absorption line (analyzed value). Figure 9 shows a measured spectrum. The absorption lines in Fig. 9 are assigned to the vibration-rotation transitions of the water molecule, and the strongest absorption line shown in the figure ($2_{02} \leftarrow 3_{03}$ transition of $\nu_1 + \nu_3$ band) was used for the analysis. The indications of the CRDS trace moisture analyzer were determined on the basis of the peak intensity of this absorption line. The three x_w values obtained independently agreed within 11 % in the range of 20 nmol/mol (ppb) to 600 nmol/mol (ppb), and it was found that the CRDS trace moisture analyzer can be used for the measurement of N_b and x_b. Using the CRDS trace moisture analyzer, we found methods of achieving $x_b < 0.15$ nmol/mol (ppb) and $N_b < 6$ nmol/h and of establishing a technology to fulfill the conditions $x_w >> x_b$ and $N >> N_b$. In the process of this research, it was also demonstrated that the CRDS trace moisture analyzer was a high-performance measuring instrument.

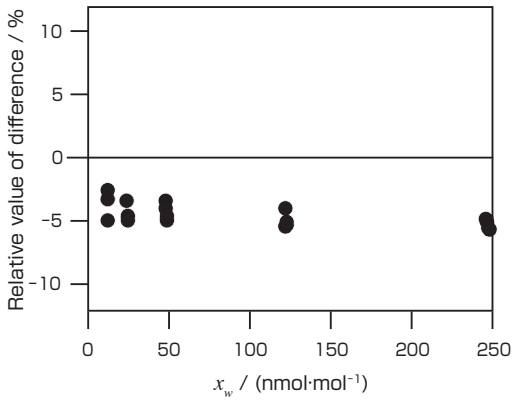

Fig. 10 Difference between indication of CRDS trace moisture analyzer and standard value after introduction of critical flow Venturi nozzle flow meter (relative value).
The dots represent [(indication – standard value) ÷ standard value] × 100.

3.2.3 Measurement of dry gas flow rate

The flow rate F of the dry gas (nitrogen) is controlled and measured using a thermal mass flow meter. We initially considered calibrating the flow meter to obtain the traceability to the SI using the Japan Calibration Service System (JCSS)[18], introduced in Japan by the Measurement Act. In late 2004, we asked the technical staff of the calibration laboratory accredited by JCSS who was capable of calibrating flow meters with the smallest uncertainty, but they replied that they had not yet had any experience of calibrating the thermal flow meter, and that they were not yet confident in performing the calibration. At that time, the accuracies of thermal flow meters stated by the manufacturers were generally 1 % of the full scale, and the uncertainty increased markedly in the small flow range relative to the full scale of the flow meter. This meant that it was necessary to set up several flow meters and calibrate all of them in order to cover the entire flow range with the small uncertainty needed in this research. Moreover, we considered that the differences in the indications and standard values observed in Fig. 8 originated mainly from the uncertainty of the flow rate measurement. For the above reasons, from about the first half of 2005, we started to consider using a flow meter with a high reliability based on a different principle.

For the measurement of a small gas flow, the most reliable flow meter is considered to be a critical-flow Venturi nozzle (sonic nozzle) mass flow meter[19][20]. The critical-flow Venturi nozzle flow meter was developed through joint research between the National Research Laboratory of Metrology and a flow meter manufacturer[21] and it became commercially available in around 2000. We introduced this type of flow meter in 2005, and the traceability to the SI was established using the JCSS. The flow rate was calculated independently using the measurements of the pressure and temperature of the gas and the information on the size of nozzle, and the operating principle was checked through the comparison between the calculated values and the indications of the flow meters. From experiments using these flow meters and by adding some improvements in controlling the flow rate, it

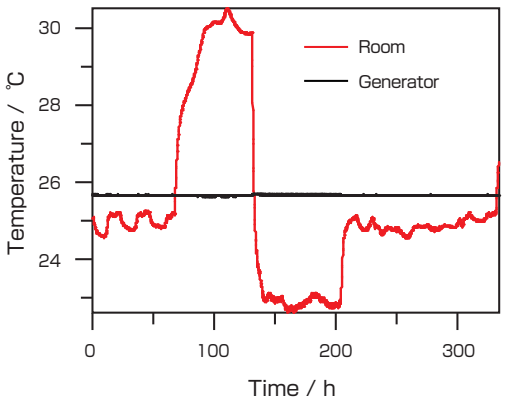

Fig. 11 Change of generation chamber temperature against change of room temperature.

was confirmed that flow rate control at a standard uncertainty of within 0.15 % was achievable[22]. Figure 10 shows the result of the comparison between the standard values of the trace moisture after introducing the critical-flow Venturi nozzle flow meters and the indications of the CRDS trace moisture analyzer. As in Fig. 8, the differences between the indications and standard values are shown as a relative value. The figure shows that the differences are within 6 % and are almost constant, in contrast to the result shown in Fig. 8 where the maximum difference reached 11 %. The remaining difference in Fig. 10 can be explained on the basis of the uncertainty due to the effect of the temperature on the absorption cross section and that due to the inaccurate value of the absorption cross section used in the CRDS trace moisture analyzer[22].

3.3 Evaluation of uncertainty

The standard uncertainty $u(x_w)$ of x_w can be expressed as follows, assuming that there is no correlation between the physical quantities on the right side of Eq. (1):

$$u(x_w) = \sqrt{\left(\frac{u(N)}{F}\right)^2 + \left(\frac{u(N_b)}{F}\right)^2 + \left(\frac{Nu(F)}{F^2}\right)^2 + u^2(x_b)} \quad (3)$$

$u(A)$ on the right side represents the standard uncertainty of physical quantity A. Moreover, $u(A)$ can be expressed as

$$u^2(A) = c_1^2 u^2(a_1) + c_2^2 u^2(a_2) + \cdots \quad (4)$$

where c_i is the sensitivity coefficient and $u(a_i)$ represents the standard uncertainty of physical quantity a_i. c_i can be determined by theoretical consideration or by experiment. It is also important to identify what physical quantities should be included in Eq. (4) as $u(a_i)$. Minor uncertainties can be ignored. However, it often takes considerable time to determine which uncertainties can actually be ignored. There are also cases where an uncertainty component considered to be negligible was in fact not negligible. In one example, we observed variability in the evaporation rate when measurements were performed several times, but this variation could not be explained on the basis of the variabilities of the pressure and temperature in the generation chamber. After gathering long-term data, we found a correlation between the evaporation rate and the room temperature, and we performed an experiment where the temperature of the room was varied intentionally using an air conditioner. Figure 11 shows the temperatures of the room and generation chamber recorded during the experiment. Although the temperature of the chamber appeared to remain constant even if the room temperature was varied, the evaporation rate measured using the magnetic suspension balance depended on the room temperature, as shown in Fig. 12(a). In order to verify that this was not an effect of temperature on the indication of the magnetic suspension balance that biased the measurement value but the effect of temperature on the evaporation rate, we also examined the effect of temperature on the indication of the CRDS trace moisture analyzer shown in Fig. 12(b). The indication of the CRDS trace moisture analyzer clearly depended on the room temperature (the bias of the indication due to the temperature change was compensated), and we concluded that the temperature affected the evaporation rate. The sensitivity coefficients for the change in evaporation rate against the change in room temperature obtained from data shown in Figs. 12(a) and 12(b) were consistent. This phenomenon is probably attributable to unexpected heat transfer, for instance, through the dry gas introduced into the generation chamber. Furthermore, the temperature of the point monitored using a thermometer (controlled to be a set value) may differ from the temperature of the water in the diffusion cell (affected by the variability of room temperature). Further details of this temperature effect were discussed in Ref. [23]. Because it appeared that the temperature of the generation chamber was well controlled, we considered that the temperature of the water in the diffusion cell was also well controlled, and we did not initially recognize this uncertainty component. However, from the above experiment, we found that the uncertainty component due to the variability of room temperature should be included in $u(N)$ as $u(a_i)$, and its

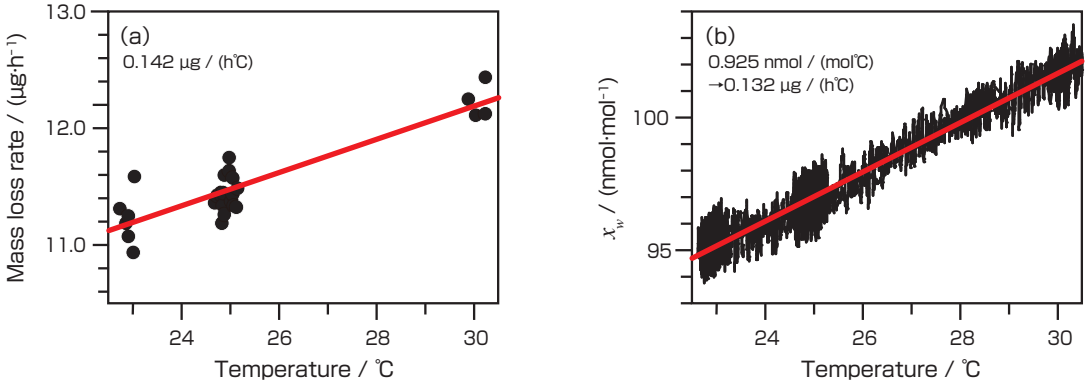

Fig. 12 Room temperature dependency of evaporation rate.
Reproduced with permission from Ref. [23].

sensitivity coefficient was determined from the experiments.

The process of evaluating uncertainty involves completing Eqs. (3) and (4) by similarly evaluating the uncertainties of other physical quantities in the two equations by an experimental or theoretical consideration. In this process, a table of uncertainty evaluation is normally created to organize and visualize the results. The uncertainty evaluation is complete only when this table is filled. The table of uncertainty evaluation that we completed in this research is shown in Table 2 (including uncertainty components in Eq. (3) only). Generally, in establishing a standard, uncertainty evaluation is as difficult as or more difficult than the development of the instruments used in the standard.

3.4 Summary of establishment of the trace moisture standard

From the results shown in sections 3.2.1-3.2.3, we finally succeeded in developing a trace moisture generator that can generate trace moisture in the range between 12 nmol/mol (ppb) and 1200 nmol/mol (ppb) (corresponding to a frost point between -100 ºC and -75 ºC at atmospheric pressure), and we achieved the goal for the generation range. An uncertainty of about 3 % was achieved, as seen in Table 2, surpassing the target of 11 % for the relative standard uncertainty at 12 nmol/mol (ppb). Thus, the trace moisture standard was established as intended.

In this research, we consider that the selection of measuring instruments was extremely important. Without the magnetic suspension balance, CRDS trace moisture analyzer, and critical-flow Venturi nozzle flow meter, we believe that the establishment of the trace moisture standard would have been extremely difficult. One key to success was that, although the CRDS trace moisture analyzer and critical-flow Venturi nozzle flow meter were initially not considered, we were able to notice the problems in the instruments used initially in the relatively early stage of the research and were able to introduce new instruments relatively soon after that. Another key factor was attributable to the effort spent on customizing the instruments used in this research. Although not hitherto mentioned in this paper, we adequately discussed our special requirements with the manufacturers before installing their instruments and also the improvement as needed after installation.

4 Preparation of the calibration service system

On the basis of the trace moisture standard established in this research, AIST initiated the calibration service for measuring instruments in the range between 12 nmol/mol (ppb) and 240 nmol/mol (ppb) (a frost point in the range between -100 ºC and -85 ºC at atmospheric pressure) in May 2007. The calibration range was expanded in May 2009, and we now provide a calibration service for the range from 12 nmol/mol (ppb) to 1200 nmol/mol (ppb) (a frost point in the range between -100 ºC and -75 ºC at atmospheric pressure). The Chemicals Evaluation and Research Institute (CERI) developed a trace moisture generator using a diffusion tube method without a magnetic suspension balance, and using the CRDS trace moisture analyzer as a reference standard for obtaining the traceability to AIST, they initiated a calibration service for the range between 12 nmol/mol (ppb) and 1200 nmol/mol (ppb) in July 2009.

Table 2 Evaluation table for uncertainty of trace moisture generation*.

Standard value	12.00	20.00	50.00	100.00	500.0	1200.0
Uncertainty component						
· Evaporation rate	0.172	0.286	0.715	0.384	1.918	4.602
· Adsorbed/desorbed moisture	0.014	0.024	0.059	0.021	0.104	0.249
· Flow rate of dry gas	0.024	0.040	0.099	0.198	0.990	2.376
· Residual moisture in zero gas	0.300	0.300	0.300	0.300	0.300	0.300
Combined standard uncertainty	0.35	0.42	0.78	0.53	2.2	5.2
Relative combined standard uncertainty [%]	2.9	2.1	1.6	0.53	0.44	0.43

*All units are in nmol/mol (except relative combined standard uncertainty)

5 High-performance trace moisture analyzer

Even if a trace moisture standard and calibration service are available, highly reliable measurement cannot be realized without a high-performance trace moisture measuring instrument, and the task of finding such an instrument is necessary. Here, the results of a preliminary performance test using the AIST trace moisture standard on some commercially available trace moisture analyzers are described.

5.1 Performance test on CRDS trace moisture analyzer

As mentioned in section 3.2.2, in the process of establishing the standard, we found that the CRDS trace moisture analyzer has an excellent performance in measuring trace moisture. Figure 13 shows an example of the measurement of trace moisture using the CRDS trace moisture analyzer. It is demonstrated that the CRDS trace moisture analyzer has good response and stability. The result also demonstrates the good response and stability of trace moisture generation using the AIST standard generator. It was confirmed that the CRDS trace moisture analyzer is capable of measuring trace moisture as low as about 1 nmol/mol (ppb) with excellent linearity in the range between 12 nmol/mol (ppb) and 1400 nmol/mol (ppb) from the comparison with the standard.

5.2 Performance tests on other commercial measuring instruments

A similar performance test was carried out on some conventional chilled-mirror hygrometers. The tested hygrometers were the products of two companies, and the lower limit of the measurement range specified in the catalogs was a frost point of -100 ºC (14 nmol/mol (ppb)) or less for both products. The results are shown in Fig. 14. In this test, the product of Company A presented problems

in the accuracy and stability of the indications, while the product of Company B showed problems in response and accuracy. Because a chilled-mirror hygrometer can directly measure the frost point, it has the highest reliability among instruments measuring humidity. However, it was demonstrated from this result that the measurement in the trace moisture range was not straightforward, even with the chilled-mirror hygrometer.

Performance tests were carried out on some conventional aluminum oxide capacitive sensors that are widely used in industry. The products of five companies were tested, and the lower limit of the measurement range specified in the catalogs was a frost point of -100 ºC (14 nmol/mol (ppb)) or less for all the products. The flow rate of gas introduced into the sensor was 0.6 L/min to 1.0 L/min, and dry purging (the removal of residual moisture) was performed for over one month before the test using the trace moisture of 12 nmol/mol (ppb). The long purging period was set to investigate the performance under actual conditions, because conventional aluminum oxide capacitive sensors are often used as an alarm indicating the presence of moisture in the processes that involve drying, and the sensors are exposed to an extremely dry atmosphere for a long time. The test was performed in the range of between approximately 12 nmol/mol (ppb) and 100 nmol/mol (ppb) (a frost point between about -100 ºC and -90 ºC). The product of one of the five companies indicated an error, that the level of moisture was below the measurement range, and measurement was not possible. The results of the products of the other four companies are shown in Fig. 15. The sensors exhibited problems in response, accuracy, and sensitivity of the indications. Similar tests were performed in joint research between BOC Edwards, Ltd. and NPL, and it was reported that some sensors required 8 h or more to respond to the moisture concentration change of 300 nmol/mol (ppb) → 850 nmol/mol (ppb)[24]. Because the conventional aluminum oxide capacitive sensor uses the equilibrium of moisture adsorption in the pores of aluminum oxide, it is deduced that the slow response is responsible for the long time required to reach the equilibrium state in the trace moisture range. Therefore, technological development to increase the response is necessary for the measurement using conventional aluminum oxide capacitive sensors in the trace moisture region.

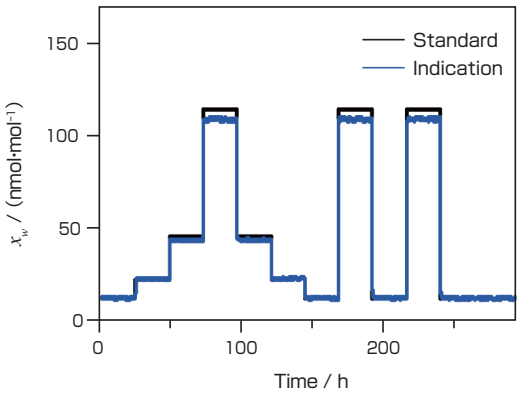

Fig. 13 Comparison of standard value and indication of CRDS trace moisture analyzer.

Previously, there was no trace moisture standard, and it was difficult to change the moisture concentration quickly in the trace moisture region. Therefore, there was no method of reliably testing the performance of trace moisture analyzers, particularly in terms of response, but recently, this has become technologically possible owing to the development of the trace moisture standard and trace moisture generation technology.

6 Future developments

Trace moisture measurement traceable to the SI was realized by establishing the national standard, the commercial availability of the CRDS trace moisture analyzer, and organizing the calibration service system. The results of the performance tests presented in chapter 5 clearly show that the reliability of the trace moisture measurement has increased markedly in recent years. However, a CRDS trace moisture analyzer is necessary to perform such measurements, which not all people can easily access because it is an expensive instrument. Therefore, from the perspective of the dissemination of highly reliable trace moisture analyzer, there are still issues concerning the commercial availability of a measuring instrument. To solve these issues, we must wait for the development of low-cost high-performance trace moisture analyzers. Manufacturers are advancing

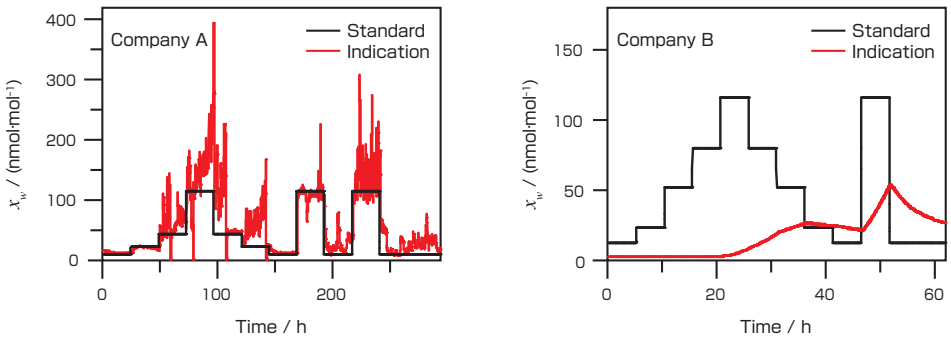

Fig. 14 Comparison of standard value and indication of chilled-mirror hygrometer.

the research and development of measuring instruments to increase the response of sensors based on the adsorption of water molecules, and some sensors have already been commercialized. Increasing the performance and lowering the cost of these sensors are awaited. For the development of other trace moisture analyzers, a specialized Fourier transform infrared spectrometer (FTIR) for trace moisture measurement has been recently developed and marketed, and the appearance of trace moisture analyzers using absorption spectroscopy techniques other than CRDS is also awaited. In this research, only a few of the conventionally used trace moisture analyzers were tested, and there may be high-performance trace moisture analyzers among those not tested in this research. In order to demonstrate the performance of an instrument, regardless of whether it is a new or conventionally used analyzer, the author considers that it is crucial to carry out performance tests (particularly for response, sensitivity, and drift) using a method traceable to the trace moisture standard. Clearly presenting the test results to other users will lead to the dissemination of a highly reliable trace moisture analyzer.

All the topics in this paper pertain to the trace moisture measurement in nitrogen, but the demand for trace moisture measurement in gases other than nitrogen is increasing in the semiconductor industry. However, there is no trace moisture standard for other gases at present, and it is necessary to consider this issue.

Recently, trace moisture standards have been established in countries other than Japan, and these standards are currently being developed or maintained by individual countries. In general, the equivalence of national standards maintained by different countries is confirmed through an international comparison. A preliminary international comparison has been initiated recently for the trace moisture standard. Four national metrology institutes, namely, the National Metrology Institute of Japan (NMIJ), National Physical Laboratory (NPL) of UK, National Institute of Standards and Technology (NIST) of the USA, and Physikalisch-Technischen Bundesanstalt (PTB) of Germany, are participating in this international comparison, which is now in progress (as of July 2009). The result will be reported separately. Because an international comparison is extremely important for further increasing the reliability of the AIST trace moisture standard and for its recognition by other countries, we plan to participate actively in such endeavors in the future.

Acknowledgements

I thank the staff and researchers of NMIJ: Hiroshi Kitano, Chief of Humidity Standards Section; Dr. Chiharu Takahashi (former Head of Temperature and Humidity Division); Masaaki Ueki, Senior Research Scientist; Dr. Shin-ichi Nakao (former Chief of Flow Standards Section); Michito Imae, Head of Time Frequency Division; Dr. Hideyuki Tanaka, Researcher; Dr. Naoki Kuramoto, Researcher; Dr. Nobuhiro Matsumoto, Researcher; and all other people who cooperated and gave us advice on the development of the instrument and the uncertainty evaluation. I am grateful to the technical staff of Rubotherm GmbH, Tiger Optics LLC, K.K. Hirai, DKK-Toa Corporation, Wakana Design Office,

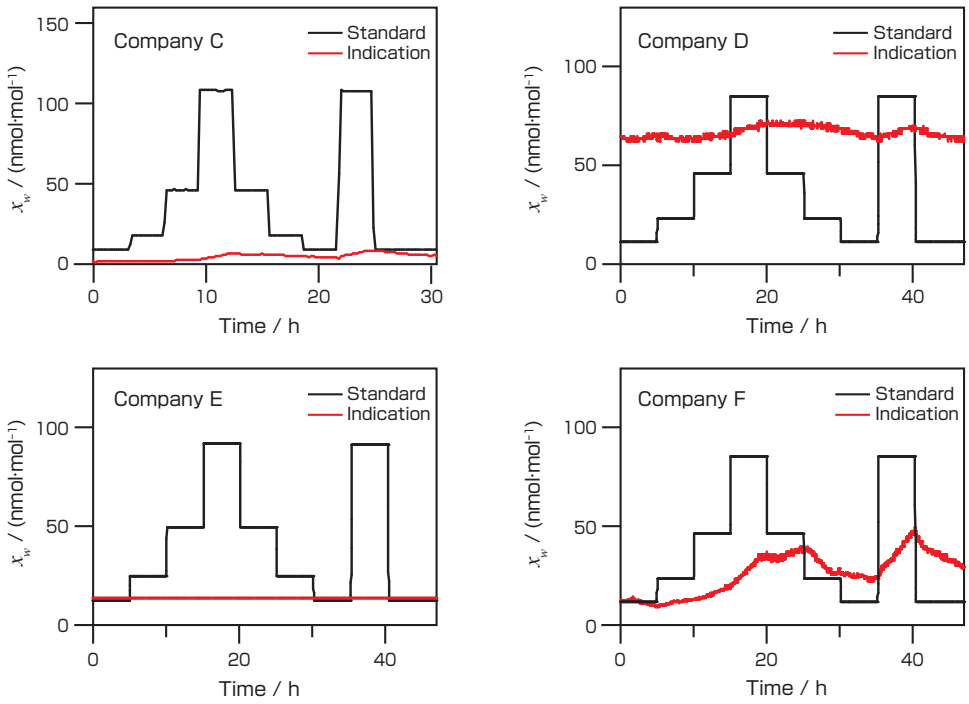

Fig. 15 Comparison of standard value and indication of conventional aluminum oxide capacitive sensor.

Horiba STEC Co., Ltd., and Fujikin Inc., who cooperated in the development and improvement of the instruments. I am also thankful to Mr. Masaaki Maruyama of the Chemicals Evaluation and Research Institute, who helped organize the calibration service system. I thank all those who exchanged opinions about the performance of the conventional trace moisture analyzers.

Terminology

Term 1. Frost point: The temperature at which condensation occurs when a humid gas is cooled under constant pressure is called the dew point. The dew point is lower when there is less vapor content of the gas. It is called a frost point when frosting occurs below a freezing point.

Term 2. Primary measurement standard: A standard that is established without relation to a measurement standard of a quantity of the same kind. It has the highest metrological quality and must be given a complete table of uncertainty according to the SI.

Term 3. Enhancement factor: A coefficient to compensate for the change in vapor pressure caused by the presence of dry air; in the presence of dry air, the vapor pressure obtained is different from that when there is vapor only, because of the additional pressure generated by the dry air, dissolution into water, and interaction with water molecules.

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Discussion with Reviewers

1 Reliability of the vapor pressure equation of ice

Question and comment (Akira Ono, Vice President, AIST)

In many national metrology institutes in countries other than Japan, the national standard for trace moisture is created using the Sonntag's vapor pressure equation for ice. When deriving the Sonntag's equation in the first place, absolute measurements of the trace moisture must have been performed in a manner traceable to the SI. What was the method? Is there an uncertainty attached to the Sonntag's equation?

In this research using a diffusion tube method, the absolute measurement of trace moisture was performed in a manner traceable to the SI. What are the possibilities of investigating the adequacy of Sonntag's equation, or deriving a new vapor pressure equation for ice with an uncertainty using this method?

Answer (Hisashi Abe)

Many of the national metrology institutes other than that in Japan generate a humid gas so that a frost point becomes a set value and constant using a frost point method, and therefore, the traceability to the SI is through temperature. The Sonntag's vapor pressure equation for ice is used when expressing the amount of moisture in a gas as an amount-of-substance fraction and for the calculations when changing the frost point by varying the gas pressure.

The Sonntag's vapor pressure equation for ice was published in Paper [8] in 1990, and this was a recalculation of the existing Wexler's vapor pressure equation for ice (*J. Res. Nat. Bur. Stands.* 1977, vol. 81A, pages 5-20) in accordance with ITS-90, which is the current international temperature scale. Wexler's equation was calculated on the basis of IPTS-68, which was the international practical temperature scale in 1977. Both the Sonntag's and Wexler's equations are obtained by integrating the Clausius-Clapeyron equation. The gas constant and compressibility factor for saturated water vapor over ice needed for the calculation were taken from the values recommended by the Committee on Data for Science and Technology (CODATA) and the values reported in the literature. For details of the calculation, please refer to Wexler's paper.

The relative standard uncertainty of the Sonntag's equation is reported to be less than 0.5 % in the -100 ºC to +0.01 ºC range[8]. Comparisons with experimental data have been performed[9], but relatively large deviations are observed in the trace moisture range (frost point of about -75 ºC or less).

The verification of Sonntag's equation is possible to some degree by comparing the experimental results of AIST's diffusion tube method and the experimental results of the frost point method obtained by NPL and NIST through the international comparison that is currently in progress. This is because, as mentioned above, Sonntag's equation is used when converting the results of the frost point method to the amount-of-substance fraction for comparison. I say "to some degree" here because the verification by this alone is insufficient; the uncertainty of the international comparison (combining the uncertainties of the standards of the two national metrology institutes and the uncertainty caused by the comparison) is larger than the uncertainty of Sonntag's equation in the trace moisture range that requires verification. In this international comparison, we question the validity of Sonntag's equation as a possible factor only when the equivalence of the AIST standard and the NPL or NIST standard cannot be confirmed within the uncertainty.

The derivation of the vapor pressure equation for ice using the diffusion tube method is possible by measuring the frost point of gas generated by the diffusion tube method. However, considering the accuracy of the frost point measurement and the current uncertainty of the diffusion tube method, it is difficult to obtain significant results. I think there is a better possibility of deriving the equation by some other method such as simultaneous measurements of ice temperature and vapor pressure.

2 Calibration for matrix gases other than nitrogen

Question and comment (Akira Ono)

In this research, you established the trace moisture standard by the diffusion tube method using nitrogen as a matrix gas, but what other gases can be used as the matrix gas? For other matrix gases, will it be necessary to individually establish the trace moisture standard using diffusion tube methods? If the standard can be established for one matrix gas (such as nitrogen), can the standard be set easily by relative measurement against other types of matrix gas?

Answer (Hisashi Abe)

In the field of semiconductor manufacturing, matrix gases other than nitrogen that require the trace moisture standards include hydrogen, argon, helium, oxygen, ammonia, and so forth. Although I think it is possible to establish their trace moisture standards and provide a calibration service using the diffusion tube method (although a method must be devised for a gas that is highly soluble in water such as ammonia), it seems not to be highly realistic considering the development cost and user's convenience. Rather, I think it is more realistic to perform a relative measurement to obtain a conversion factor as you mentioned in your question. Please also refer to the second half of the answer to the next question.

3 Use of cavity ring-down spectroscopy for various matrix gases

Question and comment (Akira Ono)

It appears that the absolute values determined using cavity ring-down spectroscopy are consistent with the standard values obtained using the diffusion tube method. Does this mean that the absolute value of the absorption cross section of the water molecule has already been accurately obtained at the wavelength of the laser?

It is mentioned that the absorption cross section is considerably temperature-dependent, but what do you think of the degree of change caused by different matrix gases? If the dependence of the absorption cross section on the matrix gases is small, then can you use the CRDS trace moisture analyzer for measuring trace moisture in other matrix gases if the analyzer was calibrated against a gas generated using the diffusion tube method?

Answer (Hisashi Abe)

A number of studies have reported the line strength of the absorption line monitored in this research used for determining

the water concentration, but there is variation in the values. For example, a difference of approximately 6 % was reported between a value in one study and another value cited in the same study for comparison (*JQSRT* 2005, vol. 94, pages 51-107). Similarly, the maximum difference is 20 % or more in another study (*JQSRT* 2002, vol. 75, pages 493-505). Therefore, rather than already having a sufficiently accurate value, I have an impression that the CRDS trace moisture analyzer used a value which happen to agree relatively well with the standard values. I hope that, by feeding back the results obtained using a method traceable to the SI, the uncertainty of the absorption cross section will be reduced, and the reliability of trace moisture measurement using cavity ring-down spectroscopy will increase further.

The matrix gas dependence of the absorption cross section is not small, for example, there is a twofold difference in values for the absorption line between nitrogen and oxygen (*Appl. Opt.* vol. 44, pages 611-619). However, this relationship can be considered to be almost constant in a small temperature range, and therefore, once the conversion factor is accurately determined at a temperature around which measurement is performed, only the calibration of the instrument for nitrogen will be necessary, and the measurement of trace moisture in oxygen can be performed using the conversion factor. By this method, for matrix gases for which the conversion factor can be obtained with small uncertainty, the reliability of the measurement can be maintained even if there is no trace moisture standard for those matrix gases. Furthermore, the users can skip the calibration for those matrix gases and reduce the management cost. Therefore, I think this is an effective method of trace moisture measurement in gases other than nitrogen. However, when the absorption line of a matrix gas lies in the same wavelength range as the absorption line of water (such as in the case of NH_3), we need to develop an analysis technology to compensate for this effect and to extract information on the moisture concentration. Depending on the type of gas, measurement cannot be performed at all because the absorption of the matrix gas is too strong, and this method may not be effective for all gas types. It is necessary to investigate for which type of gas this method is applicable by considering the uncertainty and the experimental difficulties of determining the conversion factor.

4 National measurement standards and commercially available measuring instruments

Question and comment (Akira Ono)

By using the results of this research, it was clarified that many of the trace moisture analyzers that are already commercially available do not meet the specifications. I think this is an example where a significant social loss may occur owing to the unreliability of commercially available measuring instruments whose measurement capabilities are not adequately demonstrated because of the lack of the corresponding national measurement standard. What lesson should we learn from this?

Answer (Hisashi Abe)

Regarding the conventional commercially available measuring instruments, my understanding is that this situation arose because the instrument manufacturers and retailers endeavored to meet the demands of users who were actually seeking a measurement method of trace moisture, and this situation continued without the national standard for a long time. I think this must be considered as a serious matter, and we must strongly recognize the social importance and impact of standards. Considering the situation of the trace moisture standard of other countries and the period when the CRDS trace moisture analyzer, which was essential for realizing the standard using the diffusion tube method, became commercially available, I think it was technologically difficult to establish the trace moisture standard before 2001. Ideally, even in such a situation, there should have been a shared awareness among the users, instrument manufacturers and retailers, and researchers of the national metrology institute concerning the technological reasons that prevented the development of the standard and the reliability of the trace moisture measurement without the standard. This is not just the case for trace moisture measurement but can be said to apply to other measurements in a similar situation. How can the three parties share a similar awareness? In particular, this is very difficult to achieve in a situation wherein the researchers have negative opinions (although they are not necessarily correct) about the users' demands. How do we obtain opportunities to publish and discuss such negative matters? It is not easy to bring up negative matters as topics at academic societies and in scientific papers, and even if such a matter is brought up, the probability that this information reaches the users, instrument manufacturers, and retailers is not high. If a technological issue is clarified, it can be published in *Synthesiology* as a research scenario, but even this is difficult without accompanying successful scenarios. One solution is to deepen the understanding of measurement among the users, instrument companies and retailers, and researchers by exchanging opinions using the occasions of visiting the laboratory and by attending the meetings of the Metrology Club organized by the NMIJ. I cannot think of any other particularly effective solution. I am still not fully able to understand what lesson can be learned from this case.

However, concerning the past "social loss" related to trace moisture measurements, I believe the following. Before the trace moisture standard was established, there was no way of objectively investigating the measurement capabilities of trace moisture analyzers, and this fact itself was discovered only after the standard was established. Considering this situation, I think it is a bit too much to say that there was a social loss caused by the unreliable measurements. At the time, no major problems occurred because of the measurements (at least, on the surface), and rather than calling it a social loss, I think we should express it more positively: recently, it has been discovered that there is plenty of room for improvement in trace moisture control in industry.

5 Application of diffusion tube method to organic standard gases

Question and comment (Akira Ono)

In this research, the diffusion tube method was applied to water to generate a standard gas of trace moisture. I think that this method is also applicable to organic substances for the generation of standard gases with a known concentration value. What do you think about this possibility?

Answer (Hisashi Abe)

AIST is the first to succeed in using a diffusion tube method as a primary measurement standard, but this method was originally used as a method for producing a low-concentration standard gas for various substances including organic substances (*Crit. Rev. Anal. Chem.* 2005, vol. 35, pages 31-55). Although there are some limitations, such as it can be used only for substances with a certain degree of vapor pressure at the experimental temperature and for those that do not suffer from the dissolution of the matrix gas, it is generally applicable to substances other than water. In particular, for highly adsorbent substances and those that are unstable and for which long-term storage is difficult, it is difficult to maintain their reliability as a standard gas supplied in a high-pressure container, and the method of continuously generating the standard gas (dynamic method) such as the diffusion tube method is more feasible. The diffusion tube method is introduced in the International Organization for Standardization (ISO 6145-8) as well as in the Japan Industrial Standard (JIS K 0225) as a

simple method to generate a standard gas. However, as found in this research, it should be noted that the reliability of this method in the trace concentration region may not be maintained without using a magnetic suspension balance when the evaporation rate is an extremely small value.

6 Setting goal values for uncertainty

Question and comment (Kazuo Igarashi, Measurement Solution Research Center, AIST)

In chapter 2, you mentioned that the reason for setting 11 % (corresponding to a frost point of 0.5 ºC) as the target value of the relative standard uncertainty near the frost point of -100 ºC is that the goal of NPL for the standard uncertainty at a frost point of -95 ºC was 0.5 ºC at the time. The research yielded the excellent result of maintaining the relative combined standard uncertainty at about 3 % by reducing the uncertainties of many components. However, I would like to know the suitability of the 11 % goal because the method of water vapor generation and the accompanying processes were different.

Answer (Hisashi Abe)

Because the significance of selecting the diffusion tube method would be lost if the uncertainty was much greater than that for the frost point method, to begin with we used the target value of the frost point method for the diffusion tube method. Furthermore, we expected that the residual moisture in the zero gas and adsorption/desorption moisture would be major uncertainties in the diffusion tube method, and for the high-performance purifiers used to remove the moisture for zero gas generation, the catalogs from many manufacturers state that the residual moisture is 1 nmol/mol or less. Assuming that the uncertainty caused by the adsorption/desorption of moisture would be at this level, the combined uncertainty of these components would be 1.4 nmol/mol ($\sqrt{1^2 + 1^2} \approx 1.4$), which corresponds to 10 % at 14 nmol/mol. Considering that other components of uncertainty would be included, we did not think at the time that we could achieve the 11 % figure easily. Note that NPL presently sets the lower limit of standard generation by the frost point method to a frost point of -90 ºC , and reports that the standard uncertainty at -90 ºC is 0.2 ºC (about 3.7 %). Therefore, the target value of 0.5 ºC for a standard uncertainty at -95 ºC initially set by NPL was perhaps slightly too high.

7 Cause of temperature difference in the generation chamber and diffusion cell

Question and comment (Kazuo Igarashi)

The evaluation of uncertainty is performed in section 3.3, and you mention that it arises from the difference between the temperature of water inside the generation chamber and the diffusion cell. It seems that the reasons for the temperature difference may be attributable to the temperature control depending on the flow rate of the nitrogen gas and the low heat conductivity of the SUS stainless steel used as material of the diffusion cell. If you have considered these factors, please describe them, as it will be of use in detailed investigations.

Answer (Hisashi Abe)

I added the assumption of the factors that produced the temperature difference to the text. Investigating the reasons why the room temperature change affects the measurement results is an important issue when considering the validity of the evaluated uncertainty, or the uncertainty of the uncertainty (here, this means the uncertainty of the sensitivity coefficient determined by experiment). However, we have not yet performed experiments regarding this issue or to investigate the flow rate dependence as you indicated, so the clear reason is still unknown. However, for the uncertainty of the sensitivity coefficient, from the discussions in Ref. [23], I do not think that it is particularly large under the current experimental conditions. If the obtained sensitivity coefficient is correct, reducing the uncertainty caused by the change in room temperature is not difficult compared with reducing the uncertainties of other components at this stage, and therefore, we have not performed any additional investigation of the reasons for the cause of the temperature difference.

8 Effect of purity of water

Question and comment (Kazuo Igarashi)

In generating trace water vapor, you analyzed several factors such as the adsorption/desorption of moisture, residual moisture in the zero gas, and the gas flow meter, but you do not refer to the purity of water itself. Is this because you think that it is not a problem as long as the purity level does not affect the vapor pressure?

Answer (Hisashi Abe)

I often receive questions regarding the purity of water, but I did not discuss this in the text because of the space limitation of the journal, so I shall explain it here. In this research, we used highly purified water obtained from an ultrapure water production system. However, the purity of water itself is not a major issue. This is because the evaporation rate is measured as the mass change of the diffusion cell per unit time, and therefore, even if there is an impurity in the water, it will not be an issue as long as it remains in the diffusion cell. If a gas is dissolved in the water, it may become a factor of uncertainty because it evaporates with water, but such a gas is probably removed by the aforementioned ultrapure water production system, and I think that the effect can be ignored. Furthermore, the measurements are performed after placing the diffusion tube inside the generation chamber into which highly pure nitrogen is introduced for a sufficiently long time (normally 10 days or more). Therefore, even if there is a residual gas, it will be replaced by nitrogen. Finally, for the nitrogen dissolved in water, even if it is saturated at a pressure of 150 kPa and a temperature of 25 ºC inside the generation chamber, its amount-of-substance fraction is 0.002 % or less, which can be ignored in this research.

Development of battery-operated portable high-energy X-ray sources

— Innovation in X-ray nondestructive evaluation —

Ryoichi Suzuki

[Translation from *Synthesiology*, Vol.2, No.3, p.237-243 (2009)]

We have developed a practical portable high-energy X-ray source, which can generate high energy X-rays with energies greater than 100 keV enabling the taking of high-definition X-ray transmission images using an R6 (AA) battery as a power source. This result is a consequence of the integration of the compact and energy-saving electron accelerator technologies of AIST and the carbon nanostructure technologies of private companies. In this paper, we discuss these elemental technologies and how to integrate them.

Keywords : X-ray, non-destructive evaluation, battery operated, electron accelerator, energy saving

1 Introduction

In Japan, there are many concrete structures and plant facilities that were built during the period of rapid economic growth, and aging of these structures has become an issue. To continue using the structures safely, it is necessary to deploy nondestructive evaluation in which the evaluation devices are carried onto the site to determine the deterioration condition without destroying the structure. There are various nondestructive evaluation methods using ultrasound, electromagnetic radiation, X-rays, or other radioactive rays. X-ray transmission allows imaging the inside of an object, just as in the roentgen exams of the human body. It is used in various fields since the test results can be seen and understood easily.

However, when using X-ray transmission such as for wastage evaluation of pipes in a plant, high-energy X-ray is necessary for steel pipes, and since the device for X-ray source necessary for such tests is bulky and heavy, testing in small spaces is difficult. Also, an X-ray device with large output has further disadvantages. It requires a long time for the heater or filament to warm up and cannot be used instantly when it is necessary, and its energy consumption is large. In many cases evaluation using X-ray transmission may be dismissed due to poor usability. If such issues are solved, inspections can be done more frequently, thus reducing the possibility for failures and accidents.

The author *et al.* set as an objective the development of a portable X-ray generation device that allows generation of high-energy high-output X-rays of 100 keV or over, and this device should be operable by power sources readily available such as dry cell batteries. As a result, we succeeded in the development of a high-energy X-ray generator operable by batteries using a carbon nanostructure as an electron source. This X-ray generator produces X-rays of 100 keV or over with only one AA battery. In some conditions, 100 or more shots of high-resolution X-ray transmission images can be photographed. This performance is practical for a portable X-ray source. When used, the X-ray transmission evaluation can be conducted by carrying the X-ray source onto the site without an AC source or heavy batteries, and it is hoped to be a new X-ray source for nondestructive evaluations.

In this paper, the development of the battery-operated high-energy X-ray generator is described, and consideration will be made of how the researches that we have been doing produced this result.

2 A battery-operated portable high-energy X-ray source

The X-ray can be generated by accelerating the electron beam emitted from the cathode and injecting the beam onto a metal target. A conventional X-ray tube uses the thermionic emission phenomenon of the filament or heater to obtain thermionic electron emission from the cathode. Because of its structure, one must wait until the cathode temperature becomes constant, electricity must be constantly supplied to the heater, and power is consumed even when the X-ray is not being generated. The usability of the device as a portable X-ray source is compromised due to such issues.

In the X-ray source using carbon nanostructure that was developed by the author *et al.*[1], X-rays at output equivalent or higher than the conventional X-ray source can be generated without the heater or the filament, and the issues of warm up

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and energy consumption can be solved.

The carbon nanostructure used in the newly created X-ray tube was developed by the companies engaging in joint research. The graphene sheet composed of carbon has a coniferous form, and the tip has a nanometer size tubular structure that becomes thicker on the substrate side. It is therefore mechanically stable and the electric field readily concentrates at the tip, and this allows electron emissions with high current density of 100 mA/cm^2 or higher at room temperature.

Initially, an X-ray tube was created as a prototype with this carbon nanostructure as an electron source, molybdenum mesh as an extraction electrode, and a metal plate as a target. However, in this structure, the mesh electrode became hot and emitted gas, and abnormal electrical discharge occurred. The carbon nanostructure had a critical problem where the nanostructure was damaged by discharge and the emission property deteriorated, and this caused frequent abnormal discharges. To overcome this flaw, we designed and fabricated an X-ray tube, using the simulation code used in accelerator development, with a structure in which the electron beam focused on the target efficiently without using mesh electrodes. This X-ray tube is a bipolar X-ray tube where the electron source of the cathode is charged with a negative high voltage and the target of the anode is charged with a positive high voltage (Fig. 1).

The fabrication process of the X-ray tube using carbon nanostructure involved the stabilization treatment of the emission current called aging. Since the abnormal discharge in this phase damaged the electron source, we searched by trial-and-error the processing condition that allowed stabilization of the emission current without discharge. As a result, we obtained a cold cathode X-ray tube that could generate high-output X-rays of almost the equivalent to that of the thermionic electron emission X-ray tube.

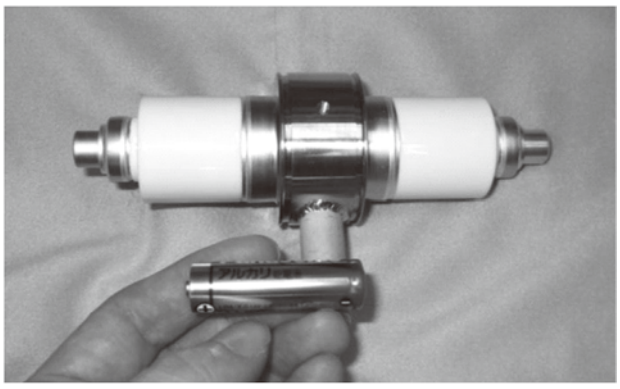

Fig. 1 X-ray tube using the carbon nanostructure electron source.

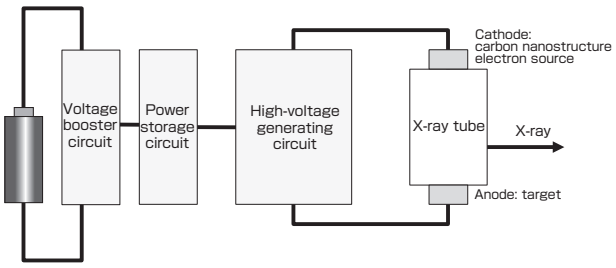

Fig. 2 Diagram of battery-operated X-ray generator.

Since this X-ray tube did not have the heater or filament and this characteristic could be utilized in a portable X-ray evaluation, we created a battery-operated portable X-ray generator as shown in Fig. 2. This X-ray generator produces X-rays by storing power temporarily in the power storage circuit using one AA battery as the power source. X-rays are produced as the X-ray tube is activated by high voltage generated in the high-voltage generating circuit when power necessary to generate X-rays is accumulated. The high-voltage generating circuit can generate voltages of ±50 kV or more and X-rays of 100 keV or more. This X-ray tube has extremely high energy efficiency since the energy consumption falls to almost zero when it is not generating X-rays. Also, it is convenient as a portable X-ray source since it does not require warming up and can generate X-rays immediately. It is also portable since the total weight is 5 kg or less including the power source.

Figure 3 shows the X-ray transmission image of a table tap shot using the X-ray tube activated on 2 J power from the power storage circuit in Fig. 2 supplied to the high voltage generating circuit. The openness of the electrodes in the outlet can be visualized. However, the resolution is insufficient at 2 J, as noises stand out in the fine structures when the irradiated area is large.

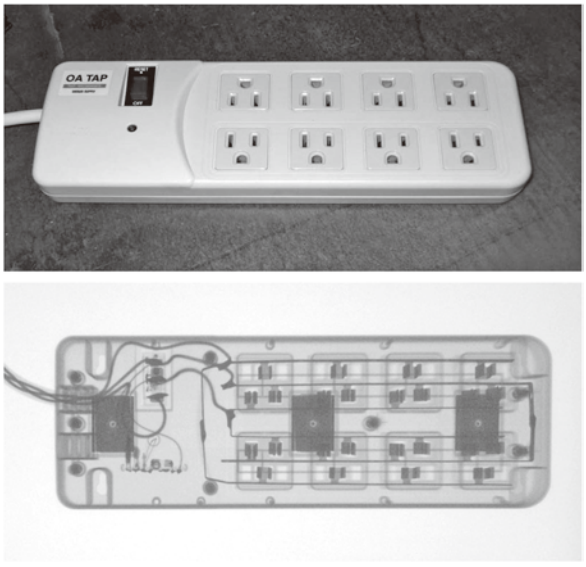

Fig. 3 (Top) Device under test: table tap. (Bottom) X-ray transmission image photographed using the battery-operated X-ray source. Input power 2 J.

Figure 4 is an X-ray transmission image shot by supplying 20 J power to the high voltage generating circuit. The LSI chips in the laptop PC can be photographed at resolutions of 0.2 mm or less. Also, the electrodes in the ceramic insulator about 10 cm in diameter can be seen clearly. With energy of about 20 J, high-resolution X-ray transmission images of various objects can be photographed well for practical purposes. It was confirmed that when X-ray transmission images were photographed at energy of 20 J per shot, over 100 shots could be taken using one AA size nickel metal hydride battery (capacity: 2000 mAh), or over 300 shots with two batteries. The X-ray tube did not show deterioration after X-ray generation for 10^6 shots at 50 J energy per shot. It was confirmed that it can be used as a portable X-ray source without problem.

Moreover, the maximum emission current of this X-ray tube was 50 mA or higher, and X-rays with high output could be generated in a short time. This allows short-time exposure of 1 millisecond or less. Using this high output property, it can be used as an X-ray tube for computer tomography (CT) that requires high output X-rays, as well as for ordinary X-ray transmission image photography.

If the X-ray source technology is likened to a radio, the conventional X-ray source is a vacuum tube radio where one has to mind the battery all the time when carrying it around, while the carbon nanostructure X-ray source is equivalent to a transistor radio, and the portability of the X-ray source increases dramatically. This will allow nondestructive evaluation and diagnosis by X-rays to be done easily on site, and new innovations in X-ray evaluation can be expected.

3 Circumstances leading to achievement of the results

The author *et al.* have been involved in the development and practical application of the electron accelerators, and the development of the new X-ray source described in chapter 2 was realized by fusing the portable ultra-small accelerator and X-ray source technology of AIST and the carbon nanostructure electron source technology developed by companies. Moreover, the technologies for ultra-small accelerators and X-ray sources were based on the technologies for energy saving and downsizing of the electron accelerator. The elemental technologies that became the basis of the development of a new X-ray source are described as follows.

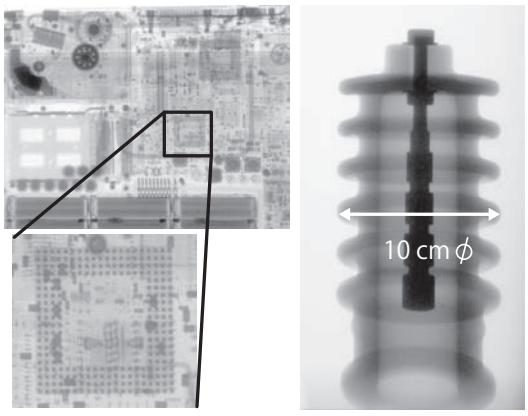

Fig. 4 X-ray transmission image with input power 20 J. (Left) X-ray transmission image of laptop PC. (Right) X-ray transmission image of alumina insulator with test electrode.

3.1 Energy saving in electron accelerator facility

The author *et al.* have been involved in the management, operation, and research using the S-band electron linac (linear accelerator) with maximum energy 400 MeV at AIST. This accelerator was completed in 1979, and has been used as the electron storage ring TERAS for synchrotron radiation, the electron storage ring NIJI-IV for free electron laser, the high-intensity slow positron beam source for material evaluation experiments, and others[2]. Energy-saving measures were conducted for the entire accelerator facility when the aged air conditioning system was renovated in FY 2005.

Before the energy-saving measures were executed, the power consumed when the electron linac was in operation momentarily reached 600 kW, and the annual amount of power used was about 2.5 GWh or more. Estimating the electron linac beam power truly needed during electron injection to the storage ring, it was only 0.01 % of the power consumed when the accelerator was actually in operation, at 320 (MeV) × 100 (mA) × 1 (μs) × 2(pps) = 64 (W). In case of the positron experiment, it was 70 (MeV) × 100 (mA) × 1 (μs) × 100 (pps) = 700 (W) or several hundredth of the actual power consumed.

There were several factors for this extremely low efficiency. The major factor was because originally, the air conditioning and water heating/cooling systems for this electron linac and the accompanying facilities were designed for generation of high-output electron beams of several 10 kW in order to handle various experiments. Therefore, they were not optimized for low-energy modes such as the positron experiment or low pulse rate modes such as injection to electron storage rings.

To solve this issue, total measures were necessary for the electron linac itself as well as its air conditioning and water cooling/heating systems. Therefore, energy-saving measures were considered by combining the accelerator technology accumulated over time and the latest technology for air conditioning, water cooling/heating, and power source systems. In executing these energy-saving measures, the following basic principles were set.

1. Energy is used only for the amount it is needed.

2. Energy is used only during the time when it is needed.
3. Energy is used only at the place where it is needed.
4. Latest technology with high energy efficiency is used.

To determine the specifications of the devices that would be renovated or newly acquired based on the above principles, it was necessary to see when, where, and how much energy (electrical power) was consumed. Therefore, the power consumed in each section was surveyed, and measures were taken within the budget allotted for renovations[3].

The main measures were decentralization of the water cooling/heating and air conditioning systems and the renewal of the high-power microwave generator of the electron linac. By estimating and considering the consumed power under various experimental conditions for the water cooling/heating and air conditioning systems, it was found that drastic reduction in power consumption could be achieved in a decentralized system rather than a centralized one. Therefore, a decentralized system was introduced that allowed ON/OFF of the water cooling/heating and air conditioning of different parts according to the type of experiment.

The S-band electron linac used eight 22 MW klystron devices, which were state-of-the-art at the time of construction, to generate high-power microwaves for electron acceleration. Currently, 80 MW klystron with about four times the output has been developed, and three to four old devices were replaced with one newest device. Since the power consumption of the conventional klystron did not stabilize at low pulse rate, pulse rate of 50 pps or over had to be used even when it was for injecting electrons to the storage ring, and therefore, the power consumption became high at about 30 kW per device or about 100 kW for three devices. The newly installed 80 MW klystron could be operated at 2 pps that was more optimal for electron injection to the storage ring, and the average power consumption was reduced to 10 kW or less. In this section alone, the renovation brought about the decrease of energy consumption to 1/10. This allowed reduction in the capacity of water cooling/heating and air conditioning systems, and large-scale energy savings became possible.

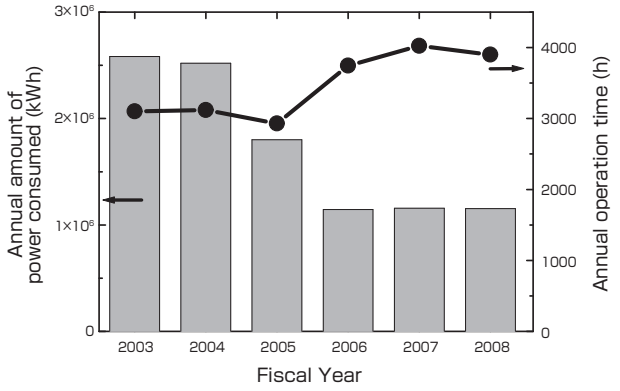

Fig. 5 Annual amount of power used at the electron linac building (Tsukuba Central 2-4 Bldg., AIST) (bar graph) and annual operation time of the electron linac (line graph).

Various other measures were taken based on the above principles, and, as shown in Fig. 5, 60 % reduction in the amount of power used was achieved for the total facility compared to before renovations. Through these measures, the operation time of the electron linac increased and the researches could be conducted more efficiently.

The energy-saving know-hows obtained by being directly involved in these measures led to the development of a low-power driven circuit for an ultra-small electron accelerator and a new X-ray source.

3.2 Development of C-band small electron accelerators

The research using high-intensity slow positron beam was conducted using the aforementioned S-band electron linac. However, since this was a time-shared accelerator, the positron experiment had to be interrupted when the storage ring injection was done, and the time for experiments was limited. Also, since the accelerator was not built for the purpose of positron generation, the generation efficiency of positrons was poor. To solve these issues, an exclusive electron linac was needed, and it was necessary to install a small electron linac with good positron generation efficiency within the limited shielded space.

Moreover, in experiments using the positron beam, high pulse rate of the accelerator was desirable to prevent the detector from becoming saturated. Therefore, we selected the C-band electron accelerator which has a short microwave filling time in the accelerating tube and with which the pulse rate could be set high, and developed the components for this system (Fig. 6)[4]. This C-band electron accelerator, because it had a smaller resonator, had the advantage over the conventional S-band electron linac since the sizes of accelerating tube and waveguide could be downsized.

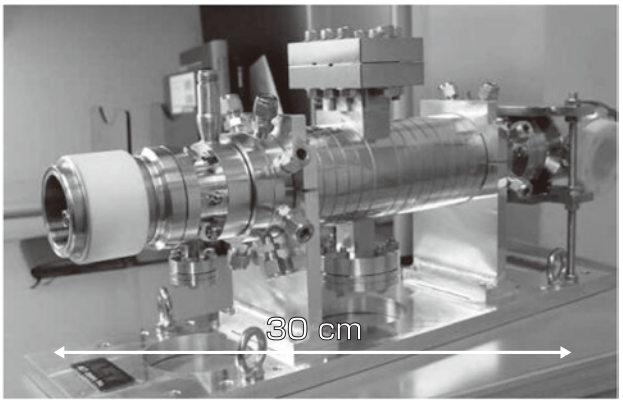

Fig. 6 First part of C-band electron accelerator.

For this accelerator, new circuit using a high-voltage high-current semiconductor switch was developed for the generation circuit of the high-voltage pulses to be supplied to the microwave amplifying tube (klystron). High-power microwaves of about 2 MW were generated by running the C-band klystron with small semiconductor switches of about 18 cm × 7 cm × 7 cm, and the electron beam was successfully accelerated.

The attainment, in the process of R&D of this C-band accelerator, of higher frequency of microwaves, the high-voltage semiconductor switch, and the high-voltage pulse generation was the technological base for realizing the new X-ray source.

3.3 Battery-operated ultra-small electron accelerators

In the 2000s, there was an increase in accidents due to aging of pipes in plants as well as leakage of steam from the pipes in nuclear power plants, and the social demand for on-site evaluation increased. Pipes with covering materials such as heat insulations were extremely troublesome to inspect since the covering had to be removed, and an inspection method without removing the covering material was desired. At that time, the author *et al.* were conducting research on downsizing and energy savings in electron accelerators as described in sections 3.1 and 3.2. As mentioned in chapter 1, thinking that the findings from our research could fulfill the social demands, we did a conceptual design for an electron accelerator and an X-ray source system that pursued ultimate downsizing and energy savings. Here, we returned to the basics to redesign the conventional accelerator technologies such as efficient high-voltage pulse generation, microwave generation, electron generation, and control technology. We developed and fabricated prototypes based on the details obtained, and succeeded in generating high-energy X-rays by operating an ultra-small electron accelerator with AA batteries.

This ultra-small electron accelerator is composed of the electron gun, the accelerating tube, the microwave source, the vacuum pump, the pulse generator, and the control system, just as in large electron accelerators. Since the conventional accelerator had several accelerating tubes, temperature controlled water was necessary to synchronize their resonance frequencies. Since the new accelerator has only one accelerating tube, no cooling is necessary as the heat load is low when operated on dry cell batteries. This accelerator employs a method of synchronizing the resonance frequency by varying the frequencies rather than keeping the resonance frequency constant by controlling the temperature of the accelerating tube. It is a system without a water cooling/heating system that required large amounts of power consumption like in the conventional accelerator. The vacuum pump is an ion pump that uses a little amount of power in high vacuum.

In this accelerator, a 9.4 GHz X-band pulse magnetron tube that has higher frequency than the aforementioned C-band is used as a microwave source. To operate this magnetron and the electron gun of the accelerator, the battery power source is boosted to 12 kV or higher, the electric energy is stored in the storage circuit, and the high-voltage pulses of about 100 kW are generated at intervals of about 1 microsecond using the semiconductor switch. By supplying to the accelerating tube the 9.4 GHz microwaves generated in the magnetron tube with the high voltage pulses, the electron beam is accelerated and a high-energy electron beam of 100 keV or over is generated. X-rays are generated by injecting this electron beam onto the heavy metal target.

Figure 7 shows a photograph of the prototype of the main body of the accelerating tube composed of the electron gun, the accelerating tube, the vacuum pump (ion pump), the X-ray target (gold film), and the X-ray emission window. It is about palm size (the accelerating tube is about 3 cm) and the weight is about 1.5 kg. The flange and valve in the photograph are necessary only in the prototype, and the weight will be reduced to half when they are removed. This main body and the components for microwave and power sources can be fit in a small camera case and carried easily with one hand.

This ultra-small accelerator has a peak electric consumption of 100 kW order, but since the pulse width is 1 microsecond, the average power consumption can be 20 W or less by lowering the pulse rate, and X-rays can be generated with 10 to 12 AA batteries. X-ray transmission imaging is possible by combining this X-ray source with the X-ray imaging system[5][6]. To complete this system, we created the technology to generate high-voltage high-power pulses from dry cell batteries by trial-and-error. This could be applied to the electron emission property tests for carbon nanostructures and X-ray generators mentioned below, and

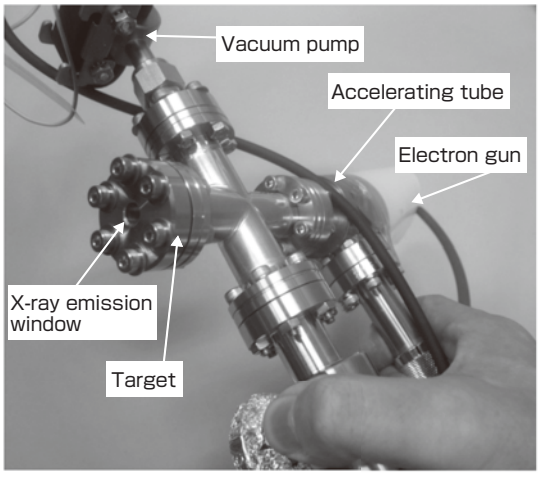

Fig. 7 Main body of the X-band ultra-small electron accelerator.

led to the advancement of the device development in a short time period.

3.4 Application of the carbon nanostructure electron source to the X-ray source

The electron source of the aforementioned ultra-small electron accelerator must have the heater power source ON at all times including when X-rays are not being generated, since it employs the principle of thermionic electron emission, and can operate for only about four hours on 10 AA batteries. Therefore, dry cell batteries tend to be used as emergency power source, and this is not an X-ray source that could be truly used any time, anywhere. This problem of thermionic electron emission electron sources does not only apply to high-frequency electron acceleration but also applies to general portable X-ray sources.

This issue can be solved if there is a high-performance electron source that does not require a heater or a filament, but carbon nanotubes (CNT) that produce electron emission at room temperature have a disadvantage in that the structure is destroyed and deteriorates readily under a strong electric field like X-ray tubes[7]. Therefore we looked for a cold cathode electron source with high stability under high electric field, and focused on a carbon nanostructure (CNX) electron source developed by certain companies. This electron source had a coniferous form that became thicker on the substrate side, and the tip was a nanometer structure as in CNT and the electric field concentrated at the tip. Since it was considered to be more stable in high electric fields than CNT, we expected it to be a hopeful X-ray source.

Therefore, we started the product development in July 2008, after checking its function in a preliminary experiment using the CNX electron source. Other carbon-based cold cathode electron sources were commercially available, but we selected the CNX electron source because the company brought the manufacturing machine for the electron source to AIST, so an environment that allowed free trial-and-error became available for the development of the X-ray source. Experiments under various conditions were possible, and the new X-ray generator described in chapter 2 was realized.

4 Discussion

Here, the findings from the development of the battery-operated high-energy X-ray generator are discussed.

The development of the battery-operated high-energy X-ray generator using carbon nanostructures was realized through the combination of the researches for energy saving and downsizing of the electron accelerator and the technology for a battery-operated ultra-small electron accelerator that were being conducted at AIST, and the technology for the carbon nanostructure electron source of private sector companies.

The motivation for the downsizing research of electron accelerators was from the necessity that arose in the course of research, to solve the issues of the large electron accelerator that was owned by AIST. We became aware that the energy saving and downsizing of the accelerator might find wide industrial applications in nondestructive evaluation, medicine, and sterilization, and set them as new development topics. The development of the components for small C-band accelerators led to the development of the energy saving technology and of ultra-small accelerators. In the C-band small electron accelerators and the X-band ultra-small electron accelerators, the human resources and facilities including those for the conventional S-band electron accelerators, radiation detection technology, and radiation-controlled area were greatly useful. This result would not have been achieved without the large electron accelerator facility.

For energy savings in accelerators, we were able to execute various energy saving measures, rather than leave them as mere desk plans, because we ran into an opportunity of renovating the aged air conditioning and water cooling/heating systems. The measures included experimental ones that would not have been employed in an ordinary electron accelerator system. We were able to accumulate the technologies and know-hows for energy savings in accelerators by observing the effectiveness of the various measures.

The development of the X-band ultra-small electron accelerator was the result of the combination of the social demand for a portable high-energy X-ray source and the necessity for downsizing and energy savings in the electron accelerator at AIST. Although this accelerator had a heater for thermionic electron emissions, and could not be called a truly practical portable X-ray source, by presenting our technological level by publishing our result to the outside world, we were able to find a new technology called the carbon nanostructure electron source. Moreover, the companies worked on this development with passion, brought the manufacturing machines for the electron source to AIST, and through repeated trial-and-error and concentrated effort, we were able to develop a truly practical X-ray generator in just half a year.

The results of this research is the result of the integration of various factors in addition to individual technologies, including facilities, people, change in the research environment, accumulation of technology, social demands, and the publication of the results. However, the factors do not lead to new results if they are simply collected. For example, if the linac was in operation without any problem, one would not think actively about energy savings or downsizing or employing an electron source with carbon nanostructure, and the developments that followed might not have occurred.

Actively looking for incompleteness or problems in elemental technologies and solving problems one at a time by incorporating different elemental technologies, may lead to new results because the linkages with other elemental technologies become clearer and stronger.

5 Summary and future issues

This paper described how the elemental technologies linked together toward the development of an X-ray source that operates on one AA battery and that can photograph high-resolution X-ray transmission images. The developed X-ray source not only can replace the conventional X-ray source as a single X-ray source, but it can be carried anywhere without warm-up, and progress of new X-ray nondestructive evaluations can be expected. To realize this, it is necessary to create a total system including a detector and a safety device as well as the X-ray source. Also, development of higher energy X-ray source using microwave acceleration techniques is necessary to handle inspections of large-scale structures.

In the future, we wish to work on these topics and continue research that may bring true innovation to the field of X-ray evaluation.

Acknowledgement

The development of the X-ray source using the carbon nanostructure was a joint research with Dialight Japan Co., Ltd. and Life Technology Research Institute Inc., under the 2008 R&D Project (to support small and medium businesses), AIST. Part of the development for the C-band electron accelerator was supported by the Budget for Nuclear Research of the Ministry of Education, Culture, Sports, Science and Technology of Japan, based on screening and counseling by the Atomic Energy Commission. We obtained cooperation of the Electron Accelerator Group, AIST. For energy saving renovation of the electron accelerator, we obtained cooperation of the Research Facilities Department, AIST and the Monitoring Room. Shingo Ichimura, Director, AIST introduced us to the companies manufacturing the carbon nanostructure. I am grateful to all the people who cooperated.

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Author

Ryoichi Suzuki

Completed the master's course at the Graduate School of Engineering, University of Tsukuba in 1987. Obtained doctorate (Engineering) in 1991. Joined the Electrotechnical Laboratory, Agency of Industrial Science and Technology, Ministry of International Trade and Industry in 1987. Became the Senior Researcher of AIST in 2001. Engages in the generation and measurement of high-energy X-ray using the electron accelerator, and the development and application material evaluation technology using the high-intensity slow positron beam. Received the Ohm Technology Award (The Promotion Foundation for Electrical Science and Engineering) in 2004. Also received the Ichimura Award (New Technology Development Foundation) in 2005.

Discussion with Reviewers

1 Original technological development for downsizing the electron accelerator

Question and comment (Hisao Ichijo, Evaluation Division, AIST)

In the description about downsizing the electron accelerator, it cannot be readily understood that it was your original technological development. I think you should cite your papers and patents and emphasize the fact that this was an original research result.

Answer (Ryoichi Suzuki)

In the electron accelerator research field, it has been widely known that downsizing could be accomplished by raising the acceleration frequency, and that itself is not original. However, we were the first to develop a small electron accelerator for generating positrons. I cited the paper that reported this technology at an academic society. Also, since this result is not the main theme of the paper, I added it to the section on the circumstances surrounding this research.

2 Effort on the conceptual design of the battery-operated ultra-small electron accelerator

Question and comment (Hisao Ichijo)

You wrote the "conceptual design" in the description for the development of the battery-operated ultra-small electron accelerator. I think you should include how you utilized the

already accomplished energy saving measures in this conceptual design, focusing on the technological aspect.

Answer (Ryoichi Suzuki)

Although I cannot write the details since it is an intellectual property, I added some descriptions on the technologies we considered in the conceptual design.

3 Application to nondestructive evaluation and issues

Question and comment (Hisao Ichijo)

In the manuscript, I think the originality will become clearer if the author adds his innovative and ingenious undertakings in the last paragraph. Also I think you should describe the possibility of application to nondestructive evaluation and the issues that must be solved.

Answer (Ryoichi Suzuki)

I integrated the wordings in the figures and the text. I added an explanation for the system without cold/hot water in the ultra-small accelerator. In this paragraph and the following one, I added how the topics of ultra-small accelerators and the development of the carbon nanostructure X-ray source linked together.

4 Components of the research

Question and comment (Mitsuru Tanaka, Research Coordinator, AIST)

I believe the components of this research are energy saving, downsizing, and the introduction of electron source. Wide-ranging technologies to support those components were nurtured in the research using the accelerator facility, and the actual measures to meet the powerful demands for energy saving and downsizing spurred the practical application, and the employment of important technology for high-performance electron source from exterior institutes also played the role in spurring the development. I think those were the syntheses of the *Full Research*.

Answer (Ryoichi Suzuki)

It was exactly as you pointed out. I rearranged the paper for better reading.

5 The efficacy of the carbon nanostructure as the X-ray electron source

Question and comment (Hisao Ichijo)

In the manuscript, pertaining to the carbon nanostructure electron source and the X-ray source, you only describe that it is hopeful as an electron source for X-rays, but can you add descriptions and figures that explain how it is effective. Also, if you describe the various methods considered to solve the technological issues, the improvements that were expected by incorporating the carbon nanostructure electron source, and the process that led to this technological integration, I think the selection and integration of the elemental technologies will become clearer.

Answer (Ryoichi Suzuki)

Also considering the comments of Reviewer Tanaka, since this paragraph is the main theme of this paper, I summarized the points and placed them before the development process. I added the experimental results including the figures and provided detailed explanations. For the integration of carbon nanostructure technology, some explanations were added in the section on the circumstances.

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 June 18, 2008
Revised October 24, 2008
Revised March 23, 2009

1 Types of contributions

Research papers or editorials and manuscripts to the "Readers' Forum" should be submitted to the Editorial Board.

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 editorials should also comply with the same structure and format except subtitles and abstracts are unnecessary. Manuscripts for "Readers' Forum" shall be comments on or impressions of articles in *Synthesiology*, or beneficial information for the readers, and should be written in a free style of no more than 1,200 words. Editorials and manuscripts for "Readers' Forum" will be reviewed by the Editorial Board prior to being approved for publication.

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 and editorials shall have front covers and the category of the articles (research paper or editorial) 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 with names of reviewers disclosed, 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. The edited discussion will be attached to the main body of the paper as part of the article.

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.
3.3.2 The text should be in formal style. 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, clear originals that can be used for printing or image files (resolution 350 dpi or higher) should be submitted. In principle, the final print will be 15 cm × 15 cm or smaller, in black and white.
3.3.5 For photographs, clear prints (color accepted) or image files should be submitted. Image files should specify file types: tiff, jpeg, pdf, etc. explicitly (resolution 350 dpi or higher) . In principle, the final print will be 7.2 cm × 7.2 cm or smaller, 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).

4 Submission

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

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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 are allowed in the proofreading stage.

6 Responsibility

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

7 Copyright

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Letter from the editor

Synthesiology Volume 2 Issue 3 has been published. I shall be in charge of the "Letter from the Editor" for this and the next issues.

The synthetic approach to research that is the subject of *Synthesiology* has been discussed intensively in order to set a methodology for the core of the R&D principle of AIST, ever since the establishment of this institution. The need for a journal was indicated from the beginning. About 100 workshops were held and many researchers participated in the discussions, and the range of discussions widened. After spending much time to prepare for the publication of the journal, it was finally launched last year with the effort and cooperation of many people. I am very glad to see that its publication process is going relatively smoothly. As one of the editors of *Synthesiology*, I am grateful to all people involved.

In this issue, papers from diverse research fields are published. The topics include: the rupture probability of active faults, the diffusion of advanced technology to households, the development of IT devices, the development of a device for x-ray nondestructive test, and the performance evaluation of measuring instruments. Conventionally, these papers were submitted to academic journals of their respective disciplines, and could not be easily understood by researchers and engineers of different fields. However, the readers who have read some of the papers of *Synthesiology* may realize that they can not only understand what is written, but may be surprised that they are actually inspired by the articles. These papers hint that there may be a common methodology in synthetic approach to research, transcending the boundaries of the technological fields.

The papers in this issue point to the importance of the role of various stakeholders in synthesizing the individual elemental technologies and then actually applying them to society, sites of production, and people's lives. Specifically, they include the relationship of government and citizen in the active fault research, the role of theoretical lead user and perceptual lead user in the technology used at households, the collaboration with manufacture device companies in the IT device development, the integration of research facilities and people in the development of x-ray nondestructive test devices, and the networking of Japanese and overseas institutions in the performance assessment of measuring instruments. The social diffusion of technology and product was not addressed in the past analytical research papers whose purposes were to develop elemental technologies. We hope that as more results of synthetic approach to research are submitted to *Synthesiology* and diverse results are accumulated, innovations that are necessary to build a sustainable society that is strongly demanded by society can be achieved more efficiently. For this reason, we expect more submissions to *Synthesiology* by the researchers and engineers of industries and universities, as well as from people of AIST.

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