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Development of a framework for risk tradeoff analysis of chemical substance substitution

A first empirical analysis of JIS lifespan

Development of EUPS for analyzing electronic states of topmost atomic layer

A revolutionary technical development to revitalize Japanese forestry

Synthesiology editorial board

2016

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Highlights of the Papers in *Synthesiology* Volume 9 Issue 4 (Japanese version Nov. 2016)

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

***Synthesiology* Editorial Board**

Development of a framework for risk tradeoff analysis of chemical substance substitution
— An approach to risk assessment using relative comparison —
Masashi Gamo *et al.*
While it is important to reduce the risks of chemical substances, if there is a possibility that substitute substances may cause different risks, the way to assess these risks is extremely important. Gamo (AIST) *et al*. present a methodology for introducing a common index to conduct relative quantitative comparison for the size of risks with differing factors, and the way to actually apply this method to risk assessment. The method is applied to existing case studies to actually investigate the effectiveness. These results can be applied to risk assessments of various fields, not limited to chemical substances, and this is a highly significant research.

A first empirical analysis of JIS lifespan
— Implications for the review system of de jure standards —
Suguru Tamura
The Japan Industrial Standard (JIS) is a set of standards and references for various products in Japan, and while it is expected to contribute to innovation, the review of the standards is done according to regular schedule and lacks flexibility. Tamura (Research Institute of Economy, Trade and Industry) statistically calculates how the sustained period of standards differs, by utilizing various data such as the form of standard, technological category, and presence of ISO standards for about 4,500 JIS standards. Based on this result, he discusses the necessities for optimizing the review period of standards that may contribute to innovation and for achieving the efficiency of standard management. It is an insightful paper.

Development of EUPS for analyzing electronic states of topmost atomic layer
— Materialization of laser-produced plasma source application and EUPS observed fascinating surface —
Toshihisa Tomie *et al.*
The topmost atom layer of substances is an important region that controls the catalyst functions, surface reactions, electric conductivity, and others, and its observation and control are immediate issues for many precision material design. Tomie (AIST) *et al.* have, over the years, engaged in the development of extreme ultraviolet-excited photoelectron spectroscopy (EUPS) that uses a plasma source produced by ultra-short pulse lasers, as well as surface measurement technology using this spectroscopy. This paper is a comprehensive report on the development and use of the technology. Specifically, extremely interesting cases of application are shown including observation of trace contamination of the topmost atom layer, a non-charge phenomenon of insulating films, electric conductivity, and catalyst activity. Since characteristic surface analysis that is difficult by other methods can be done, it is hoped that more users will utilize this method in the future.

A revolutionary technical development to revitalize Japanese forestry
— A proposal for a portable tree felling manipulator to address specific properties of Japanese forestry —
Yuko Shirai
Forestry in Japan is facing difficulty to be established as an industry, due to unique natural environment and social systems. Shirai (Waseda University) states the necessity of technological development that enables industrial autonomy, and presents demonstration experiments of an originally developed tree-felling manipulator and its potential for utilization in the industry. The paper is characteristic because the specific scenario and course are laid down for the revitalization of forestry, and a unique “technological design theory that thoroughly emphasizes the interaction with environment” is developed. The methodology can be applied to various other fields in the future, and is very interesting.

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Synthesiology – English edition Vol.9 No.4 (Mar. 2017)

Contents

Highlights of the Papers in *Synthesiology*

Research papers

Development of a framework for risk tradeoff analysis of chemical substance substitution 185 – 196
— *An approach to risk assessment using relative comparison* —
- - - M. Gamo and J. Takeshita

A first empirical analysis of JIS lifespan 197 – 212
— *Implications for the review system of de jure standards* —
- - - S. Tamura

Development of EUPS for analyzing electronic states of topmost atomic layer 213 – 233
— *Materialization of laser-produced plasma source application and EUPS observed fascinating surface* —
- - - T. Tomie and T.Ishitsuka

A revolutionary technical development to revitalize Japanese forestry 234 – 251
— *A proposal for a portable tree felling manipulator to address specific properties of Japanese forestry* —
- - - Y. Shirai

Editorial policy 252 – 253

Instructions for authors 254 – 255

Vol.9 table of contents (2016) 256 – 257

Letter from the editor 258

Aim of *Synthesiology*

Development of a framework for risk tradeoff analysis of chemical substance substitution

— An approach to risk assessment using relative comparison —

Masashi GAMO* **and Jun-ichi TAKESHITA**

[Translation from *Synthesiology*, Vol.9, No.4, p.187–197 (2016)]

A chemical substance is often substituted for another to reduce risks associated with use of the original substance. However, the replacement may be associated with new risks, and this introduces a risk tradeoff problem. Although the concept of risk tradeoff analysis has been discussed in this context, no feasible method has yet been developed. In this study, a novel assessment method was proposed, based on relative risk comparison among substances, through an examination of some possible approaches. Case studies were also conducted to assess the efficacy of the method.

Keywords **:** Chemical substances, risk, tradeoff, relative comparison, assessment methodology

1 Introduction

While the use of chemical substances is necessary for achieving a prosperous and sustainable society, there is concern about related risks against humans and ecosystems. The production, use, and emission of chemical substances determined to pose significant risks are regulated by laws and through self-management. For example, such substances as polychlorinated biphenyls (PCBs), some chloride agrichemicals, brominated flame retardants, and heavy metals such as mercury, cadmium, and lead are strictly regulated, while the emissions of substances like air pollutants such as toluene are declining every year due to voluntary emission control.[1]

Although the risk associated with a particular substance can be reduced by regulating the use and emission of that substance, related measures may be costly; and replacement by different, less hazardous substances is often made, while maintaining the functionality of the product in which the original substance was used. For example, the brominated flame retardant, decabromodiphenyl ether (decaBDE), has been replaced by substances such as bisphenol-A bis(diphenyl phosphate) (BDP);[2] lead solder alloys by 'lead-free solder alloys,' such as tin-silver-copper alloys, which do not contain lead;[3] and chlorinated solvents by carbohydrate or aqueous varieties, for use as industrial cleansers.[4]

The occurrence of new risk when reducing a certain risk is called 'risk trade off.' To assess whether the replacement of a given substance constitutes appropriate risk management, it is insufficient merely to demonstrate reduction in the risks associated with the original substance. It is necessary also to consider the potential risks associated with the replacement substance. It is necessary to determine whether the overall risk of the replacement substance is less than that of the original substance; and in addition, by comparing the risks before and after substitution, to assess whether the given substitution produces a risk reduction effect that exceeds the cost of countermeasures.

Kishimoto[5] argued for the necessity of developing an assessment method for human health risks associated with chemical substance use, which would reflect social requirements, including the new social demand for "comparing risks of different types of chemical substances, and the assessment of cost-effectiveness of emission reduction measures;" and presented a case study involving toluene risk assessment. Here, he proposed a method for quantifying human health risks, using quality of life (QOL) as the risk index, by backcasting from the demand. The method enabled the comparison of the cost-effectiveness of risk reduction measures for toluene with that of risk reduction measures for other chemical substances, infectious diseases, accidents, disasters, etc.

However, Kishimoto's proposal is, in practice, applicable only to substances such as toluene, for which abundant toxicity information is available. In risk tradeoff analysis associated with substance substitution, the substitution often involves replacing a substance for which there is relatively abundant information, with one for which there is insufficient information. For such risk tradeoff analysis to become a

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realistic assessment method, then, it is necessary to develop a technique whereby Kishimoto's risk comparison conception is executable.

In this paper, we discuss the approaches taken in this regard, as well as the development of basic techniques enabling risk tradeoff analysis of chemical substance substitution, and the results of related case studies. We also discuss future prospects for this analytical method.

2 Investigation and discussion of approaches

Here, we consider the merits and demerits of current approaches to risk tradeoff analysis, in terms of the comparative risk before and after substance substitution. Figure 1 shows the relationship between the basic techniques and the complex of analytical methods considered below.

2.1 General risk analysis method

The first approach involves the application of a general assessment method for chemical substance risks. In this method, the nature and level of risk is assessed through comparison of levels of tolerance to those of exposure to a given chemical substance. Tolerance level, the exposure level at which the manifestation of toxicity is not a concern, is often calculated by applying an uncertainty factor (safety factor) to the results of animal tests and human epidemiological studies; whereas, exposure level is determined by multiplying the concentration in environmental media (air, water, food, etc.) by the amount of intake of the media. The concentration in environmental media is obtained by actual measurement or by simulation-based prediction. There is considered to be no risk if the exposure level is less than the tolerance level, and risk if the former is greater the latter.

However, since the comparison of exposure and tolerance levels is conducted separately for each individual substance, if the respective ratios of exposure to tolerance level for two substances are calculated to be 0.1 and 0.5, for example, neither is considered to involve risk, and no relation is established between them. This lack of relation becomes especially problematic when the types of toxicity differ between the two substances. With this approach, then, any difference in the level of risk before and after substance substitution can be considered only as difference in the presence (or absence) of risk.

In addition, calculation of the tolerance level requires the results of animal tests and human epidemiological studies; and since sufficient information on toxicity may not be available to enable accurate tolerance level calculation for a given substance after substitution, it must be concluded that risk tradeoff analysis based on this method is difficult.

2.2 Method using a common risk index

To solve the problem faced by such general risk analysis methods, wherein the relative risks of multiple and varied substances cannot be compared, Kishimoto proposed an approach involving quantitative expression of the magnitude of health risks, based on a common index.[5] As common indices, lifespan reduction due to adverse health effects, or similar reduction but with adjustment for quality of life, are both widely used. For example, Gamo *et al*.[6] used an index called lost life expectancy (lifespan reduction) to evaluate replacement of a termite control agent; and Gamo *et al*.[7] used this same index to rank major environmental pollutants. Kishimoto[5] used an index based on loss of quality-adjusted life-years (QALYs: lifespan adjusted for quality of life) in the detailed risk evaluation of toluene; and Cohen *et al*.[8] used this index to compare the benefit of polysaturated fatty acid, with the risk of methyl mercury, in fish consumption. Disability-adjusted life expectancy years (lifespan with adjustment for disabilities caused by disease) is used as an index in calculations of the worldwide burden of disease by the World Health Organization (WHO);[9] and Havelaar *et al*.[10] used this index to compare the risk of bromate

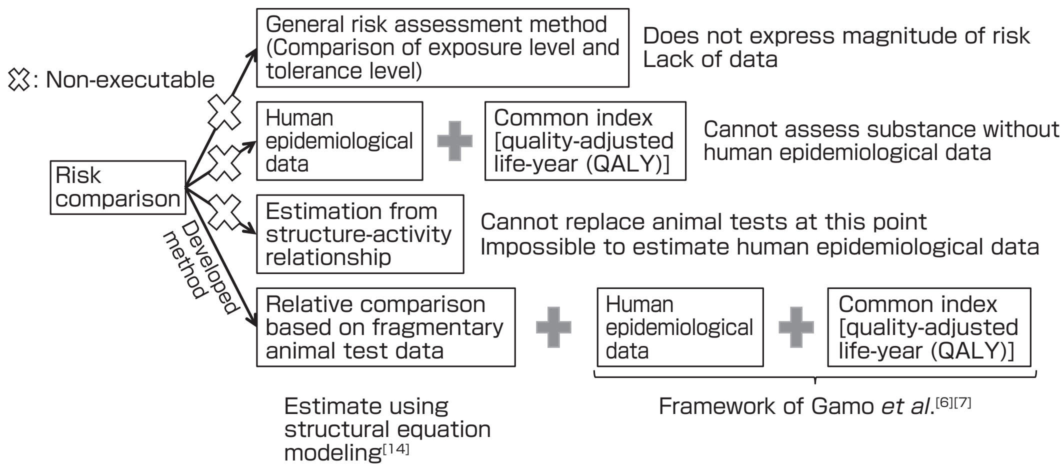


Fig. 1 Scenario for developing the methodology for risk tradeoff analysis, and the basic techniques

byproducts, with risk reduction in infectious disease by the disinfection of drinking water.

The problem with this approach is that its estimations require the results of human epidemiological studies, and many potential replacement substances do not even have sufficient animal test data, let alone human epidemiological study results. In sum, this approach is less executable than the general risk analysis method described in Subchapter 2.1.

2.3 Estimation based on structure-activity relationships and/or cell-based assays

In light of the problems faced by the foregoing methods, one way to counter the lack of information on the potential toxicity of substances after substitution is a risk estimation approach based on structure-activity relationships and/or cell-based assays. The structure-activity relationship is an expression relating the structure of a given chemical substance to its activity (in this case, toxicity). By constructing the expression, based on data for multiple chemical substances, it is possible to estimate the potential toxicity of substances without animal or human study data. Several related studies have been conducted, and recently a prediction system based on categorization for repeated dose toxicity, called the Hazard Evaluation Support System (HESS), was developed in Japan.[11]

In a similar manner, the approach based on cell-based assays involves conducting tests using cells instead of animals, and assessing the presence and degree of toxicity of the target substance based on these tests. The Ames test (detection of mutagenicity using bacteria), which evaluates the mutagenicity of chemical substances, is well known, and several cell-test methods are described in the guidelines of the Organisation for Economic Co-operation and Development (OECD). However, the general consensus is that it is difficult to correlate the effect on cells with the effect on individual organisms.

While use of these methods is expected to increase in the future, due to demands for cost reduction in animal testing and from the perspective of animal rights, they cannot replace animal tests at this point; and it is technically impossible to estimate information equivalent to human epidemiological studies, based on the results of these methods. Moreover, even if technological development progresses to the point where it becomes possible to determine the nature and level of risk for each substance, this alone would not enable multiple-substance risk comparison, as discussed above with regard to the general risk evaluation method (Subchapter 2.1).

2.4 Method of multiple-substance relative comparison

To enable comparative risk tradeoff analysis of substance substitution, we have proposed an approach involving the relative comparison of substance risks, based on a quantitative risk evaluation method (Subchapter 2.2), to enable relevant estimation when human epidemiological data is unavailable. In this approach, the substance for which the risk can be evaluated using a common scale based on human epidemiological data is established as a reference substance, and hazard assessment of the target substance is conducted by relative comparison with the reference substance. This avoids the difficulty of estimating the relevant human epidemiological data based on animal test results (or on cell-based assays or structure-activity relationships).

Several reports have been published on the approach of assessing substance risk without sufficient test data, based on such comparison. For example, Maier[12] proposed the "parallelogram approach," where workplace tolerance concentrations are established by comparing the activities of pharmaceutical intermediates without hazard data, with those of substances with sufficient animal test data, based on cell-based assays. This is similar to our proposal in the sense that the results of cell-based assays are not directly used for estimating individual effects, but instead as a tool for relative comparison. Nakanishi *et al*.[13] proposed a "two-axis approach" for conducting risk evaluation of carbon nanotubes (CNT). To evaluate the risk to workers of inhalation exposure to various CNTs, the inhalation exposure test, which is standard but costly, is conducted only for representative CNTs, while the simpler method of an intertracheal administration test (where a CNT suspension is applied to an animal's trachea) is conducted for other CNTs, in order to establish a wide range of comparison.

In the case of the method discussed here, the established method[6][7] was chosen as a basis for quantifying the risk of reference substances. However, as there is no existing research on a method for relative risk comparison of reference and target substances, it was necessary to develop an original method for conducting such comparison based on limited information.[14] The details of this method will be presented in the next chapter.

3 Framework of assessment and basic techniques

3.1 Overall structure

It is assumed that, for the target substance, there is only fragmentary animal test data, and no human epidemiological data, to enable the sort of quantitative risk assessment as is described in Subchapter 2.2. Here, fragmentary animal test data denotes cases where, for example, there exist data or papers characterizing the specific effects of the substance on the liver, but none describing all the effects on all organs; and human epidemiological data refers specifically to the dose-response relationship (relationship between the exposure level and occurrence rate of the effect) in humans.

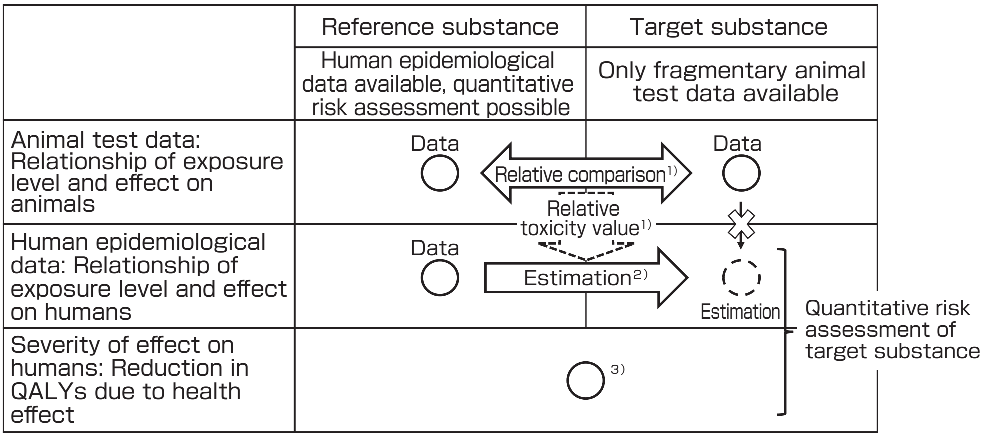
In reality, it is impossible to directly estimate such target substance human epidemiological data based solely on fragmentary animal test data, given the species difference between humans and animals. Therefore, we devised a framework for conducting such an estimation with the following procedure (Fig. 2 shows a conceptual diagram of the procedure).

1) The substance with human epidemiological data, and for which quantitative risk assessment is possible, is established as the reference substance.
2) A relative comparison is made between the reference substance and the target substance, at the level of animal test data, to calculate the relative toxicity value, which is the ratio of the exposure level of the target substance, to that of the reference substance at which both substances present the same level of toxicity.
3) The dose-response relationship of the target substance is estimated by multiplying the relative toxicity value calculated in (2) with the known dose-response relationship of the reference substance (Fig. 3). In this case, it is desirable to establish a confidence interval for the estimated dose-response relationship equation.
4) The quantitative risk assessment is conducted by combining the dose-response relationship of the target substance and the value of severity of the expected health effect as expressed in terms of a common index.

Typically, chemical substances affect multiple organs, but the level of exposure required to trigger an adverse effect varies with the given organ. Therefore, to reflect the diversity in substance toxicity, it is desirable to conduct this four-step estimation for each organ. Thus, we decided to treat the liver and kidney independently, as they were major organs in which adverse effects occurred. The reference substances were established and relative toxicity values were calculated for each organ.

3.2 Reference substances and dose-response relationships

In establishing the reference substances for liver and kidney



1) Relative comparison between the reference substance and the target substance, calculation of relative toxicity value from Takeshita *et al.*[14] (Subchapter 3.2)
2) The conceptual diagram for estimating the human epidemiological data for the target substance, based on the human epidemiological data and relative toxicity values of the reference substance, is shown in Fig. 3.
3) These values are general values for types of effects, and it was assumed that they are not dependent on types of exposed substance.

Fig. 2 Framework of the risk tradeoff analysis based on relative assessment of substances

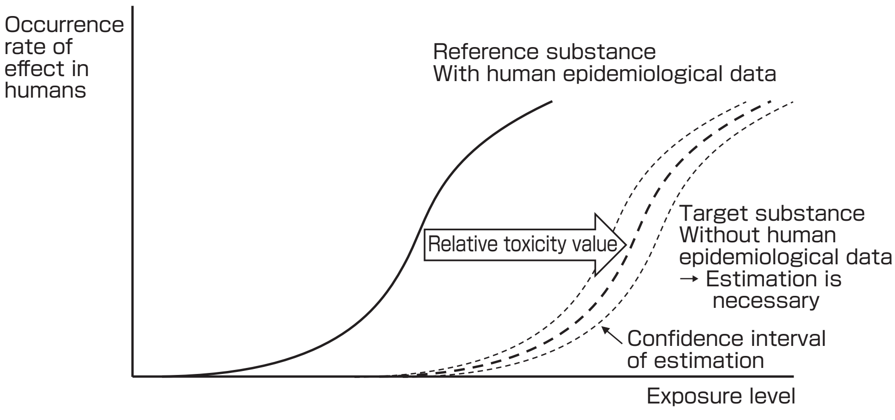


Fig. 3 Estimation of the dose-response relationship of the target substance, based on the dose-response relationship of the reference substance and the relative toxicity value

effects, the CHE Toxicant and Disease Database[15] of the Collaborative on Health and the Environment (CHE), an international environmental group, was used to search for chemical substances that were known to cause adverse effects on organs. This database, which is based on three famous textbooks on toxicology, enables searches for chemical substances that may be the cause of disease.

We selected substances for which the evidence was “strong” for liver and kidney effects, and then narrowed the search to substances with published human epidemiological data, by referring to existing documents on hazard assessment. As a result, we established as reference substances vinyl chloride monomer (chloroethene) for liver, and cadmium for kidney effects. The human dose-response relationship information for each substance is as follows.

3.2.1 Liver effect

Ho *et al*.[16] studied workers who were exposed to vinyl chloride monomer in the air, and reported that of 271 subjects exposed to 1–20 ppm (equivalent to 2.5–50 mg/m^3), 12 showed liver dysfunction (4 had liver enlargement, 4 had liver and spleen enlargement, and 4 had spleen enlargement). As this was occurrence rate data for a specific concentration range, we decided to apply the distribution of sensitivities to noncarcinogenic effects, as proposed by Huijbregts *et al*.;[17] that is, a geometric standard deviation of 1.82 when the lognormal distribution is assumed for individual differences. As a result, for inhalation exposure to vinyl chloride monomer, the dose-response relationship of liver effect was set as the lognormal distribution (with a geometric mean value of 31 mg/m^3, and a geometric standard deviation of 1.82). In using this value for the risk assessment of oral exposure, the concentration in air was converted to a daily intake amount (unit: mg/kg/day) by using assumed values for respiratory volume and weight.

3.2.2 Kidney effect

The effects of cadmium on humans have been studied in detail, and renal tubular disorder is known as a highly sensitive adverse effect. In this case, then, the established dose-response relationship value was used. Renal tubular disorder was defined as the situation where the urine β_2-microglobulin concentration exceeded 1000 μg/g creatinine. The sensitivity of this tubular disorder is age dependent, and Gamo *et al*.[18] and Nakanishi *et al*.[19] summarize the dose-response relationship parameters for each age.

3.3 Derivation method for the relative toxicity value[14][20]

The following two factors were considered requisite for a relative toxicity derivation method to be developed.

- It is possible to complement missing data: On the assumption that the available animal test data is fragmentary, it may be the case that either or both the data for liver and kidney are lacking. In such a case, it is difficult to use the data in actual risk tradeoff analysis unless the relative toxicity values can be estimated for both organs. The method in which the significance of effects in multiple organs are mutually estimated is called the quantitative activity-activity relationship (QAAR).
- It is possible to estimate the confidence interval: Estimation is inherently uncertain, yet the estimation of uncertainty is essential in considering the results of risk tradeoff analysis. For example, in cases where it seems that the risk has decreased with the replacement of a given substance, the estimation of a confidence interval is necessary to determine whether the decrease is dubious or not.

With these factors and the methodological aim in mind, we decided to apply structural equation modeling (SEM), which is a statistical analysis method encompassing linear regression analysis and factor analysis. In SEM, the statistical model parameters used in establishing relationships between the variables are determined such that the variance and covariance calculated from the model will best correspond to the variance and covariance calculated from the actual data. Figure 4 shows an abstract diagram of this procedure.

To construct the training data set, existing test data were obtained from literature on roughly 165 substances listed in the Japanese Pollutant Release and Transfer Register as Class I Designated Chemical Substances. There are 45 toxicity endpoint items in total, for the combination of target organs (liver, kidney, blood, urine, body weight, death, spleen, digestive tract, respiratory organ, brain, etc.), test animal species (rat, mouse), and administration methods (oral exposure, inhalation exposure).

Using the constructed model, the no-observed-effect level (NOEL) is estimated for each item and substance, and based on this result, the relative toxicity value of the two substances is estimated. Let Substance A be the reference, Substance B be the target, and $\hat{a}$, $\widehat{\sigma_a}$, $\hat{b}$, $\widehat{\sigma_b}$ be, respectively, the logarithmic NOEL value for Substance A, its standard deviation, the logarithmic NOEL value for Substance B, and its standard deviation. Then, the relative toxicity value of Substance B versus Substance A (the reference) and its 95 % prediction interval are calculated by the following equations.

Relative toxicity value: $\exp(\hat{b}-\hat{a})$
95 % prediction interval:
$$[\exp(\hat{b}-\hat{a}-2\sqrt{\widehat{\sigma_a}^2+\widehat{\sigma_b}^2}),\ \exp(\hat{b}-\hat{a}+2\sqrt{\widehat{\sigma_a}^2+\widehat{\sigma_b}^2})]$$

The estimation accuracy of the respective NOELs for each substance was quantified according to the OECD principles of verification, by applying the leave-one-out cross-validation method, which dictates that each observed value is removed and then estimated by the remaining observed values. This is repeated for all values, and then the estimated values are compared with the true values. In this case, the correlation coefficient between the observed and estimated values was 0.89; and in terms of estimation accuracy, 93 % of the observed values were included in the 95 % prediction interval, and 97 % of the ratios of estimated and observed values were less than 10.

3.4 Common index of the effects

As noted in Subchapter 2.2, among the common indices of human health effect are lost life expectancy, quality-adjusted life-years (QALYs), and disability-adjusted life-years. Here, we used the QALY index, which incorporates both lifespan and QOL reduction. In the case of QOL reduction, various values are typically reported, according to the disease and its state, even if the same organ is affected; but here we set a general value, without assuming any specific disease state. As the liver and kidney effect information obtainable for the reference substances differed, the estimation method differed for the two organs.

3.4.1 Liver effect

The liver dysfunctions reported by Ho *et al.*,[16] as discussed in Section 3.2.1, were based on the observation of workers, and were considered to reflect a relatively light, chronic effect due to long-term exposure to chemical substances. In the risk ranking of environmental pollutants, Gamo *et al.*[7] conducted an assessment by specifying as one year the reduction in lifespan due to health conditions accompanied by some expression of subjective symptoms, and our present evaluation adopted this assessment. For QOL, the data compiled and organized by Tengs *et al.*[21] was reviewed, and the QOL value of 0.01 was used, in the QALY calculation, for the lightest liver disorder among the liver diseases. This value means that, for example, when an individual lives (for about 80 years) in this state of health, the disorder will be considered to have the same degree of severity as a lifespan reduction of 80 yr × 0.01 = 0.8 yr.

3.4.2 Kidney effect

Renal tubular disorder by cadmium exposure is thought to occur in those over 50 years old, and it has been reported that the mortality of people in this health state is, respectively, 1.57 times (males) and 1.81 times (females) the mortality rate of healthy individuals.[22] Based on this information, lifespan reduction was calculated based on the life table (a table of mortality by age, which enables calculation of average lifespan and other values). For the QOL reduction due to kidney disease, the data compiled and organized by Tengs *et al.*[21] was similarly reviewed, and the QOL value of 0.01 was used for the lightest kidney disorder.

4 Case study

The above method was used for the three risk tradeoff analyses[2]–[4] conducted by the Research Institute of Science

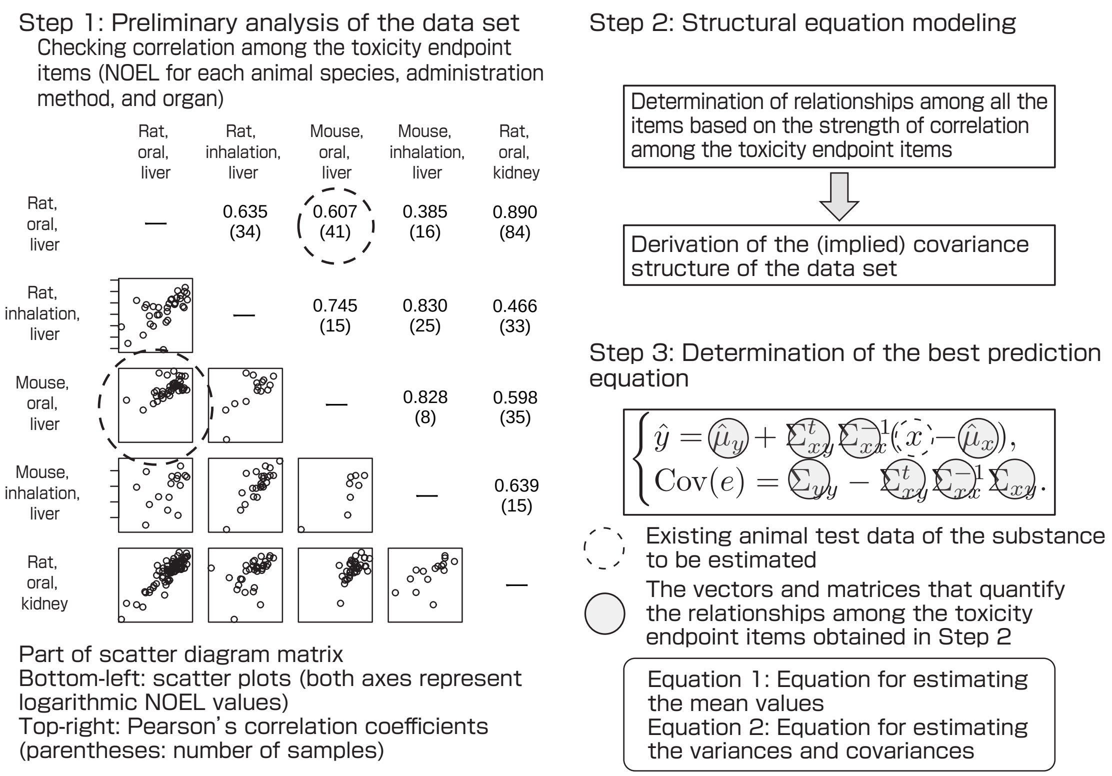


Fig. 4 Procedure for developing the QAAR method using structural equation modeling

The dashed circle in the scatter diagram matrix in Step 1 shows the Pearson's correlation coefficient and scatter diagram, when the NOEL of the liver effect due to oral exposure is compared for rats and mice (horizontal axis: rat value; vertical axis: mouse value).

Table 1. Change in estimated exposure level (decaBDE equivalent) with the substitution of flame retardant: Liver effect

		Estimated exposure level (mg/kg/day: decaBDE equivalent)			
		decaBDE	BDP	TPP	Total
Scenario with substitution	When mean value of relative toxicity value is used	2.1×10^{-4}	8.8×10^{-5}	4.0×10^{-7}	2.9×10^{-4}
	When 95 % upper limit of relative toxicity value is used	2.1×10^{-4}	4.1×10^{-4}	7.9×10^{-6}	6.3×10^{-4}
Scenario without substitution		3.3×10^{-4}	–	–	3.3×10^{-4}

Table 2. Change in estimated exposure level (decaBDE equivalent) with the substitution of flame retardant: Kidney effect

		Estimated exposure level (mg/kg/day: decaBDE equivalent)			
		decaBDE	BDP	TPP	Total
Scenario with substitution	When mean value of relative toxicity value is used	2.1×10^{-4}	6.9×10^{-5}	5.1×10^{-8}	2.7×10^{-4}
	When 95 % upper limit of relative toxicity value is used	2.1×10^{-4}	4.2×10^{-4}	3.1×10^{-7}	6.2×10^{-4}
Scenario without substitution		3.3×10^{-4}	–	–	3.3×10^{-4}

Table 3. Risk associated with the flame retardant in each substitution scenario[2] (day : quality-adjusted life-year : value per person for lifetime exposure)

	With substitution (current situation)		Without substitution (imaginary situation)
	When mean value of relative toxicity value is used	When 95 % upper limit of relative toxicity value is used	
Liver effect	<< 0.001 (2.8×10^{-57})	<< 0.001 (2.0×10^{-53})	<< 0.001 (9.5×10^{-57})
Kidney effect	<< 0.001 (1.4×10^{-140})	<< 0.001 (1.0×10^{-122})	<< 0.001 (8.8×10^{-137})
Total	<< 0.001 (2.8×10^{-57})	<< 0.001 (2.0×10^{-53})	<< 0.001 (9.5×10^{-57})

for Safety and Sustainability at AIST. Here, we present a summary of the replacement of a flame retardant used as a plastic additive.[2] A study was done on a scenario where decabromodiphenyl ether (decaBDE) was partially replaced by bisphenol-A bis(diphenyl phosphate) (BDP) [which includes triphenyl phosphate (TPP) as impurity], and an imaginary scenario where a replacement for decaDBE was not sought. For these scenarios, we conducted a material flow analysis consisting of the assessment of the demand volume, community-acquired stock volume, and waste volume of each substance. Then the respective exposure levels of each substance, through indoor air, environment, and food, was estimated. The risk tradeoff analysis was conducted based on the estimated exposure levels for each substance in each scenario.

Before conducting the quantitative risk analysis based on common indices, we first investigated the qualitative difference in risk due to substitution. DecaBDE, the original substance, was set as the reference substance, and the relative toxicity values for BDP and TPP were calculated; after which, the estimated exposure levels in each scenario were calculated and totaled. The overall exposure level was expressed as an equivalent amount of decaBDE (mg/kg/day). Table 1 shows the liver effects, and Table 2 the kidney effects. In the scenario with substitution, the estimated exposure level of decaBDE equivalent was less than in the scenario without substitution. Even with the addition of BDP and TPP, it could be determined, using the estimated mean value of relative toxicity, that the exposure level of decaBDE equivalent decreased slightly in the scenario with substitution. However, when the estimated 95 % upper limit of the relative toxicity value was used, the exposure level of decaBDE equivalent increased with substance substitution. This means that, when the uncertainty involved in the estimation of the replacement substance's toxicity was considered, it could not be determined whether the substance substitution would necessarily contribute to risk reduction. Here, the ratios of the 95 % upper limit and the mean of the relative toxicity value for the liver effect, for BDP and TPP, were 4.7 and 20, respectively; and for the kidney effect, 6.0 and 6.0, respectively. The difference in the ratio values for the different substances and organs reflects the fact that the availability and/or reported values of NOEL varied.

Table 3 shows the results of the quantitative risk comparison by common indices, based on the method described in Chapter 3.[2] The magnitude of risk is expressed as the QALYs (days) per person, through lifetime exposure. In both scenarios, extremely small values, both for liver and for kidney effect, were found. A lifetime probability of cancer occurrence of 10^{-5}, often used as the upper limit of the tolerable risk level, is equivalent to a lifespan reduction of about 0.04 d.[23] Therefore, when the loss in QALYs is less than 0.001 d, it can be concluded that the effectiveness of the substance substitution, in terms of risk reduction, cannot be determined.

5 Discussion

The substance substitution risk tradeoff analyzed in the case studies of the flame retardant[2] (Chapter 4), lead solder alloy,[3] and industrial cleanser,[4] arose from concern for the risk presented by the original substances. To determine whether such substitutions are appropriate from the perspective of risk reduction, quantitative risk analysis based on scientific evidence is essential, and the aforementioned studies performed at the Research Institute of Science for Safety and Sustainability were the first instances of such analysis. This was primarily made possible by the development of the assessment method described in this paper. Kishimoto[5] developed the concept, and we engaged in developmental research on basic techniques to realize it.

Our risk tradeoff analysis involved chemical substance substitution aimed at preservation of the original substance's functionality; for example, preserving the flame-retardant property in plastic. However, related measures may involve simply reducing the amount used of a certain substance, to reduce the associated risks. In such cases, a tradeoff occurs which may involve risks other than chemical substance toxicity alone. To take two examples, the reduction of product functionality may increase the risk of fires or accidents, and reducing energy efficiency may increase the risk of global warming. While we have constructed a provisional framework for comparing the risks associated with different chemical substances, there remains the question of how to compare risk types other than toxicity, with the attendant questions of how to express such different risk types in terms of common indices, and how society should understand tradeoff involving risks with greatly varying properties. Such questions must be addressed, for example, in the risk analysis of substances (such as nanomaterials) used in future technologies, for which, as practical application is lacking, there is, as yet, no risk tradeoff concern, though strict regulation may be applied. However, if such regulation should limit the possibilities for future technology, there is a chance of forsaking future risk reduction in some fields, and this too must be considered.

The method of chemical substance risk tradeoff analysis presented in this paper must also be improved in the following respects. The toxicity data that formed the basis for the QAAR model constructed for the relative comparison of toxicity consists of animal test results published in general toxicological journals, and the reliability of the data has not been carefully investigated. Currently, government agencies tend to utilize highly reliable test data, collected within the framework of the Law Concerning the Examination and Regulation of Manufacture, etc. of Chemical Substances, for the purpose of creating a structure-activity relationship model for toxicity. The rebuilding of models, based on such data, will increase the reliability of the assessment results. Also, this paper's QAAR model is based solely on the correlation among the toxicity endpoint items observed in animals; however, to enable the assessment of substances for which no animal test results are available, NOEL estimation values based on structure-activity relationships, as well as the results of cell-based assays, and chemical substance structure descriptors, should be used as variables in structural equation modeling. In addition, to increase the model's reliability, both variable selection and establishment of cause-effect relationships can be performed based on the information on the mode of action of chemical substances. Finally, in this study, the focus was on the effects on major organs (here, the liver and kidney). However, chemical substances may also have neurotoxicity and/or sensitizing effects, and future research topics must include the selection of reference substances, the understanding of dose-response relationships, and the estimation of QALYs, for these other effects.

Improvement is also needed in the treatment of uncertainty. In this paper, we drew attention to the importance of the estimation of uncertainty in the tradeoff analysis, and quantified the uncertainty in the estimation of relative toxicity values, based on the correlation among toxicity endpoint items. However, other relevant uncertainties were not treated here explicitly. Notable among these are the uncertainties involved in the selection of reference substances (even if the focus is on 'liver effect,' it may be necessary to establish different reference substances for different substances), the estimation of dose-response relationships for reference substances (uncertainty in the reliability of the human epidemiological data on the given reference substance, and/or uncertainty accompanying the derivation method for obtaining the given dose-response relationship), and the determination of QALYs for a given effect (even if the focus is on 'liver effect,' the degree of severity may differ among substances). Also, in terms of exposure assessment, there are uncertainties in the establishment of the substitution scenarios and estimation of exposure levels. We must, to some extent, accept the fact that uncertainty is an inherent feature of assessment. For example, though in the research and development of assessment methods, the main aim is to reduce the uncertainty in factors with a large degree of uncertainty, and much R&D work is focused on this aim, there are cases where there is a limit on reducing uncertainty, and/or there is little or no effect on decision-making despite the presence of large uncertainty (for example, in the context of the present study, when the superiority of Substance A over Substance B is very clear, even with uncertainty considered). Nonetheless, to develop a risk tradeoff analysis method that contributes to effective decision making in future, the quantitative estimation of uncertainty, and the formation of consensus regarding the magnitude of uncertainty, will become increasingly important.

In presenting our novel method for risk tradeoff analysis, and the related case study, our hope is that consideration of the risk tradeoffs involved chemical substance substitution will ultimately extend to society at large; and in this way, it may be possible to avoid unhelpful regulations or the substitution of substances based on poor scientific evidence. As noted earlier, substance substitution incurs costs, and the risk may even increase in some cases. Effective risk tradeoff analysis is necessary both for the companies that manufacture chemical substances and for the general public that uses them. While there is no move for incorporating such assessment in specific regulations at this point, the OECD, for example, has established an ad hoc group focused on the substitution of hazardous chemical substances, and is discussing the development of methods to support safe assessment of substance substitution.[24] We, for our part, are making timely presentations of our assessment method and case studies to such bodies.

It is expected that the relative comparison-based approach to risk assessment, as described here, will be applied to risk assessments in diverse fields, not only to chemical substances. Particularly in the case of new technologies that may replace conventional ones, rather than simply estimating the risks that may be introduced by such new technologies, it will be more realistic and convincing to society at large to perform relative comparisons of the two, based on a clear understanding of the current risks posed by the conventional technologies. Future topics to be studied, then, include the opportunities and requirements for applying the relative comparison approach, the identification and organization of uncertainties inherent in such assessment, and the relationship between such uncertainties and acceptability of the assessment by society.

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Discussions with Reviewers

1 Overall

Comment (Akira Ono, AIST)

This paper proposes a novel, practical risk assessment methodology, based on the technique of introducing an index that can be broadly employed to quantitatively compare the magnitude of risks with differing factors. It should be noted that this method was applied to existing cases to verify its effectiveness.

Some different scenarios for developing the practical risk assessment methodology were compared. The processes of scenario selection are described, and this paper is appropriate as a research paper for *Synthesiology*.

2 Uncertainty in the risk estimation

Question (Akira Ono)

I value highly the fact that you considered and executed the method for estimating the uncertainty of risk (95 % confidence interval: 2σ equivalent). Uncertainty is an important concept, and I think it will be at the core of future research development. Based on this way of thinking, I pose the following question, as well as the subsequent questions and comments.

I would like to understand the specific magnitude of uncertainty. In Tables 1, 2, and 3, how many times greater is the uncertainty (2σ) than the mean value?

You estimate the relative toxicity value of the target substance without human epidemiological data using a statistical method, but what are the factors of uncertainty in this case? Please give examples of factors that have large effects (for example, the quality and amount of animal test data is insufficient, the correlation among toxicity endpoint items is low, or the similarity between the selected reference substance and the target substance is low).

I think one of the key challenges in this research is how to decrease the uncertainty involved in estimating the relative toxicity value of the target substance. To do so, I think it is necessary to clarify the factors that enhance the uncertainty, and make efforts to reduce them, beginning with the largest contributor. What do you think?

Answer (Masashi Gamo)

We did not show the relative toxicity value itself; but the ratios between its mean and 95 % upper limit values for the liver effect, for BDP and TPP, were 4.7 and 20, respectively; and for the kidney effect, 6.0 and 6.0, respectively. The differences in the ratio values for different substances and organs reflect the fact that there are differences in the availability and/or reported values of NOEL.

In this paper, we highlighted the importance of the estimation of uncertainty in the tradeoff analysis, and quantified the uncertainty involved in the estimation of relative toxicity values based on the correlation among toxicity endpoint items. On the other hand, as you indicated, there are several uncertainties that are not treated explicitly. Notable among these are the uncertainties involved in the selection of reference substances (even if the focus is on 'liver effect,' it may be necessary to establish different reference substances for different substances), the estimation of dose-response relationships for reference substances (uncertainty in the reliability of the human epidemiological data on the given reference substance, and/or uncertainty accompanying the derivation method for obtaining the given dose-response relationship), and the determination of QALYs for a given effect (even if the focus is on 'liver effect,' the degree of severity may differ among substances). Also, in terms of exposure assessment, there are uncertainties in the establishment of the substitution scenarios and estimation of exposure levels. We must, to some extent, accept the fact that uncertainty is an inherent feature of assessment. For example, though in the research and development of assessment methods, the main aim is to reduce the uncertainty in factors with a large degree of uncertainty, and much R&D work is focused on this aim, there are cases where there is a limit on reducing uncertainty, and/or there is little or no effect on decision-making despite the presence of large uncertainty (for example, in the context of the present study, when the superiority of Substance A over Substance B is very clear, even with uncertainty considered). Nonetheless, to develop a risk tradeoff analysis method that contributes to effective decision making in future, the quantitative estimation of uncertainty, and the formation of consensus regarding the magnitude of uncertainty, will become increasingly important.

I have added these comments to the text.

3 Verification of the adequacy of the estimation method for uncertainty

Question (Akira Ono)

You estimate the uncertainty of risk estimate values using statistical methods. Is there any way to verify this method's adequacy?

For example, let's say you select several reference substances for a certain target substance, and see how much variation there is in the risk estimate values of the target substance with respect to each reference substance; can you use this as an index of the adequacy of the method?

Or, for example, say you select two substances that have sufficient human epidemiological data, position one of these as the reference substance and the other as the target substance, and then estimate the target substance risk using the method described in this paper. Here, you do not use the human epidemiological data for the target substance, but only the animal test data. I think you could do the same thing by interchanging the reference substance and the target substance. Might not the difference between the highly reliable estimate value based on the human epidemiological data, and the estimate value based only on animal test data, function as an index of the adequacy of the method?

Answer (Masashi Gamo)

Of the methods presented in the paper, we reported the results of the leave-one-out cross-validation method concerning the adequacy of the estimation of uncertainty involved in the estimation of relative toxicity value, as shown in Subchapter 3.3. We found that 93 % of the observed values were included in the 95 % prediction interval, and we believe the estimation of uncertainty was adequate. This method is the most widely accepted, by various bodies (including the OECD), as verification of statistical estimation.

On the other hand, for the method of estimating the uncertainty involved in the establishment of reference substances, I think it is effective, as you suggested, to specify several such substances and compare the estimate results based on these, or to mutually estimate and verify the NOEL of reference substances; although there are not many substances with human epidemiological data, which means that this is one of the topics to be investigated in the future.

4 Use of data from the scenario that was not selected

Comment (Akira Ono)

In Fig. 1, you compare four scenarios and select the fourth scenario for this research, regarding the three non-selected scenarios as non-executable. However, isn't it true that the exposure/tolerance concentration data, structure-activity relationships, and cell-based assay data in the scenarios that you deemed non-executable contain a certain amount of useful information on toxicity, although it is impossible to individually execute the related risk tradeoff analysis? And if you could incorporate these data into this research in some form, could you not thereby reduce even more the uncertainty involved in the risk assessment?

As a future direction in Paragraph 3 of Chapter 5 (Discussion), you note that the point in increasing the reliability of estimation results is to incorporate and integrate such data into the methodology of this study. Please explain, even if it is just your current thoughts, by what mechanism you can reduce the uncertainty by integrating this data.

Answer (Masashi Gamo)

As you indicated, we think that the three non-selected scenarios may contain data and methods that can be utilized effectively in the risk tradeoff analysis.

For substances that have absolutely no animal test data, it is important, from the perspective of animal rights, to use the structure-activity relationships and cell-based assay results. One possible approach is to use the NOEL estimation values based on structure-activity relationships, cell-based assay results, and the chemical substance structure descriptors, as observation variables in the structural equation modeling. On the other hand, in the method presented in this paper, the characterization of the correlation among the variables in the structural equation modeling depended only on the correlation among toxicity endpoint items. However, we can also select variables, and establish the cause-effect relationships among them, based on the information on the mode of action of chemical substances, which is also discussed in the treatment of the general risk assessment method.

Paragraph 3 of Chapter5 (Discussion) was modified to reflect these concerns.

5 Reason for replacing the flame retardant

Question (Akira Ono)

Table 3 shows the results of risk assessment, taking the example of a conventional flame retardant. I understand that the right column of the table (with no substitution, the imaginary situation) shows the reduction in QALYs if the conventional flame retardant is used, compared to when we stop using that flame retardant.

The conventional flame retardant and replacement substance both have very small risk estimation values, in fact dramatically small compared to the reduction in QALYs of 0.04 days, which is the upper limit of the tolerable risk level. What is the reason for the extremely low risk values obtained in the assessment? Does this show that the risk has been overrated, and is in fact very small? Or is it due to your introduction of the common index of QALYs? Please explain.

And another question along the same lines. According to this assessment result, the original-substance reduction in QALYs is negligibly small, and you conclude that the substance substitution was scientifically meaningless. I think this conclusion is correct; but then, why was this flame retardant substituted for the original? What was the reason? The reason might not have been scientifically sound, but it must have been convincing enough at the time. Please provide the authors' view, to the extent of your knowledge, on what the reason was.

Answer (Masashi Gamo)

In Table 3, the magnitude relationships between risk estimation values (QALYs) in the scenarios with and without substitution are reversed, depending on whether one uses the mean or the 95 % upper limit of the relative toxicity value. This is true also in Tables 1 and 2, but it indicates that one cannot determine whether the substitution of a given substance necessarily contributes to risk reduction, considering the uncertainty involved in the assessment.

In this specific case study, it was calculated that the risk estimation value was extremely small. I think we would have reached the conclusion that there was no risk, using the general risk assessment method (comparison of exposure and tolerance levels) as well. However, as explained in Subchapter 2.1, the general risk assessment method could only provide the conclusion that 'there is no risk,' whereas in this study, the magnitude of risk could be specifically presented in terms of QALYs. By doing so, it was clarified that the risk was not at a level that necessitated discussion of risk reduction measures.

The reason for the flame retardant substitution was probably that people were interested only in the toxicity of the conventional flame retardant, and there was widespread desire to avoid this substance. I do not think even a general risk assessment was done at the time, and comparison of the hazards or risks of the original and replacement substances was not done in detail, unlike the careful analysis performed in this paper.

A first empirical analysis of JIS lifespan

— Implications for the review system of de jure standards —

Suguru Tamura[1,2,3]

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In this study, we normatively discuss the road map scenario to improve the management system of standards and then to improve the national innovation system. In AIST, there are many research projects of standards' generation, but the research about the management of established standards is rare. For this purpose, factors related to the lifespan of de jure standards are examined. We especially focus on the effect of technological categories of standards on lifespans. Under the system used by the Japanese Industrial Standards Committee, the review period for standards is five years, and it has not been changed for several decades. The system of ISO has been in the same situation for several decades as that of Japan Industrial Standards (JIS). By using the record of about 4500 JIS standards, the de jure standards of some industrial technology areas are shown to have a tendency toward longer lifespans. Depending on the obtained study results, we proposed a road map scenario to improve the national innovation system through the management of standards, which incurs less administrative costs and makes timely market creation.

Keywords **:** Lifespan, de jure standard, review interval, technological category

1 Introduction

In this study, we normatively discuss innovation to improve the management system of standards in a science and innovation policy perspective. For this purpose, we focus on the review system of the standards. Through this study, we have found that the review intervals of standards are fixed regardless of technological fields and the system has been the same for decades from the previous century. This system is the same for international standardization organization such as International Organization for Standardization (ISO) and de jure standards in Japan.

The research on standards in terms of innovation management is still in its introductory phase.[1] This study focuses on de jure standards, as set by governmental agencies, rather than on de facto standards, which are the results of market competition. De jure standards are fundamental for innovation. For example, MPEG (Motion Picture Experts Group), the digital format for exchanging moving pictures, is standardized as a de jure standard and is widely used to exchange digital movies.[2] A topic of research in the formation of the standards is the parallel development of standards and R&D activities. However, in the case of R&D of a public research institution in Germany (BAM: Federal Institute for Materials Research and Testing), previous research pointed out that standardization does not move parallel to the R&D results of published papers in the field of basic research.[3] This implies that the formation of standards does not necessarily contribute directly to innovation. In the case of the US research institute for standards, the National Institute of Standards and Technology (NIST), the evaluation of individual R&D projects is still in the preliminary stage.[4] This case also shows that the formation of standards (pre-formation) itself is only a part of the national innovation system and we need to explore how to manage standards (post-formation). In this study, we explore the management system of formed standards, with the aim to achieve an efficient national innovation system. We suggest a road map scenario, which includes both pre-standardization and post-standardization steps to improve the national innovation system through an efficient management system of standards. We base our analysis on the survey of a number of research articles related to standardization released by the National Institute of Advanced Industrial Science and Technology (AIST).

In this study, we focus on a public national research institution, AIST in Japan, as in the above-mentioned research in Germany.[3] In AIST, there are many research projects focusing on standards' generation, but there seem to be no research on how to manage already established standards, in their innovation scenarios. There has been little research and almost no discussion about the role of standards after their formation for the transformation of laboratory technologies into market practices. More discussion is needed on this issue in terms of integration of related elements including both social and technological factors.[4] We especially need further discussion to clarify

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the links among R&D results, standards' formation (pre-standards' formation) and standards' management (post-standards' formation) in different innovation scenarios, both nationally and internationally. The OECD Frascati manual, the international guideline for innovation measurement since 1963, does not discuss how to manage already formed standards.[5] From the economic perspective, researchers have been focusing more on why and how standards are formed, rather than how we normatively manage already formed standards.[6]–[9]

How do scholars and practitioners approach the topic of standards' management after they are formed? One fundamental aspect is the lifespan of standards. The importance of this aspect is easily inferred from the case of patents and copyrights. The legal lifespan of patents and copyrights is a key factor to determine the value of patents and copyrights, after they are formed. The legal lifespan of patents is 20 years in Japan, but in some technology areas, like biotechnology, it can be extended to protect the value of patents. The lifespan of patents is a matter of value management for innovation. We ask whether we already have sufficient knowledge about the lifespan of standards to manage existing standards. It seems clear that we do not. We have surveyed the existing knowledge and several factors related to the lifespan of de jure standards, which are examined from the standards' management prospective. Among all factors, we focus on the effect of the technological category of standards on their lifespans. Our results suggest a management system of standards leading to less administrative costs and achieving timely market creation. This management system is normatively presented in a following road map scenario for innovation.

2 Background

In AIST, there are many research projects reflecting a wide range of technology sectors. In addition, several research projects involve standardization. AIST is organized into 5 departments and 2 centers, which range from life science and information technologies. Its budget is about 1269 million USD for 2014. AIST is conducting research with a focus on industrialization. It has about 2200 researchers and it is one of the largest R&D institutions in Japan. The institution is also in charge of national measurement standards in Japan, like the National Institute of Standards and Technology (NIST) in the US. It promotes international standardization as part of its open innovation strategy.[10] In terms of policies, AIST is an affiliated agency of the Ministry of Economy, Trade and Industry (METI). METI is in charge of the management and formation of de jure standards in Japan, known as Japanese Industrial Standards (JIS), and of the country's innovation policy. Because of this twofold organizational structure, there is much research related to standardization conducted at AIST, in various technology fields. In addition, the AIST staff plays a key role in both the committees of the International Organization for Standardization (ISO) and the International Electrotechnical Commission (IEC). While 48 researchers from AIST have served as chairs, secretaries and conveners, 258 researchers have participated in meetings of those organizations as experts.[10]

In terms of standardization, the research conducted at AIST includes the following topics:

1. Environmental analysis methods of hazardous chemicals;[11]
2. Production and utilization of thermophysical property data;[12]
3. High pressure gaseous hydrogen;[13]
4. Four dimensional radiotherapy system;[14]
5. Secure password authentication schemes;[15]
6. Methodology for designing cryptographic systems;[16]
7. Utilization of thermophysical property data;[17]
8. SOFC systems;[18]
9. Font size for elderly people;[19]
10. SOFC cell/stack power generation performance tests;[20]
11. Utilization of observational data;[21]
12. Analysis method for oxygen impurity in magnesium and its alloys;[22]
13. Automotive navigation and route guidance system;[23]
14. Thermoelectric hydrogen gas sensor;[24]
15. Safe usage of moving images;[25]
16. Evaluation device of cosmetics for UV protection;[26]
17. Cryptographic modules;[27]
18. Three-dimensional shape for supporting industry;[28]
19. Geological map;[29]
20. Accessible design for senior citizens.[30]

However, these studies mainly focus on the formation process of standardization (pre-standards' formation), without consideration of the management of standards after their formation is completed (post-standards' formation). In our study, we also present a roadmap to innovation after the standards' formation is completed so as to integrate R&D results and standardization activities more effectively and comprehensively. This knowledge could improve the results of R&D in social settings, reducing their management costs and increasing efficiency.

In Japan, de jure standards are prepared by the Japanese Industrial Standards Committee (JISC). Such standards are reviewed every 5 years to decide whether standards are to be terminated, revised, or continued. The review interval has been fixed to a 5-year period, regardless of technological differences for several decades. However, some standards may not need to be reviewed so often. Previous studies did not show the distribution of the lifespan of each standard and this led to a fixed review interval. If the statistical evidence regarding the lifespan of standards is provided, a more appropriate review interval can be considered, following the academic evidence. We can use the knowledge of the lifespan for the international standardization organizations such as ISO since ISO also has been using the fixed interval review system for several decades.

Producing new standards and then maintaining them requires both human and financial resources. Under the current JISC rules, standards are reviewed every 5 years. Is the fixed 5-year review interval the most adequate, in a scientific perspective? This is the fundamental research question of this study. Our results show that the standards in specific technological categories tend to have longer lifespans. These standards can be revised to make their review periods longer.

For the sake of this study, the lifespan, defined as the number of years between the establishment of a standard and its end, was the dependent variable in our model. Several factors supposed to be related to the lifespan are used for statistical analysis. Specifically, the following factors are considered: 1) technological category; 2) relationship with an international standard; 3) legal status (e.g., whether the standard has been incorporated into legislation yet); 4) revisions (e.g., revision of contents); and 5) type of standard. The relationship among these variables is defined as follows:

Lifespan of standard = f (technological category, relationship with an international standard, legal status, revision, type of standard). (1)

Technological category is supposed to have a relationship with lifespans because the product lifecycle (e.g., technology lifecycle) is related to the lifespan of standards. In addition, international standards are supposed to have an effect on lifespans because changes in an international standard, such as a standard of the ISO or IEC, can lead to corresponding amendments in the JIS system. The revision status of a standard may have a relationship with lifespans because revisions are presumed to lead to a renewal of the technology targeted by the standard. The type of standards may also have a relationship with lifespans because the production standards will no longer be necessary once a product has left the market.

For the sake of this study, the e-JISC, the electric database of reference for METI, was used. This database is used for administrative purposes, and it has been used for this type of analysis for the first time in this paper.

In this study, about 4500 JIS are surveyed. Our major contributions are as follows:

1) The lifespans of JIS in each technological category is first investigated and the data we obtained are shown in figures;
2) We found statistically significant differences in the marginal effects of technological categories on lifespans. As a result, the standards of certain technological sectors are observed to have longer lifespans than others. This evidence leads to support a flexible interval system;
3) Based on our results, we proposed a road map scenario to improve national innovation systems through the management of standards.

3 Literature review and hypothesis formation

3.1 Management of standards

The existing research on the management of standards is mainly focused on how to form standards.[31] How to manage already formed standards in terms of innovation systems has not been recognized as a fundamental research topic so far, for the following reasons:

1) The formation of standards is still the main interest among researchers and research on the management of standards is still in its introductory phase;
2) Lack of available data for the purpose of standards' research.[32]

3.2 JIS preparation process

JIS are mainly prepared to meet the needs of the private sector. Around 80–90 % of JIS are newly established or revised as a consequence of proposals from the private sector under Article 12 of the Industrial Standardization Act.[33] In the formation process of standards, a draft for the JIS is prepared by a group of interest. This draft is then submitted to a drafting committee whose participants are drawn from producers, users, and third parties. If this step is successful, then, as the next step, the confirmed draft is sent to JISC. Finally, JISC deliberates about the draft and the standard may be authorized.[34]

3.3 Effective terms of de facto standards

Several studies have focused on the effective terms of de facto standards, but they do not include de jure standards in their scope. Known as the most famous case study on the effective terms of de facto technology standards, David[35] investigates the standard of QWERTY typewriters. In his research, it was noted that such technology standards lasted for about 100 years without revision, not even after more efficient keyboard arrangements were developed. The key arrangement that was first developed is not the most efficient arrangement possible and was, in fact, designed to reduce typing speed. This feature of the design was important at the time of its introduction about 100 years ago because the typing speed of humans was faster than the mechanical capabilities of typewriters.

Today, nearly all typewriters have been replaced by personal computers. Inputs can even be provided to personal computers through a virtual touch screen keyboard instead of a physical keyboard. Hence, replacing the arrangement of keyboards would improve efficiency. Nevertheless, the QWERTY keyboard layout is still in use, even in touch screen interfaces. This case shows a lock-in effect, strong enough to effectively prevent changes in the basic interface of personal computers. David used this case to illustrate the persistency of standards.[35]

Another case study focusing on de facto standards in the fields of audio-visual and information technologies was conducted by Yamada.[36] This research showed that a de facto standard is established when the market share of a product reaches 2 %–3 %. David's research explained the persistency of standards in terms of a lock-in effect, focusing on human learning, but not all factors related to market dynamics were analyzed. Yamada's research gives some guidelines about the timing of formation for de facto standards, but not all determinants other than market share were discussed. In both cases, the focus is on de facto standards.

Although these studies discussed the effective terms of standards, they did not normatively discuss a scenario to improve the management system of standards, depending on their research results. Our study suggests a way to improve innovation systems through the effective management of standards.

3.4 Other related research

There are several related studies to be taken into account.

3.4.1 International standards

The relationship between international standards and international trade flows was studied by Blind and Jungmittach.[37] As for the consistency between JIS and international standards, about 6,000 of the 10,000 JIS were related to international standards.[33] Harmonizing with international standards has become more important after the introduction of the World Trade Organization's Technical Barriers to Trade (TBT) agreement in 1995. Since then, JISC has been promoting consistency between JIS and international de jure standards, such as those published by the ISO and IEC. However, the relationship between the JIS lifespan and international standards has not been studied yet, even after the TBT enforcement. We control for this effect in the evaluation of the influence of the technological categories.

3.4.2 Legal usage

JIS are used in some laws and regulations, such as the Pharmaceutical Affairs Act, the Fire Service Act, and the Human Resource Development Promotion Act in Japan. JIS are cited around 6,500 times in the Japanese law.[33] Nevertheless, the relationship between legal citations and JIS lifespans has not been studied previously. We control for this effect in the evaluation of the influence of technological categories.

3.5 Selection of relevant elements

3.5.1 Overview

The purpose of this study is to find a way to improve the management of standards. For this purpose, we need to identify which technological category influences the lifespan of standards. In addition, we need to introduce a set of control variables.

In this study, the hypothesis that technological category effects the lifespan of standards is formulated and examined through statistical estimation. The economic value of standards can be measured in several ways. Lifespan is a way of assessing their value. Under JISC rules, JIS are reviewed every 5 years; in the review, it is decided whether to terminate a standard or not, taking into account the opinion of the related industrial sector. This means that, if a standard does not seem to be needed in the 5-year review, such standards will be terminated. In this research, the lifespan of a standard is used as a proxy for the economic value of standards.

Although details vary across technological categories, the lifespan of a standard is supposed to be related to a certain stage in the product life cycle. When a product leaves the market, the related standard is supposed to be terminated. Each standard is associated with a specific technological category. In the JIS classification scheme, there is a category for management standards. Management standards are rule-related standards that are used in organizations and in society as a whole. This research includes management standards within the scope of its analysis.

3.5.2 Control variables

3.5.2.1 International standard

Some JIS were prepared on the basis of international standards to ensure standards to be domestically and internationally harmonized. In this analysis, "international standards" refers to ISO and IEC standards. When an international standard is converted into JIS, it is likely that there will be both positive and negative effects on the lifespan. The contents of the associated international standards are used in more areas and countries than in the case of JIS. Hence, the relationship with an international standard tends to produce a strong lock-in effect, and the standard is less likely to be terminated. Because of this, the lifespan of locked-in standards will tend to be longer. To control for this effect on the lifespan, a variable related to international standards needs to be included in the estimation of the model.

3.5.2.2 Legal usage

Some standards have legal effects, and one of the important roles of JIS is to provide national rules for Japan, where JIS represent the de jure set of standards. Some laws use JIS for quantitative regulation and for reference. As such, this usage requires stability, to be in line with the regulative purpose, hoping that such standards will stay in force. In addition, to change laws and administrative rules that are based on JIS, a formal process, typically involving Congress or the Cabinet, is needed. As a result, JIS in legal usage are usually thought to have a longer lifespan. To control for this effect on lifespan, a variable related to legal usage needs to be included in the model.

3.5.2.3 Revision

The revision of standards is likely to extend their lifespan because, when revisions are made, technological progress is incorporated into the revised standards. Hence, technological progress will be reflected in the contents of such standards, and, therefore, a revision should extend the lifespan of a standard. To control for this effect on lifespan, a variable related to revision needs to be included in the model.

3.5.2.4 Type of standard

The type of a standard may be related to its lifespan. For example, in the case of measurement standards, the described measurement method may be used to gather information about the quality of products. However, the need for standards concerning specific products will diminish as those products leave the market. Hence, measurement standards seem, in general, to be useful over a longer span than product-related standards. Nevertheless, it is also possible to conceive a relationship in the opposite direction. In industries where radical innovation is more frequent than incremental innovation, innovation in products and measurement cannot lag behind. Thus, innovation in products and measurement will happen together. When an obsolete product leaves the market, the associated measurement methods will also leave the market. In such industries, measurement standards may have lifespans similar to those of product standards. This means that technological replacement will be associated with the replacement of measurement methods. In short, in industrial sectors with frequent and radical innovations, measurement standards will be less static. For example, when digital media such as CDs (compact discs) were introduced, the technology related to analog storage media (like LP records) disappeared from the market. To control for this effect on the lifespan, a variable related to the type of standards needs to be included in the model. In addition to the categories such as 1) product and 2) measurement, there is the type of standards, which relate to a design and a mark. We formulate standards of the design and mark.

3.6 Hypothesis

To evaluate the effect of technological categories, we control for the above-mentioned elements. The following hypothesis is used in this study for the empirical analysis and scenario formation:

Hypothesis (H). The technological category of a standard affects positively or negatively the lifespan of a standard.

4 Method and Models

In this study, the relationship among the above-mentioned elements is statistically analyzed.

4.1 Model formation

We estimate the following regression to show the relationship among relevant elements and test the above-mentioned hypothesis. The dependent variable in the models is the lifespan, measured in years.

$$Model1: LIF = constant + \sum_{i=1}^{18} \beta i\ TECi + control\ variables + u, \quad (2)$$

where the following is referred:
control variables: ISO, LEG, REV, ESY, and ENY
LIF: lifespan of a standard;
TEC: category of a standard (dummy);
ISO: international standard status (dummy);
LEG: legal status (dummy);
REV: revision (dummy);
ESY: establishment year of a standard (ten-year interval categories (dummy));
ENY: end year of a standard (ten-year interval categories (dummy));
constant: constant term; and
u: error term.
In addition, to check the robustness of Model 1, we formulate Model 2, in which the type of standards is added as a control variable. In Model 2, to evaluate the effect of the type of standards (e.g., a) production; b) measurement; c) design and mark), we add an additional control variable (dummy variable), as follows:

$$Model2: LIF = constant + \sum_{i=1}^{18} \beta i\ TECi + control\ variables + u, \quad (3)$$

where control variables include ISO, LEG, REV, ESY, ENY, and TOS, and TOS is a dummy variable for the type of standards. All other variables are the same as in Model 1.

4.2 Method

The ordinary least squares (OLS) analysis is used to estimate the coefficients of both models and to test our hypothesis. The statistical package STATA is used for the estimation.

4.2.1 Dataset

In this study, we used data from the e-JISC, the electric database of reference for the METI officials. The e-JISC provides data including the information of the *JIS Handbook*.[38] For example, besides the contents of each standard presented in the *JIS Handbook*, the e-JISC provides data relating to JIS, such as, 1) starting time; 2) ending time; 3) amendment time; 4) title; 5) identification number in a database format. Currently, the e-JISC is prepared and maintained by METI and used for administrative purposes. For this research, we used the database under the academic cooperation between METI and RIETI (Research Institution of Economy, Trade and Industry). In this study, we use such information to build a new data set and we conduct a comprehensive analysis of the JIS lifespan for the first time.

At present, around 12,000 JIS are in effect, and in the past about 7,600 have been ended. Only standards for which complete data are available were chosen. We ended up with 4,483 standards (observations). We first analyzed the lifespan distribution in each technological category. The distributions are presented in Fig. 1. The distribution of the lifespan of standards varies across technological categories.

4.2.2 Variables

A detailed explanation of each variable is reported in Table 1. The categories specified in JIS were used as technological categories in our models, and dummy variables were introduced for each category in Table 2. The type of standards was determined from the description in the title of each standard, and categorized into 1) product; 2) measurement; and 3) design and mark. Dummy variables for the time when each standard was established (beginning) and the time when each standard was terminated (ending) were introduced, using ten-year intervals.

The dependent variable in the models is lifespan, measured in years. As shown in Table 2, we introduce the variables “c1” to “c19” corresponding to the technological categories of JIS. The variable “iso_iec” indicates the relationship with international standards. The variable “legal” indicates the use of a standard in legislation or for regulatory purposes. The variable “re” indicates whether a standard has been revised or not. For the purpose of controlling for the generation effect, we introduce the dummy variables “year10b#” and “year10e#” (where # represents an index), which represent the introduction and end years of a standard, respectively.
Among the categorical variables, c1 “A: Civil engineering and architecture” (technological category), year10b1 (starting year), year10e1 (ending year), and p_type (type of standard) are used as baseline categories for the empirical estimation. “A: Civil engineering and architecture” was selected as the default industrial category due to its adequate number of observations.

5 Results and discussion

Table 3 shows the descriptive statistics for all variables. The OLS regression results are shown in Table 4.

In Model 1, looking at the coefficient on the industrial sector, the categories (c8) “H: Non-ferrous materials and metallurgy” and (c13) “Q: Management system” are not statistically significant. The categories (c11) “M: Mining” and (c15) “S: Domestic wares” show a tendency to be significant ($p < 0.10$). The other sectors’ coefficients are all statistically significant. This result supports our hypothesis. Only the category (c17) “W: Aircraft and aviation” shows a negative coefficient, although this is relative to that of the baseline category (c1) “A: Civil engineering and architecture.” In Model 2, we use the type of standards as a control variable to check the robustness of the results of Model 1. In both Model 1 and Model 2, the significance of the results is the same. From the above results, the model is rewritten as

$$\text{Lifespan of standard} = \text{f (technological category (+/−))} \quad (4).$$

Standards are reviewed at 5-year intervals, but those standards that are likely to have a longer lifespan would benefit from longer review intervals. Among all technological categories, those with larger coefficients tend to have longer lifespans. Coefficients larger than 2 are highlighted in Table 5, and include (c6) “F: Shipbuilding” and (c2) “B: Mechanical engineering.” In terms of sectors showing a shorter lifespan, (c17) “W: Aircraft and aviation” is statistically significant, but the coefficient is about -1, which is not large in this context. The difference in the coefficients between the two industrial categories does not seem large enough to suggest shortening the review period.

As for the theoretical model of the lifespan of the standards in the dynamic innovation process, transitions to newer technologies occur after the market for a prevailing technology is saturated, and new standards are required corresponding to the emergence of new markets.[39]–[41] Repetition of the sequence generates sequential innovation. The observed result empirically shows that the time-series behavior of the innovation processes differs according to technological categories.

6 Future scenario and policy implications

In Fig. 2, we present a road map scenario for the future development and further integration of the elements presented. We propose a scenario that covers both short-term and long-term outcome goals. The current scenario, as implicitly shown in AIST research, only focuses on a short-term scenario. Our scenario includes both pre-standardization and post-standardization management, while the current

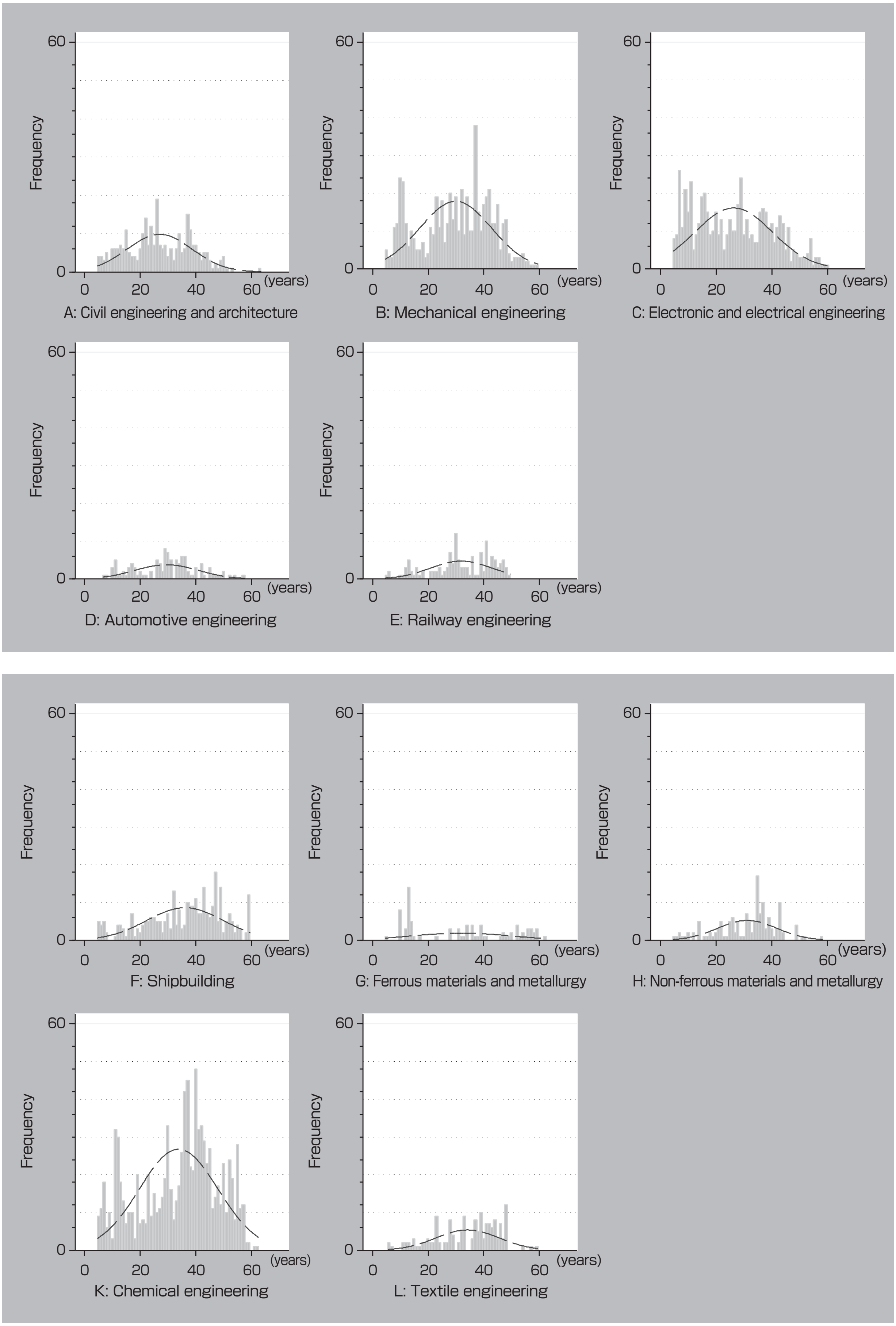

Fig. 1 Lifespan distribution of JIS standards

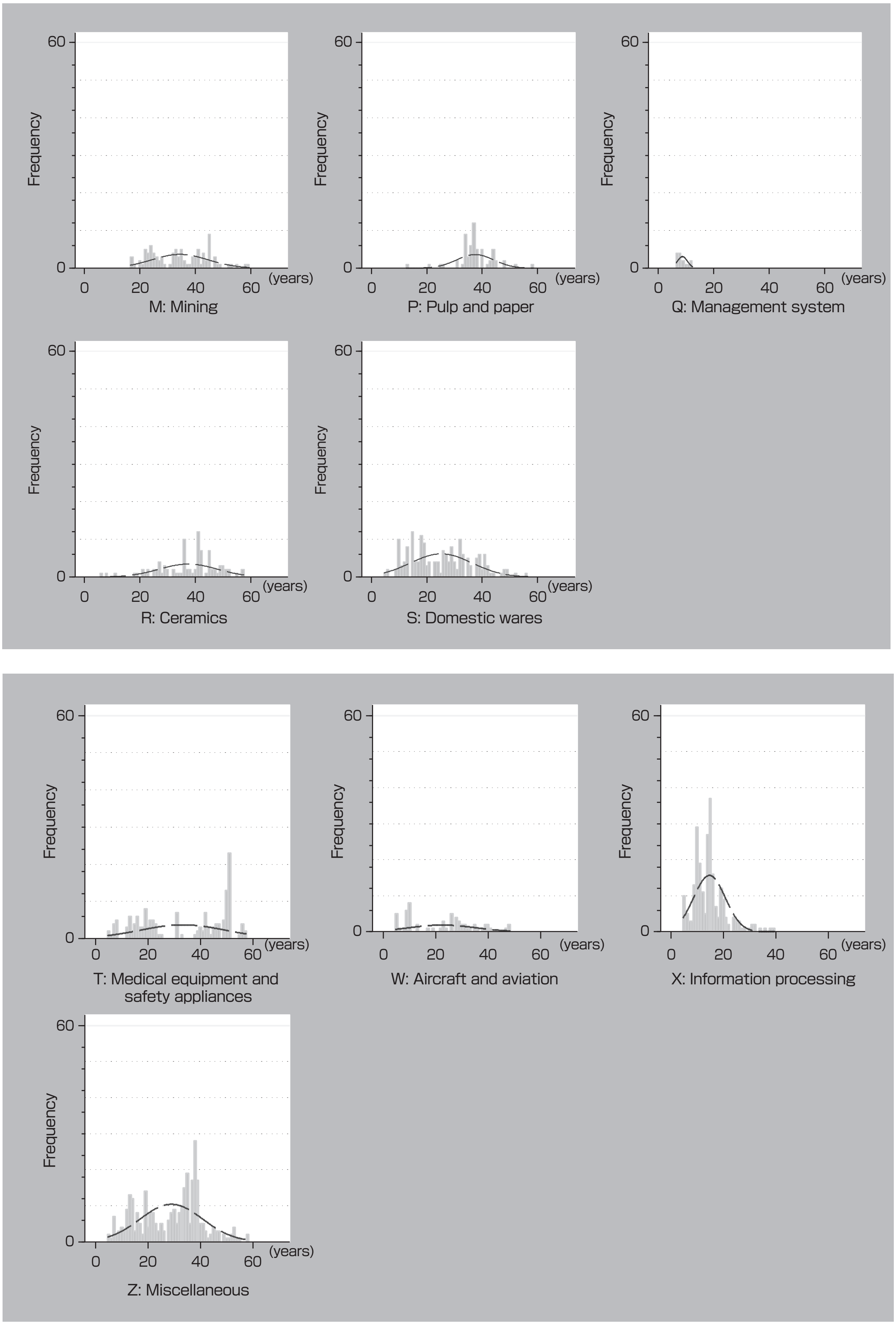

Fig. 1 Lifespan distribution of JIS standards

Table 1. Explanation of variables

Variable	Notation in analytical results	Explanation	Source	Notes
LIF	lif	*Lifespan of standard* : The number of years while the standard have been in place	Data from Japanese Standards Association (2011) and JISC data	Dependent variable
TEC	c1, c2, c3, ···, c19	*Technological category*: Dummy variable for technological category	Data from Japanese Standards Association (2011) and JISC data	Independent variable
ISO	iso_iec	*International standardization*: Dummy variable for international standardization	Data from Japanese Standards Association (2011) and JISC data	Independent variable Control variable
LEG	legal	*Legal status*: Dummy variable for legal usage	Data from Japanese Standards Association (2011) and JISC data	Independent variable Control variable
REV	re	*Revision*: Dummy variable revised or not	Data from Japanese Standards Association (2011) and JISC data	Independent variable Control variable
ESY	year10b#	*Established year* : The year when a standard is formed (ten year categorization basis; "#" is group number.)	Data from Japanese Standards Association (2011) and JISC data	Independent variable Control variable
ENY	year10e#	*End year* : The year when a standard is terminated (ten year categorization basis; "#" is group number.)	Data from Japanese Standards Association (2011) and JISC data	Independent variable Control variable
TOS	p_type, m_type d_type	*Type of standard*: Dummy variable for standard category: i) "d_type" indicates a design and mark standard; ii) "m_type" indicates a measurement standard; iii) "p_type" indicates a production standard.	Data from Japanese Standards Association (2011) and JISC data	Independent variable Control variable

Table 2. Alphabetic JIS technology code and technology area name

Alphabetic JIS technology code and technology area name	Corresponding independent dummy variable in models 1 and 2
A: Civil engineering and architecture	c1
B: Mechanical engineering	c2
C: Electronic and electrical engineering	c3
D: Automotive engineering	c4
E: Railway engineering	c5
F: Shipbuilding	c6
G: Ferrous materials and metallurgy	c7
H: Non-ferrous materials and metallurgy	c8
K: Chemical engineering	c9
L: Textile engineering	c10
M: Mining	c11
P: Pulp and paper	c12
Q: Management system	c13
R: Ceramics	c14
S: Domestic wares	c15
T: Medical equipment and safety appliances	c16
W: Aircraft and aviation	c17
X: Information processing	c18
Z: Miscellaneous	c19

Table 3. Descriptive statistics of variables

Variable		Obs	Mean	Std.Dev.	Min	Max
1.Independent variable						
lif		4483	30.01115	13.75334	5	63
2.Dependent variable						
c1		4483	0.06402	0.244815	0	1
c2		4483	0.128485	0.334667	0	1
c3		4483	0.125139	0.330914	0	1
c4		4483	0.024091	0.153349	0	1
c5		4483	0.029445	0.169068	0	1
c6		4483	0.066473	0.249136	0	1
c7		4483	0.018961	0.136401	0	1
c8		4483	0.031229	0.173956	0	1
c9		4483	0.211912	0.408708	0	1
c10		4483	0.034352	0.182152	0	1
c11		4483	0.020968	0.143294	0	1
c12		4483	0.013607	0.115866	0	1
c13		4483	0.003123	0.055802	0	1
c14		4483	0.020299	0.141037	0	1
c15		4483	0.038367	0.192103	0	1
c16		4483	0.033906	0.181007	0	1
c17		4483	0.013384	0.114925	0	1
c18		4483	0.052866	0.223792	0	1
c19		4483	0.069373	0.254116	0	1
	Control Variable					
	iso_iec	4483	0.152353	0.359403	0	1
	legal	4483	0.003569	0.059641	0	1
	re	4483	0.711131	0.453288	0	1
	d_type	4483	0.009146	0.095205	0	1
	m_type	4483	0.167076	0.373085	0	1
	p_type	4483	0.823779	0.381051	0	1
	year10b1	4483	0.002454	0.04948	0	1
	year10b2	4483	0.348204	0.476454	0	1
	year10b3	4483	0.227303	0.419137	0	1
	year10b4	4483	0.158822	0.365551	0	1
	year10b5	4483	0.119563	0.324486	0	1
	year10b6	4483	0.107963	0.310369	0	1
	year10b7	4483	0.03569	0.185538	0	1
	year10e1	4483	0.05242	0.222898	0	1
	year10e2	4483	0.498996	0.500055	0	1
	year10e3	4483	0.326121	0.468844	0	1
	year10e4	4483	0.122463	0.327856	0	1

Table 4. Estimation results

Dependent variable: lif		
Independent variable	model1 (coefficient/t-value)	model2 (coefficient/t-value)
1.Technological category		
c2	2.0567	2.0542
	[7.30]***	[7.28]***
c3	1.1003	1.0972
	[3.86]***	[3.84]***
c4	1.2082	1.2039
	[2.75]***	[2.74]***
c5	1.1824	1.1844
	[2.91]***	[2.91]***
c6	3.8369	3.8403
	[11.89]***	[11.85]***
c7	2.2717	2.2738
	[4.63]***	[4.64]***
c8	0.2524	0.2551
	[0.63]	[0.64]
c9	1.4889	1.4918
	[5.57]***	[5.58]***
c10	2.2951	2.298
	[5.88]***	[5.89]***
c11	0.8809	0.8738
	[1.91]*	[1.90]*
c12	2.6987	2.7002
	[4.95]***	[4.92]***
c13	-1.1582	-1.158
	[-1.08]	[-1.08]
c14	1.3432	1.3335
	[2.88]***	[2.86]***
c15	0.6355	0.6399
	[1.70]*	[1.71]*
c16	2.0533	2.057
	[5.15]***	[5.15]***
c17	-1.3009	-1.2956
	[-2.36]**	[-2.35]**
c18	1.0596	1.0524
	[2.83]***	[2.78]***
c19	1.2459	1.2466
	[3.90]***	[3.90]***
constant	35.6928	35.6594
	[29.04]***	[28.98]***
2.Control variable		
ISO	yes	yes
LEG	yes	yes
REV	yes	yes
ESY	yes	yea
ENY	yes	yes
TOS	no	yes
R-squared	0.9231	0.9231
Adj-R-squared	0.9226	0.9225
N	4483	4483

NOTE: [ ] t-value, * p<0.1, ** p<0.05, *** p<0.01.
Control variables: 1)international standardization(ISO), 2)legal status(LEG), 3)revision(REV), 4)established year(ESY) and 5)end year(ENY) are included in both models. Type of standard(TOS) is only included in the model2.

Table 5. Technology categories and coefficients

	Positive coefficient (Model 1 / Model 2)	Negative coefficient (Model 1 / Model 2)	Significant level (Model 1 / Model 2)	Notes
A: Civil engineering and architecture				c1(Base group)
B: Mechanical engineering	2.05/2.05		***/***	c2
C: Electronic and electrical engineering	1.10/1.09		***/***	c3
D: Automotive engineering	1.20/1.20		***/***	c4
E: Railway engineering	1.18/1.18		***/***	c5
F: Shipbuilding	3.83/3.84		***/***	c6
G: Ferrous materials and metallurgy	2.27/2.27		***/***	c7
H: Non-ferrous materials and metallurgy				c8
K: Chemical engineering	1.48/1.49		***/***	c9
L: Textile engineering	2.29/2.29		***/***	c10
M: Mining	0.88/0.87		*/*	c11
P: Pulp and paper	2.69/2.70		***/***	c12
Q Management system				c13
R: Ceramics	1.34/1.33		***/***	c14
S: Domestic wares	0.63/0.63		*/*	c15
T: Medical equipment and safety appliances	2.05/2.05		***/***	c16
W: Aircraft and aviation		-1.30/-1.29	**/**	c17
X: Information processing	1.05/1.05		***/***	c18
Z: Miscellaneous	1.24/1.24		***/***	c19

Note: Coefficients with absolute value greater than 2 are highlighted. (*$p < 0.1$, **$p < 0.05$, ***$p < 0.01$)

roadmap includes only pre-standardization management of invented technology. Moreover, the pre-standardization management mainly focuses on the R&D perspectives. Today, standards play an important role and, in some cases, standards are essential for the formation of new product markets. Standards play an important role in the dynamic change of the product life cycle. Innovation and standards are complementary to each other.[39]–[41] Previous research mainly discussed the first stage of the proposed scenario "1. R&D and standardization." In this study, we show that we can improve the scenario focusing on "2. Integration of relevant elements." Knowing that lifespan varies across technological categories, we can improve the management system of standards, focusing on the post-standardization phase. As a result, we can introduce a third stage "3. Improvement of the management system of standards," achieving more effective management systems for the established standards and timely market creation, and obtain "4. Improvement of the innovation system," which means the establishment of a platform for the management of standards for innovation systems.

We suggest the possibility to reduce the administrative cost of maintaining standards simply by allowing longer review intervals of standards. This is the key feature of the proposed stage "3. Improvement of the management system of standards." The current interval of 5 years could be extended for some categories, as suggested by the coefficients in our estimation results. The results from Models 1 and 2 suggest that the following industrial categories are ideal candidates for less frequent reviews: (c2) "B: Mechanical engineering;" (c6) "F: Shipbuilding;" (c7) "G: Ferrous materials and metallurgy;" (c10) "L: Textile engineering;" (c12) "P: Pulp and paper;" (c16) "T: Medical equipment and safety appliances." De jure standards are prepared and used across both developed countries and developing countries, even though de facto standards are established by corporations from developed countries. The aim of this research is also to help improve administrative systems based on de jure standards, including the ISO and IEC, around the globe. Such reforms would improve national innovation systems both in developing and developed countries, through the improvement of the management system of standards.

6.1 Theoretical contribution

We identified the key determinants of the lifespan of standards and the relationship as Lifespan of standard = f (technological category (+/−)). This result leads to a different treatment of standards across technological categories. As the timely creation of a market is essential in the current innovation system, a correct timing for standards' review is important and can lead to the implementation of more valuable standards. This is expressed as

$$\max \text{(Value of standards)} = f(t^*),$$

where t* is the equilibrium point of the review period in terms of the value of standards, in each technological category.

6.2 Managerial contribution

Our result shows that we can use the lifespan of standards as tools for the management of such standards, as in patent or copyright legal systems. We can reduce the administrative and related transaction costs for reviewing standards. We can adjust the current 5-year interval to longer intervals, for some categories, as our estimation results (Table 5) seem to suggest. Through this empirical analysis, a comprehensive management scenario for both the pre-standardization and post-standardization periods is presented for the first time as an explicit conceptual framework. This result applies to both the international standards' system in ISO and IEC and to each country's de jure standards' management system. Our result has potential global implications, since de jure standards are necessary tools in both developing and developed countries.

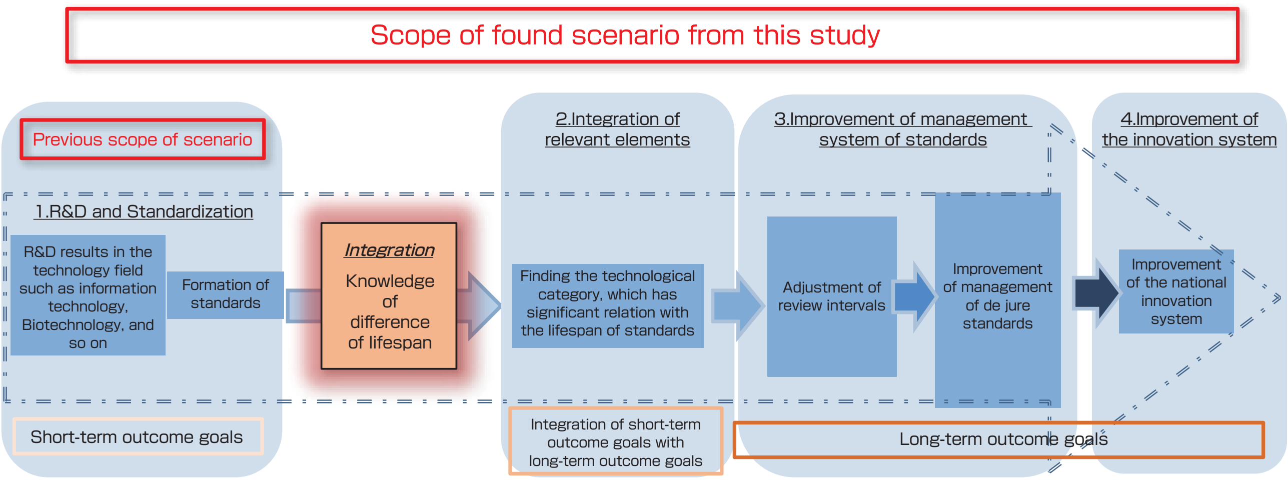

Fig. 2 Road map scenario for the improvement of the national innovation system through the management of standard

7 Further study

We study the general tendency of each technology sector. The next study will aim to investigate the lifespan and the reason behind each technology standard. For this, it is necessary to know the nature of the technology. The role of standards in terms of product life cycle should be discussed for each related product.

We proposed the scenario in Fig. 2. To improve the mindset for fostering innovation through the review term of the standardization, it is necessary to check the difference in the lifespan of each technology field when JISC plans the review schedule, which usually occurs on a yearly basis.

8 Conclusion

In our study, we presented a roadmap to innovation after the standards' formation is completed so as to integrate R&D results and standardization activities more effectively and comprehensively. For the purpose, this study focused on the lifespan of standards as the variable of interest. We normatively discussed the review interval of standards. First, we found an empirical relationship between the technology sectors and the lifespan of standards. This means that differences in technological characteristics have a strong influence on the lifespan of de jure standards. This is true for industrial sectors, such as (c6) "F: Shipbuilding" and (c2) "B: Mechanical engineering." We can optimize the review periods of standards following these results. In some contexts, extending the review interval can be appropriate. This may lead to a reduction in the cost of maintenance of standards and to adequate market creation. We also presented a road map scenario, focused on both the pre-standardization and post-standardization periods, to improve the national innovation system through the revision of the management system of standards, by referring to the research result of standardization in AIST. This result is beneficial to public agencies in Japan, as well as to international organizations, such as ISO, which deal with de jure standards with fixed review interval systems for several decades.

Acknowledgements

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Discussions with Reviewers

1 Overall

Comment (Naoto Kobayashi, Waseda University)

This paper presents original results, which are useful and interesting from the viewpoint of optimizing the review period of the standards that contribute to innovation. The process is expected to help the efficient management of standards. Therefore, this paper deserves to be published in *Synthesiology* owing to the improvement of the synthetic method of logical expression.

Comment (Hiroaki Tao, AIST)

By analyzing about 4500 JIS standards, this paper describes the influence of factors, such as technology categories, ISO standards, legislative application, review records, and the type of standards (design, measurement, and product), on the lifespan of standards. The paper is a valuable contribution to be published in *Synthesiology* as there have been no papers that address the lifespan of de jure standards and the policy implications for innovation systems based on the improvement of technology standards' management.

2 Relevance of lifespan to the standards' properties

Comment (Naoto kobayashi)

The lifespan of standards was investigated by introducing the technology category as an independent variable in the regression analysis. It is necessary to analyze the relevance of the lifespans to the properties of the lifespan distribution shown in Fig. 1. For example, in *C: Electronic and electrical engineering*, the frequency of the lifespan decreases almost monotonically toward the longer lifespan. In *T: Medical equipment and safety appliances*, it is found that many specific standards have a lifespan of about 50 years. I recommend that you describe the relevance of the results of your current analysis to the properties of standards in the corresponding technology category.

Answer (Suguru Tamura)

The aim of this study is to present a statistical analysis for the existent categories. It is possible, however, to study the characteristics of technology standards and their statistical properties in a narrower technology classification. Examining the reason why individual standards differ in age is a subject for future research. This point is added in Chapter 7, "Further research."

3 Revising the review interval of standards

Comment (Hiroaki Tao)

This study's proposition that innovation systems can be improved through the management of standards in addition to the formation of technology standards is novel and important. However, the resulting policy implications are limited to extending the review interval and, as a consequence, seem to focus only on the reduction of management costs. The recommendation is an obvious one. Could you present a recommendation on reducing the review interval that would improve innovation speed?

Answer (Suguru Tamura)

According to the results presented in Table 5, the coefficient is large enough to serve as evidence in support of our discussion on the policy implications. Several coefficients are positive and almost exceed the value two. This value corresponds to the situation where the review interval tends to be 50 % longer than the current five-year interval. Nevertheless, there are not enough large and negative coefficients. Hence, in the discussion on policy implications, we consider only the extension of the review interval.

4 Technology classification and review intervals

Comment (Hiroaki Tao)

In Fig. 1, *B (Mechanical engineering)* and *K (Chemical engineering)* seem to have two peaks. This suggests that setting a single and fixed review interval, which depends on the existing technology categories, is not rational. Could you comment on this?

Answer (Suguru Tamura)

Under the current system, reviews with a specific interval are a requirement. Hence, to formulate policy recommendations, it is necessary to consider a review interval. In our analysis, the review periods correspond to the pre-existent JIS technology categories, and the revision of the review period is suggested according to those categories. This suggestion depends on the existing framework for technology classification. In addition, we study the factors that affect the age of technology standards in each technology category as a whole. Let us consider the example of smoking and the health risk it poses from the medical point of view. There are smokers, who do not suffer from lung cancer, but, on average, the ratio of cancer sufferers is higher among smokers when we consider the difference between groups of smokers and non-smokers. When we contemplate this result in the context of policy implications and the policy framework, we consider the average figure for each group, rather than data on a single individual. This example illustrates that, for statistical analysis, the established group category is used in many cases. For classification purposes in our study, we follow the category of JIS technology standards, which has been used in the literature for a long time.

One may point out that, for example, to divide each existing technology category into subgroups and to decide the review interval according to the subgroup is theoretically possible. However, to achieve that, exploring other category classification criteria is essential. This essentially means searching for the reason for the different lifespan of each standard. We think that this is not within the research scope of this paper and the issue is discussed as a subject for further research in Chapter 7, "Further research."

5 Analysis concerning the characteristics of technology categories

Comment (Hiroaki Tao)

It is contemplated that the influence of ISO standards, legislative application, review records, and the type of standards on the lifespan is different in each technology category, but the differences in such an influence between technology categories does not seem to be analyzed in the present study. Is it difficult to address this in your study?

Moreover, emerging, mature, or obsolete technologies change over time differently in each technology category. It has been suggested that this influence manifests in the number of standards produced, the frequency of reviews, and the number of aborted standards. Is it possible to observe the technology transitions in each technology category?

Answer (Suguru Tamura)

In this study, we first control the influence of factors such as ISO standards, legislative application, review records, and the type of standards. Later, we discuss the influence of technology categories. When we observe the variance in age, we should recognize that the source of the variance is ISO or technology categories. Otherwise, our conclusions concerning the factors' influence and policy implications are erroneous. Therefore, we use the control variables to isolate the influence of the unintended factors. With this method, the factor of interest—the technology category—is analyzed separately. Certainly, if we were addressing a different research goal, we could observe the influence of ISO by treating the other factors as control variables, rather than as policy variables. The current research setting largely corresponds to our research goal. For a discussion on this theoretical issue, see, for instance, *Introductory Econometrics: A Modern Approach* by Wooldridge.

Finally, the control variables we used in this study are:

1. ESY, in order to capture the generation differences when standards were established;
2. ENY, in order to capture the generation differences when standards were abolished.

Through this treatment, we can estimate the influence of categories on age by excluding the influence of the generation background. On the other hand, we can estimate the influence of the generation difference by treating ESY and ENY as policy variables and including the technology categories as controls, although such setting diverges from our research goal. This analysis shows that, for example, the standards established in certain decades tend to have a longer—or shorter—lifespan than those established in other decades.

Development of EUPS for analyzing electronic states of topmost atomic layer

— Materialization of laser-produced plasma source application and EUPS observed fascinating surface —

Toshihisa TOMIE[1,2]* and Tomoaki ISHITSUKA[2]

[Translation from *Synthesiology*, Vol.9, No.4, p.216–234 (2016)]

A quarter century has passed since the principle of EUPS (extreme UV excited photoelectron spectroscopy) was invented as the most promising application of a laser-produced plasma source. EUPS enables analysis of electronic states of the topmost atomic layer, band bending of semiconductors, estimation of carrier density, and evaluation of electrical conductivity from secondary electron signals. These newly emerged analyses provide useful information for developing catalysts, protective insulators and other materials. These new analyses were born when problems needed to be solved were brought in by users. We can say that EUPS was sophisticated by the needs of users. In this paper, we describe the historical background leading to the invention of the principle of EUPS, the selection and development of the component technologies that constitute the EUPS system, and the birth processes of novel analyses that emerged.

Keywords : Laser-Produced Plasma, EUPS, time-of-flight, topmost atomic layer, band bending, carrier density, secondary electron

1 Introduction

1.1 EUPS pioneers new applications for photoelectron spectroscopy

In 1992, extreme ultraviolet-excited photoelectron spectroscopy (EUPS) was devised as an application of laser-produced plasma (LPP) sources.[1] Since then, for a quarter of a century, we have been developing technologies for making EUPS a practical analyzing tool and pioneering applications of photoelectron spectroscopy. As we worked on gaining users to spread the use of EUPS analysis and met users' demands, new potential of EUPS were pulled out, and it became possible to conduct various analyses that are difficult or impossible with conventional photoelectron spectroscopy. It is a joy of the inventor to watch that the much wonderful potential of EUPS has become clear. The devices to be developed in the future to maintain and advance people's quality of life will require innovative functions, not just simple improvements. We expect EUPS will be one of the innovative analysis technologies that will promote technological innovations.

Currently, the EUPS system as shown in Fig. 1 is disclosed and offered for use to general users, as one of the analysis devices of AIST Nanocharacterization Facility (ANCF) which is a part of the Nanotechnology Platform Project[2] of the Ministry of Education, Culture, Sports, Science and Technology (MEXT). In this paper, we describe the process that led to the practical use of EUPS, the configuration of EUPS, and the new analysis methods pioneered by EUPS.

In Subchapter 1.2, we outline the history of LPP application research up to the creation of EUPS, and in Subchapter 1.3, the characteristic of LPP sources will be explained. The ideas for making EUPS as a practical device using an LPP source is explained in Chapter 2, the history of prototype units that led to the current working unit is described in Chapter 3, and the details of the elemental technologies that configure the current working unit will be presented in Chapter 4. The new analyses methods pioneered by EUPS will be introduced in Chapter 5.

1.2 History of the research of laser-produced plasma (LPP) sources

First, we present the history that led to the creation of EUPS that is probably the only one LPP application technology that has been put to practical use.

When a pulse laser is focused on a solid, an ultrahigh temperature state is achieved since the energy is injected before cooling by thermal conduction occurs. Several million degrees centigrade can be achieved. In such high temperature conditions, the solid becomes plasma consisting of electrons and multivalent ions. Since the density is high and the temperature is high, ultrahigh pressure is also achieved. Plasma is ejected at high speed, the solid target that is irradiated with the laser is pushed by the reaction,

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and this force is usable as the propulsion of rockets in space. The idea for inertial confinement fusion, where the nuclear fusion is caused by creating an ultrahigh density condition by compressing the interior material by the reaction of exploding plasma, became realistic after the publication on laser oscillation by Maiman.[3] The paper published in *Nature* by Nuckolls *et al*.[4] in 1972 stated that there is a possibility that a breakeven, where the input energy and nuclear fusion output energy become equal, can be achieved with 1 kJ of energy, and this sent a shock around the world. Perhaps also because it was immediately after the Oil Crisis, three labs at the Electrotechnical Laboratory, which is one of the precursors of AIST, formed a group to tackle the inertial confinement fusion research. One of the authors (Tomie) was employed as one of the researchers, and was involved in the development of a high-power glass laser system for inertial fusion. In conducting the research for the glass laser oscillator, he discovered and clarified a phenomenon of pulse compression in a laser resonator.[5] This is the phenomenon of the principle of femto-second laser oscillation that emerged later. Initially, it was said that nuclear fusion was possible with a 1 kJ pulse laser, but later it was found that several MJ was necessary. The author felt that the research requiring such enormous laser power could not be continued by a small research group, and shifted the direction of his research to application of plasma using the high-energy laser he developed.

Shortly after the introduction of the laser, keV X-ray generation from LPP was observed,[6] and one calculation showed that the conversion efficiency would be comparable to X-ray tube X-ray source including the conversion efficiency from electricity to laser energy.[7] There was much research on using LPP as a high-intensity X-ray source. The author tried some approaches of application research. X-ray proximity lithography[8] was proposed as a shrink printing method of semiconductor circuits, and the development of its technology was conducted mainly using synchrotron radiation, and LPP was a candidate light source. The first LPP research conducted by the author was the use as a source of the X-ray proximity lithography. The conclusion that an X-ray reflecting mirror which can collect the X-rays irradiated almost isotopically was an essential requirement to utilize LPP as an industrial light source was the achievement of the author’s research done in a very short period.

His next research was the X-ray microscopy to observe fine structures of biological cells. In living conditions, cells contain a large amount of water. By using the X-rays with wavelength range between 4.4 nm (K absorption edge of carbon) and 2.3 nm (K absorption edge of oxygen), the distribution of carbon that composes the biological cell structure, *i.e.*, the structure of a cell, can be viewed by suppressing the effect of water (oxygen). In this sense, this wavelength range is called the “water window.” To observe biological cells with an electron microscope, the process of removing water is necessary, but, when the “water widow” X-rays are used, the structure of the cells can be observed in its living conditions. Much X-ray microscopy research was conducted using strong synchrotron radiation.

The reason why LPP is important as an X-ray microscopy light source is because it is an ultrahigh-intensity light source as well as a pulse source. When obtaining X-ray images by X-ray absorption, the energy density absorbed by the sample increases inversely proportional to the fourth power of spatial resolution.[9] The absorption energy density at 1 μm spatial resolution reaches several Sv (J/kg) that is a fatal dose for cells. That is, in medical diagnosis, the resolution cannot be increased too high. LPP can provide such ultrahigh-density

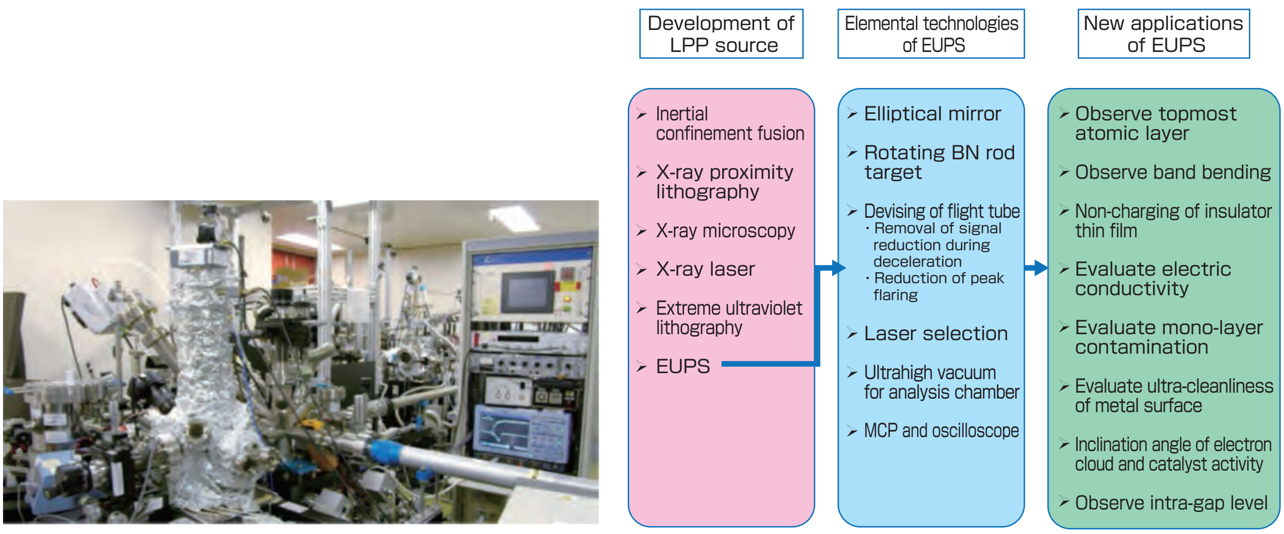


Fig. 1 EUPS Unit 2 that is available for sample analysis, and the process, configuration, and new applications of EUPS development

X-rays in an extremely short pulse of nanoseconds. At spatial resolution of several tens of nm, the necessary doze is too high and cells will instantly explode. The author *et al.* showed that a biological cell evaporated at about 60 ns using stereo X-ray photography.[10] A structure of about 50 nm was visible in the contact exposure X-ray image using LPP of a pulse width of 0.5 ns,[11] and this might have been the true structure before its destruction by thermal shock.

In LPP that has high temperature of about one million degrees and in which the electron state changes rapidly, population inversion can be generated in the vacuum ultraviolet region[12][13] and this enables the so-called "X-ray lasers." The author (Tomie) joined the X-ray laser research[14][15] team in the UK for one year in 1986, gained experience and returned home, and started research in Japan. To produce LPP for "water window" X-ray lasers, a supergiant laser system as large as we can build only one in Japan because of cost was necessary. Although it could give information of structures of living cells that was impossible with other methods, the issue was whether the use of extremely expensive giant-high-power laser system can be justified. This was also the issue faced by X-ray laser researchers around the world.

An international conference to seek the possibility of X-ray laser application to areas other than X-ray microscopy was held in the USA.[16] By reading the proceedings, the author learned of the potential of application to surface research, and stated at international conferences that the best application of X-ray lasers was to photoelectron spectroscopy.[10] The reason is because it takes the best advantage of X-ray lasers, whose greatest characteristic is extremely narrow spectrum width. In the X-ray lasers for X-ray microscopy of living cells, wavelengths outside the water window region are meaningless, high repetition rate operation of X-ray lasers is meaningless since the sample is destroyed in a single shot, and the pulse width needs to be 1 ns or less. In photoelectron spectroscopy, any wavelength can be used It is important to have a high repetition rate to obtain a large cumulative photon number in reasonably short time because decent quality spectra need large total number of detected electrons.

In the application to photoelectron spectroscopy, it is not necessary to use X-ray lasers as the light source, and an LPP source is sufficient. Moreover, downsizing is an important requirement as a practical system. The author's (Tomie) research shifted to the practical use of photoelectron spectroscopy using an LPP source from X-ray lasers, and this research is being continued to the present.

In the middle of the 1990s, extreme ultraviolet lithography (EUVL) appeared as a major application of LPP.[17] Today's information society is realized by the great progress of semiconductor technologies which was achieved by the increase of the integration by reducing the size of semiconductor circuits. This was enabled by the progress of lithography technology which prints in reduced size the circuit pattern. The progress of lithography technology was made by shortening the exposure wavelength. The shortening of the wavelength went from 248 nm of KrF lasers to 193 nm of ArF lasers, but due to the limit of the transparent wavelength of optical materials, further shortening was difficult, and a big jump of wavelength to EUV that uses reflection optics was required. The Mo/Si multilayer mirror gives a high reflectivity at 13.5 nm, and tin (Sn) LPP was selected as the source. Technologies of EUVL were developed in very big national projects in USA, Europe, and Japan. Currently, it is reported that 13.5 nm power surpassing 100 W has been obtained. Although this is an amazing figure, there is no semiconductor element made by EUVL on market yet. That is because the speed of progress of light source development failed to meet the demand of increased average power for rapid reduction of the feature size of semiconductor circuits. For the introduction to mass production plants, it is now said that several hundred watts are necessary. The author (Tomie) who was one of a few researchers working on LPP sources for X-ray microscopy was involved in EUVL since the middle of the 1990s.[17] As a person deeply involved in EUVL research for over 20 years, I hope that the increasing demand in EUV power stops and the EUVL may eventually be set for practical realization.

1.3 Characteristics of LPP source

The advantages of the LPP source[Footnote 1] include the facts that: the spectrum can be changed by selecting target material, it is short pulse, its brilliance is extremely high as shown in Fig. 2,[18] it is a point source, and others. The disadvantages are: large amount of debris contaminants (fine particles and plasma) are released as shown in the top photograph of Fig. 2, duty ratio of emission is extremely low, collection of radiation is not easy since the source emits nearly isotopically, and others.

Figure 2 shows the comparison of peak brilliance of several X-ray sources. The peak brilliance is defined by the number of photons emitted per unit solid angle per unit bandwidth per unit time from the unit surface area of the source. The data shown as black dots for LPP are values obtained in the experiments conducted by the authors, and brilliance was close to the blackbody radiation of 200-eV. This brilliance is two to three orders of magnitude larger than that of bending magnet synchrotron radiation. Compared to the X-ray tube source, LPP is 10 orders of magnitude brighter. The undulator synchrotron radiation is five orders of magnitude brighter than LPP. While the duty ratio of emission is extremely low, far lower than LPP, and the time averaged power is quite low, high-order harmonics of femtosecond lasers and X-ray lasers using LPP can have similar peak brightness to the undulator.

In the application to EUVL,[Footnote 2] the important characteristic of LPP is that it is a point source. The disadvantage of a divergent source was overcome by the development of a collecting mirror with an extremely large collecting solid angle as large as π steradian. The disadvantage of short emitting time is overcome by operating the source at high repetition rate of 10 kHz or higher.

In all LPP applications, the biggest issue is suppression of contamination of surrounding optical elements by fine particles and fast ions generated from the target. Also in the development of LPP for EUVL, the greatest obstacle for commercialization was contamination control. Historically speaking, full development of EUVL was started by the birth of an idea of using liquid Xe as a target material of LPP. Xe atom does not stick to optics at room temperature and optics will not be contaminated.[17]

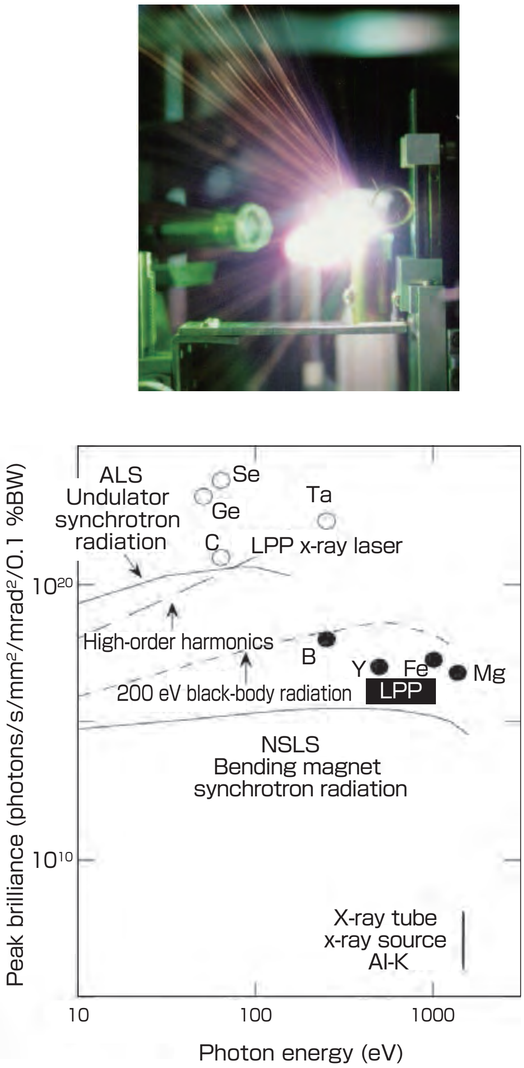


Fig. 2 [Top] Photograph of LPP. Greatest issue for practical application is stopping debris contamination. [Bottom] Comparison of peak intensity of various sources.[18]
LPP is a source with intensity 2–3 orders of magnitude higher than bending magnet synchrotron radiation and nearly 10 times higher than X-ray tube. However, emission time is as short as several nanoseconds, and the time-average power is extremely low without clever ideas

Concerning the contamination by plasma, attention is often placed only on the adhesion of atoms of the target material, but in fact, sputtering of surrounding materials by high-speed ion is more serious. In a discharge plasma source, the deterioration of electrodes by sputtering is severe. Tin is notorious as a dirty material because it adheres, but it is optimal as a material for EUVL. One of the reasons is because it is possible, in principle, to balance the adhesion of tin vapor to the surface of a multilayer reflector and the sputtering by high-speed ions. The lifetime of multilayer mirrors is extended using this principle.

2 Proposal of EUPS

2.1 Requirements of the photoelectron spectroscopy light source

As shown in Fig. 3, photoelectron spectroscopy gives information of the electronic states of a material by measuring energy of electrons released from the substance by photoexcitation. In comparison with the Auger electron spectroscopy that employs electron excitation, the good point of photoelectron spectroscopy is the high-energy resolution. In most analysis, resolution of about 1-eV is sufficient. Ordinary resolution XPS[Footnote 3] widely used in material analysis is 0.8-eV, and it is 0.3-eV in high-resolution XPS. Incidentally, resolution of 0.3-eV is confirmed for EUPS.

To obtain a photoelectron spectrum with high energy resolution, an excitation source should have a narrow bandwidth and an electron spectrometer needs to have a

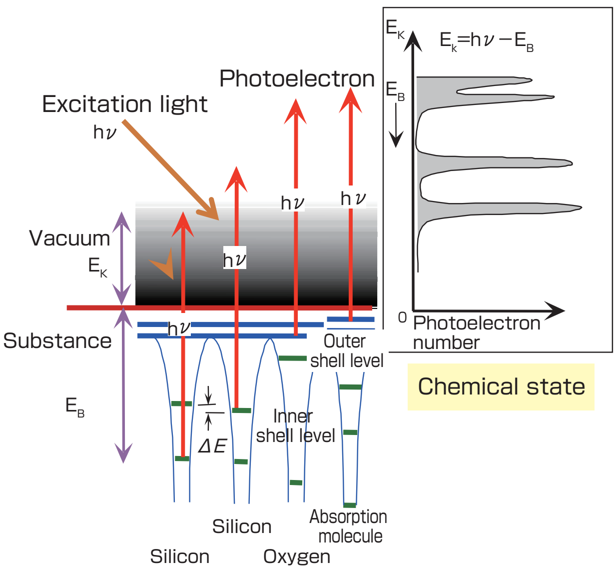


Fig. 3 Principle of photoelectron spectroscopy
Because the best characteristic of photoelectron spectroscopy is high energy resolution, narrow bandwidth of the excitation light and high energy resolution of electron spectroscopy are required.

high-energy resolution. To reduce statistical noise, large cumulative counts are necessary.

The research of photoelectron spectroscopy using LPP as an excitation source was conducted by a different group,[19] and the accumulation time needed to obtain photoelectron spectra was extremely long. If the laser pulse width is 10 ns and the emission time is the same, duty ratio of emission is only one ten-millionth of the time when repetition rate is 10 Hz. Even if a laser can be operated at 1 kHz the emission duty ratio is one hundred-thousandth. As seen in Fig. 2, even if the instantaneous peak brilliance of LPP is extremely high, the time averaged photon flux is low. The response we received when we presented the principle of EUPS at an academic society was “LPP would never be a practical source for practical photoelectron spectroscopy.”

2.2 Devising the EUPS

In EUPS, we overcame the disadvantage of LPP that the duty ratio is small by two ideas. One was not to use a spectrometer to obtain a narrow bandwidth light for excitation. The other was to use the time-of-flight (TOF) method for analyzing electron energy.

In a synchrotron radiation facility, the photon flux on the sample is considerably lower than the flux of the source. The reason is that the transmission of a diffraction grating spectrometer to obtain a narrow bandwidth light from the continuous spectrum light is extremely low, and the light intensity decreases by four to five orders of magnitude by passing through the spectrometer. With an LPP, a narrow bandwidth line can be generated by appropriate selection of target material and laser irradiation conditions. Although a single line spectrum cannot be obtained, as shown in Fig. 4, almost a single narrow bandwidth line spectrum can be selected using an appropriate filter. Since this eliminates a spectrometer, photon flux on sample at 10 Hz repetition rate can be comparable to that in a synchrotron facility.[18]

The use of TOF electron energy analysis changes the short pulse nature of LPP from a disadvantage to a big advantage.

In TOF, as shown in Fig. 5, the electrons produced in a short pulse fly a certain distance, are detected by detectors with a high temporal resolution, and the velocity distribution of electrons is recorded.

When the electrons are produced continuously in time, the energy of electrons is measured using the fact that the degree of bending of the trajectory of electrons by an electric field differs per the speed of electrons. The electric field is applied between the electrodes having double hemisphere or a double cylinder structure. Multiple electron detectors can be placed to detect electrons having different energies, but basically, only the electrons with one specific energy are detected. Therefore, the detection efficiency is extremely low, because all other energy electrons are not detected and thrown away. On the other hand, in TOF, electrons of all energies are detected in one measurement, and the detection efficiency is high. In principle, 100 % detection efficiency is possible in TOF. To suppress the radiation damage of material by inner shell excitation that is thought to occur after 10,000 excitations of one atom, high detection efficiency is absolutely important.[18] If the efficiency of collecting electrons is high, the excitation intensity of the sample is reduced, and then, the employment of the TOF method is effective also to suppress charging when observing insulators.

Being a pulse source was considered a disadvantage of LPP since the duty ratio is low, but as explained above, it is, in fact, an important advantage in the application to photoelectron spectroscopy since the TOF method having a high detection efficiency can be utilized. There were works of research on TOF electron spectroscopy using high-order harmonics of femtosecond laser pulses as an excitation source,[20] but EUPS is the first to utilize TOF with LPP as a light source.

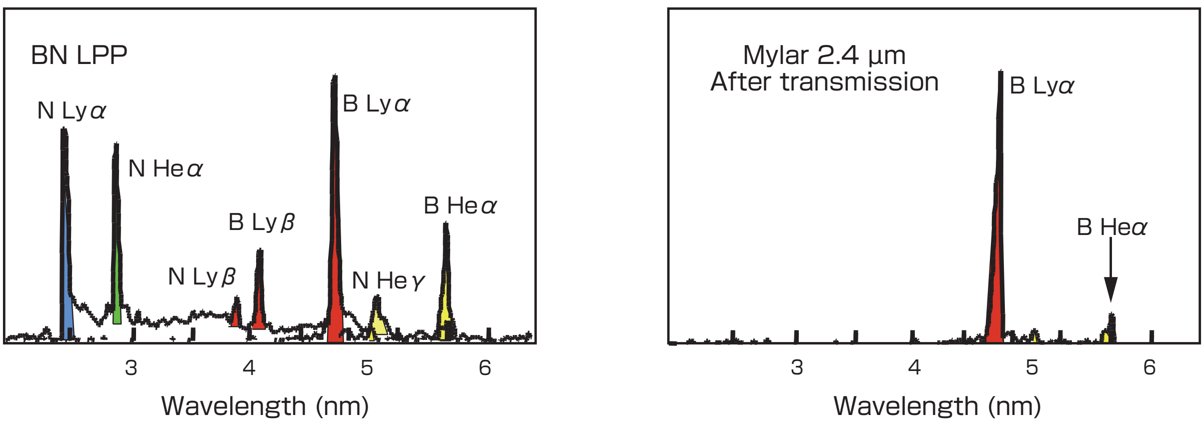


Fig. 4 LPP source spectra used in the EUPS[21]
The emission spectrum of boron nitride LPP (left) becomes almost monochromatic (4.86 nm) after passing through a Mylar film (right). Spectral loss for obtaining a narrow line is very small

3 History of the EUPS prototypes

The basic principle of EUPS is described in the patent,[1] but development and advancement of several elemental technologies were necessary for making it a practical analyzing tool.

The target material of LPP for EUPS is uniquely determined. As mentioned above, the biggest issue in any LPP application is the shielding against contaminants. In photoelectron spectroscopy, it is necessary to avoid contamination of even a mono atomic layer on the sample surface, and the use of a filter that completely shields from all substances emanating from the plasma is mandatory. Since EUV light must pass, the first requirement is that the filter must be ultrathin. On the other hand, since it must block fast ions and fine particles, the second requirement is that the filter must have certain mechanical strength with certain thickness. The filter is contaminated and damaged by ions and debris from a target, and the filter must be exchanged frequently. Therefore, the third requirement is that the filter must be low in cost. In practical use, this is extremely important.

As a material fulfilling these three requirements, we selected a carbon polymer film. Mylar, which is also a product name, can be purchased as a film of thickness of 1.4 µm at a reasonably low price.

Light of wavelengths near the long wavelength side of the K absorption edge of 4.4-nm of carbon can pass through a Mylar film at high transmissivity. The wavelength of the Lyman α line of hydrogen-like boron ions is 4.86-nm and is very compatible with Mylar. There are other emission lines emitted from a boron plasma, but they are absorbed strongly by a Mylar filter, and their intensities are negligible in practice. Boron nitride (BN) including boron can be purchased in a shape of rods. The emission of nitrogen plasma is at the short wavelength side of the K absorption edge of carbon, and it is largely absorbed by the Mylar film and does not greatly affect the photoelectron spectrum measurement.

The fact that a boron plasma emits a strong line in the wavelength region that can pass through an inexpensive Mylar film of sufficient thinness, and boron is supplied in a rod form at a reasonable price was a gift from god to us for the realization of EUPS.

To clearly state that our photoelectron spectroscopy is different from conventional ones, in 2001, we named our scheme as "EUPS" after extreme-ultraviolet-excited-photoelectron spectroscopy.

3.1 Prototype 1

The development of EUPS was started from the middle of the 1990s. In the prototype Unit 1 for the verification experiment of the basic principle, the sample to be measured was placed in a magnetic shielded flight tube of 50-cm length, a target for LPP was placed close at a few cm, and plasma was produced by focusing a pulse laser on the target.[21] The pulse energy was 100-mJ. A BN plate was used as the target material, a Mylar thin film was sandwiched in a very narrow space between the sample and the plasma, Lyman α line emission of the hydrogen-like boron ion was selected by the Mylar filter. Photoelectrons from Si2p with the kinetic energy of about 150-eV was slowed down to about 15-eV by applying about 135-V to the flight tube, hence the flight time was lengthened by reducing the speed to increase the energy resolution that was limited by time resolution. Chemical shifts of Si2P electrons in the Si substrate, SiO_2, and SiN films were observed separately in photoelectron spectra obtained by summing up the time waveforms of 96 shots,[22] Thus the principle of EUPS was confirmed. The Mylar film worked as a vacuum separator, and the vacuum of the plasma space was about 10^{-3} Pa and the

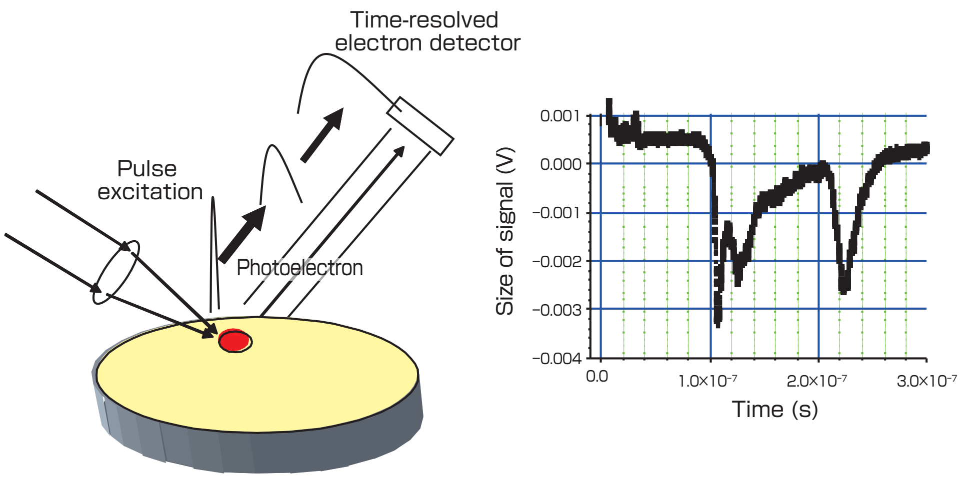


Fig. 5 Detection efficiency of TOF energy analysis is very high since particles of all energies can be detected simultaneously

vacuum of the sample space was about 10^{-4} Pa.

Through the experience with Unit 1, we could extract the issues to be solved for the realization of EUPS. The hardest thing in operating Unit 1 was the exchange of Mylar films used for wavelength selection and vacuum separation. In the Unit 1, it was necessary to set the LPP close to the sample to not weaken the EUV intensity on the sample. The Mylar film then became quickly contaminated with the debris from the target and had to be exchanged after a short time operation. Moreover, since the space between the sample and the plasma was very narrow, the exchange of the Mylar film was extremely difficult.

3.2 Unit 3 and Unit 4

After the experience with Unit 1, we fabricated Unit 2 that is currently in operation. The elemental technologies were improved as explained in the following chapters, and the current unit does not have any elements of the first Unit 2, but the basic configuration is unchanged. The prototype units with different configuration from Unit 2 were also fabricated, and we shall first describe them briefly.

In Unit 3, the efficacy of the magnetic bottle[Term 1] was investigated as it was expected to increase the capture efficiency of the electrons. The expected performance was not obtained and we decided not to adopt a magnetic bottle.

In Unit 4, the spatial resolution was pursued. Using a Schwarzschild collector optics,[Term 2] submicron beams were formed, and we succeeded in obtaining photoelectron images with a spatial resolution of less than 1 micrometer

On the surface of the convex and concave mirrors that configure a Schwarzschild mirror, it is necessary to form a multilayer film to have a high normal-incidence reflectivity. The peak reflection wavelength is at a slightly long wavelength side of 13-nm in the case of Mo/Si. Wavelength close to 6.7-nm of the La/B4C multilayer film can be also employed as an excitation light although the multilayer technology is not so mature for Mo/Si. Since it is difficult to emit an isolated, strong line emission at these wavelengths, the energy resolution of photoelectron spectra is not high when using Schwarzschild optics. Also, the collected solid angle of the source emission is small, the photon number on sample per shot is small, and a high repetition rate over 10-kHz is necessary for the source to attain a practical measurement speed. In the range that we conducted the survey, we could not find strong demands that matched the difficulty of the technology for spatial resolution of one micron. So, we stopped the development of the system using a Schwarzschild collector mirror.

4 Elemental technologies that configure the working unit

The EUPS Unit 2 was designed following the experiences gained from Unit 1, and it is currently in operation. There were three major changes from Unit 1. One was to insert an elliptical mirror between the sample and the plasma. Second was that the form of the target for LPP was changed from a plate to a rod. Third was to set the flight tube vertically. The schematic configuration of Unit 2 is shown in Fig. 6.

4.1 Use of elliptical collecting mirror

In a practical photoelectron spectroscopy system, continuous operation over a long time is mandatory. This is an extremely difficult requirement in the application of LPP that releases large amount of contaminants. It is necessary to dramatically reduce the deposition rate of contamination on the film used for vacuum separation. The primary method is to increase the distance between the plasma and the vacuum separation film.

Therefore, we used a configuration where the image of the plasma is transferred to the sample using an elliptical mirror with a long axis length of 70-cm. Since sufficient space was made between the elliptical mirror and the plasma, the Mylar film was set at 10-cm from the plasma. After some devising, currently, measurements can be done without exchanging the Mylar film for about one week.

To increase the reflectivity of 4.86-nm light, a Ni/C multilayer film was formed on the elliptical mirror, and the design value is peak reflectivity of 25 % with a bandwidth of 0.5-nm.

4.2 A rotating BN rod target

When the plasma is produced by focusing a pulse laser on solids, the area is heated to high temperature and blows off and a hole of several tens of microns diameter is formed. When the laser is focused on the hole, the density of the produced plasma is low and the emission intensity decreases greatly. To maintain the emission intensity, it is necessary to focus the laser on a flat surface. In Unit 1, the position of a BN plate was scanned for generating LPP on a flat surface, but since the scanning mechanism of the plate was complex and a large space was necessary, the form of the target was

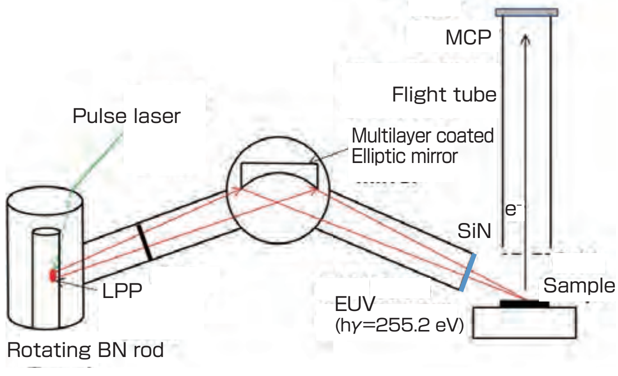


Fig. 6 Configuration of EUPS Unit 2

changed to a rod in Unit 2. In Unit 2, by rotating and moving a rod up and down (spiral motion), one rod is now employed for about one week without exchange.

4.3 Devising on the flight tube

The flight tube set horizontally in Unit 1 is set vertically in Unit 2, which allows setting a sample horizontally. By setting the sample holder horizontally, transferring a sample from the load-lock chamber to the analysis chamber became easy. The manipulator that holds a sample has five degrees of freedom, and as will be explained in Subchapter 5.6, the angle of incidence of 4.86-nm light on the sample can be changed. In ordinary EUPS measurement, in order to increase the photoelectron signal, the angle of incidence is set at 10 degrees from horizontal.

When the mass of electrons is set as m_e and the kinetic energy as E, the velocity v is $v = (2E/m_e)^{1/2}$, and the time of flight t when flight distance is L is $t = L(m_e/2E)^{1/2}$. The speed of electrons with the kinetic energy of 100-eV is 6×10^8 cm/sec, and when the flight distance is L = 48-cm, the time of flight is t = 80-ns.

The energy resolution ΔE when time resolution is Δt is

$$\Delta E = -2(\Delta t/t)E \propto \Delta t E^{3/2}$$

and when the time resolution is Δt = 3-ns, the energy resolution for 100-eV electrons is $\Delta E = 2 \times 100 \times 3/80 = 7.5$-eV. This is insufficient for photoelectron spectroscopy. If 100-eV electron is decelerated to 10-eV, the energy resolution is ΔE = 0.24-eV that is sufficiently high in energy resolution. To obtain high energy resolution, the electron is slowed down in TOF. If the sample is grounded, by setting the potential of the flight tube at −90-V, the 100-eV electron is decelerated to 10-eV.

The energy resolution increases by decelerating the electrons, but we faced a problem that the signal decreased when the electrons decelerated. Also, it was found that the low energy side of the peak flared out to disfigure the waveform. These problems that inhibited the obtainment of a good photoelectron spectrum were solved by the two patented methods.

4.3.1 Solving the problem of reduced signals when electrons are decelerated

The reason the signals decreased when the electrons were decelerated was considered to be that the electrons heading for the MCP detector would go toward the wall (and therefore the solid angle detected by a detector would become small), because the speed in the direction toward the wall did not change while the speed heading for the MCP[Term 3] detector was greatly suppressed. To solve this problem, we fabricated decelerating electrodes having a curvature, as shown in Fig. 7.[23] If electrons are generated at the center of the curvature, the direction of the motion of electrons will not change by deceleration, the collecting solid angle of the detector will not change, and the signal intensity will not change. If the electron source is set outside the curvature center, due to the retarding electric field, the velocity component heading to the cylinder wall will further decelerate, enabling “convex lens effect” that bends the electrons to the center direction of the cylinder, and thereby the signal becomes larger.

4.3.2 Solving the problem of the flaring peak

The other problem was that a large slowly decaying signal accompanied after a peak which should decay sharply. The origin of this signal was unknown for several years, but through some experiments we identified it as originating from the flight tube wall.

The photoelectrons emitted from a photo-irradiated sample are released at wide angles. Some of the electrons directly arrive at the MCP detector, while the majority of the electrons hit the flight tube wall. Then, new secondary electrons with low energy are emitted from the flight tube wall. Since the secondary electrons have low speed, they reach the MCP after the photoelectrons are directly captured by the MCP from the sample. The secondary electrons generated from the flight tube wall are also released with wide angular distribution, and the fraction of secondary electrons that arrive at MCP is high if they come from the flight tube wall close to the MCP, and is low as the distance increases. The arrival is delayed as the distance is further away from the MCP. This is the origin of the component that gradually decays after the peak. We made such an inference.

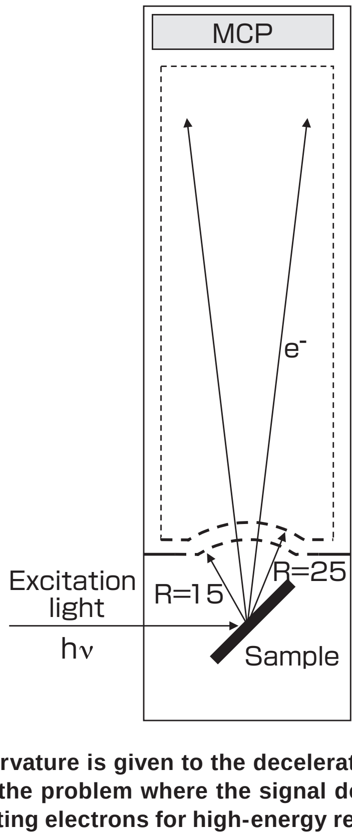


Fig. 7 Curvature is given to the decelerating electrodes to solve the problem where the signal decreases when decelerating electrons for high-energy resolution[23]

From the above consideration, we devised a way to prevent the secondary electrons from the flight tube wall to arrive at the MCP, as shown in Fig. 8.[24] Several collars were set near the exit of the flight tube near the detector, so the photoelectrons released from the sample were not blocked but the secondary electrons from the flight tube wall could not reach the MCP. The second idea was to use a mesh as the flight tube wall rather than a plate. By increasing the opening ratio of the mesh, the number of secondary electrons generated from the wall mesh is greatly reduced. Since the mesh line has a curvature, most of the secondary electrons generated at the wall mesh would not go to the MCP. By these two ideas, the large flare after the peak was reduced and became negligibly small.

4.4 Selection of a laser

As a laser for production of LPP, a laser named Infinity from Coherent, Inc. is employed. Special feature of Infinity is the use of a stimulated Brillouin scattering (SBS) mirror.[Term 4] The SBS mirror enables good Gaussian beam profile with a pulse width of 3 ns, for a very wide range of pulse energy and a repetition rate up to 100-Hz. These are extremely important characteristic of a laser for scientific experiments, particularly for EUPS.

The Q-switch YAG laser used in Unit 1 is one of widely-used lasers. Since it was not equipped with an SBS mirror, the time and spatial waveform of the laser beam change when the repetition rate or pulse energy is changed and operation at constant output power is required. The pulse width was about 10 ns, and a pulse of 2.5-ns was obtained by devising electric pulses of the Q-switch, but small pulses were present on both sides and was not a single Gaussian. With the YAG laser in Unit 1, the pulse energy of about 100-mJ was necessary to obtain the same EUV intensity obtained by Infinity operated at 30-mJ. The production of contaminant substances from the target is nearly proportional to the pulse energy, and the speed of contamination of the Mylar film increases. From the point of safety, use of second harmonic of 532-nm wavelength is the same as for Unit 1.

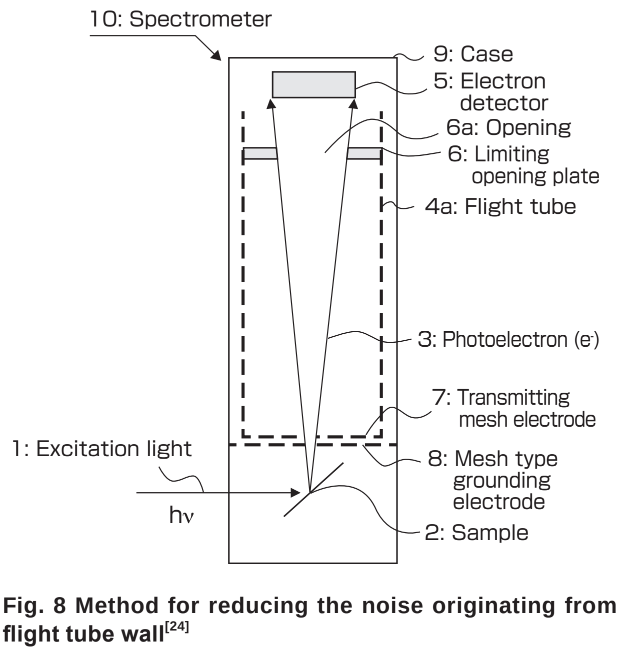


Fig. 8 Method for reducing the noise originating from flight tube wall[24]

4.5 Ultrahigh vacuum for the analysis chamber using a SiN filter of thickness of 100-nm

The vacuum separation of the LPP space and the elliptical mirror space is done using a Mylar film, but the degree of vacuum separation by the Mylar film is about six orders of magnitude, and the degree of vacuum in the elliptical mirror space remains at 10^{-4} Pa. The Mylar film deteriorates due to exposure to plasma, and the degree of vacuum separation decreases. The Mylar film is exchanged when the elliptical mirror space reaches 10^{-2} Pa.

To keep 10^{-7} Pa for the analysis chamber even when the elliptical mirror chamber reaches 10^{-2} Pa, the elliptical mirror space and the analysis chamber are vacuum separated by using a SiN film. Since the absorption coefficient of SiN film at 4.86-nm is large, to reduce the intensity drop of EUV light by the SiN filter, the SiN film of thickness of 100-nm or 50-nm is employed. The transmission at 4.86-nm are 17 % for a 3-μm Mylar and 48 % for a 100-nm SiN. Since the reflectivity of a NiC multilayer film of the elliptical mirror is 25 %, the total transmission is calculated as 2 %.

4.6 MCP and oscilloscope

In TOF energy analysis, the time resolution limits the energy resolution. Therefore, high time resolution is required for the MCP that detects the electrons and the oscilloscope that records the electron current. Products with about 1-ns time resolution were selected for the two components. To obtain large signals, it is necessary to have a large solid angle for electron capture, and MCP with a diameter of 40-mm is selected. To the author's knowledge, it was the largest commercially available product.

Since the time interval of detected electrons is long in an ordinary photoelectron spectroscopy, counting detectors with several μs dead time can be used. In EUPS, however, dozens of photoelectrons arrive in 200-ns and several tens of thousands of secondary electrons in 3-μs time, so, conventional counting detectors cannot be used. In EUPS, electrons are detected as an electric current recorded by using an oscilloscope. To reduce memory size, an average of signals of several tens to several hundred shots is calculated in the oscilloscope.

4.7 Event counting mode

Recently, we have more cases to measure very small photoelectron signals. The major problem we encountered when measuring very small signals was that good spectral waveforms were not obtained even by increasing the number of accumulating shots.

We thought the cause was the electric noise in the signal systems such as in the cables and the oscilloscope interior. Since the amplification of the MCP detector is very large, one electron signal is larger than the electric noise. However, the probability of arrival of electrons at a specific time in many shots is extremely low. On the other hand, the noise appears at the same time position in every shot. Therefore, when the signals of multiple shots are averaged, the electron signal is buried in the noise. This problem can be solved by recording TOF signals of all shots without averaging, and removing the noise from each shot signal.

The oscilloscope we selected has the sequential mode. In this mode, all signals of 1-μs duration with the 0.1-ns sampling interval can be recorded at 10–50 Hz repetition rate. Recently, the small signals are often recorded in the sequential mode. However, it is limited to a special purpose, because the data volume in one measurement is enormous, and consumes a large capacity of the data server.

A signal above a certain level is set as one event. Event counting is expected to remove all electric noise. By adopting this counting mode, it has become possible to observe ultra-weak structures as presented in Subchapter 5.7.

5 Pioneering of new photoelectron spectroscopy method using EUPS—Users' demand advanced the device

EUPS is the only one device in the world. But we were aware that it will not be widely used by people simply because it is the only one in the world. We were aware that general users would not use this device unless we became the user and demonstrated findings observable only by EUPS. Therefore, after the system technology was also completed, we embarked on using the EUPS to observe various samples for the following 10 years.

However, we did not imagine that we ourselves had not rightly evaluated the EUPS that we created. The potential of EUPS was brought forth in the process of our contacting many users, the users contacting us, and solving many problems that the users brought to us to solve. We can say that EUPS was advanced by the users. New analysis methods were born from the users' demands, and the system was improved. This relationship is shown in Fig. 9.

EUPS is capable of the following analyses that are impossible or difficult using the conventional photoelectron spectroscopy[25]:

1. Analysis of the electronic states of the topmost atomic layer
2. Evaluation of band bending in semiconductors
3. Analysis of insulating films without charge neutralization
4. Evaluation of cleanliness of nanoparticle metal surfaces
5. Evaluation of electric conductivity of nanoparticle surfaces
6. Evaluation of the inclination angle of electron clouds

5.1 Observation of the topmost atomic layer

When we initially started the development of the EUPS technology, many people commented that a wavelength of 4.86-nm is too long, and that the fact that 1s electron of carbon and oxygen and deep inner shell electrons of various elements cannot be excited was a serious deficiency as photoelectron spectroscopy. We replied that inner shells other than C, N, and F could be seen with 255-eV photon energy. But at that time, we did not realize that 4.86-nm was indeed the best wavelength before we started analyses of various samples.

The first sample observed by EUPS was a Si wafer. Only Si2p of the natural oxide layer on the surface was observed and Si2p of the substrate was not observed. The Si2p of the substrate was observed only after hydrofluoric acid treatment and Si2p of the oxide did not disappear easily. There were comments that EUPS observes only contaminations which we do not want to see. However, this was an ugly duckling. The very large signals of contaminations that normally could not be observed in conventional spectroscopy turned out to be extremely useful in surface analysis.

Figure 10 shows an example of contamination analysis conducted by researchers of Panasonic Corporation.[26]

To improve the performance of the insulating protection film on electrodes in a plasma display, Panasonic researchers were trying to improve the manufacturing process by reducing contamination. Previously, they evaluated the contamination of the insulation protection film by XPS, but the signal originating from contamination was very small. At an exhibition for analyzing devices, they knew EUPS, came to us and confirmed the sensitivity of EUPS by evaluating a sample. They conducted a joint research project with us. Figure 10 is the comparison when seeing the same sample using XPS and EUPS. Clearly EUPS has higher sensitivity. They evaluated contamination of the insulating film produced in various processes. We received a report that the actual manufacturing processes were improved based on the results of EUPS analysis.

The EUPS spectrum of a sample supplied by an AIST

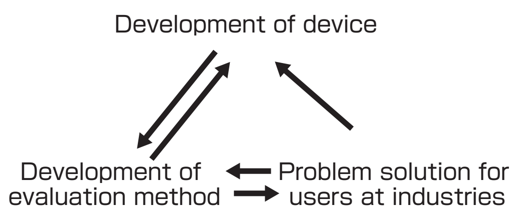


Fig. 9 Process of EUPS advancement

researcher is shown in Fig. 11. The Si2p signal of the substrate can be observed by removing the natural oxide film on the surface of a Si wafer by hydrofluoric acid treatment. In a sample with SiO_2 of thickness of 2.1 nm, substrate signal was only 10 % of that in the cleaned sample, and was hardly seen through a SiO_2 film of 4.9-nm thickness. This tells that about 0.5-nm from the surface is observed in EUPS. On the other hand, we know XPS observes an average of several nm depth from the fact that the substrate signal is greater for a SiO_2 film of 2-nm thickness.

The difference in depth resolution of XPS and EUPS is due to the difference in kinetic energy of observed photoelectrons. The “escape depth,” which is defined as a depth that the electrons escape into the vacuum without losing energy in materials, has very small dependence on materials and it is smallest at the kinetic energy of several tens to a hundred eV, and it is about 0.5-nm.[27] When observed by XPS, the kinetic energy of a Si2p electron is larger than 1-keV, and the escape depth is several nm. Figure 12 shows the escape depth observed by EUPS.[28] When the kinetic energy of the electrons decreases, the escape depth increases rapidly in insulators.

At the time when we invented the principle, we had no knowledge that the photon energy at around 200-eV used in EUPS was optimal for obtaining information of the topmost atomic layer.

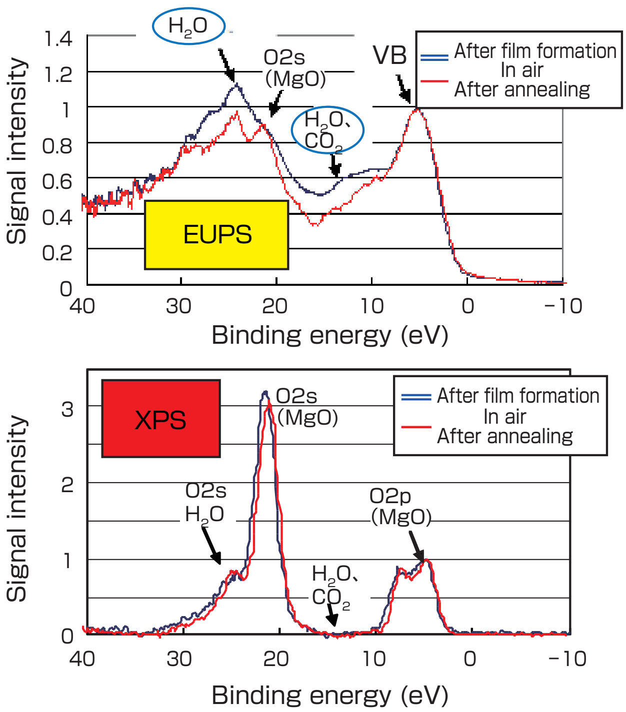


Fig. 10 Comparison of the sensitivity of EUPS and XPS in the contamination analysis for improving production process of insulator film protecting electrode in a plasma display[26]

5.2 Observation of band bending in semiconductors

The Si wafer was selected as the first sample of photoelectron spectroscopy using EUPS, to see whether the n-type and p-type could be distinguished. It was found that the peak positions of Si2p differed in n- and p-types but they did not agree with the difference of the Fermi level of different doping.Term 5 Also, we noticed that the peak positions changed

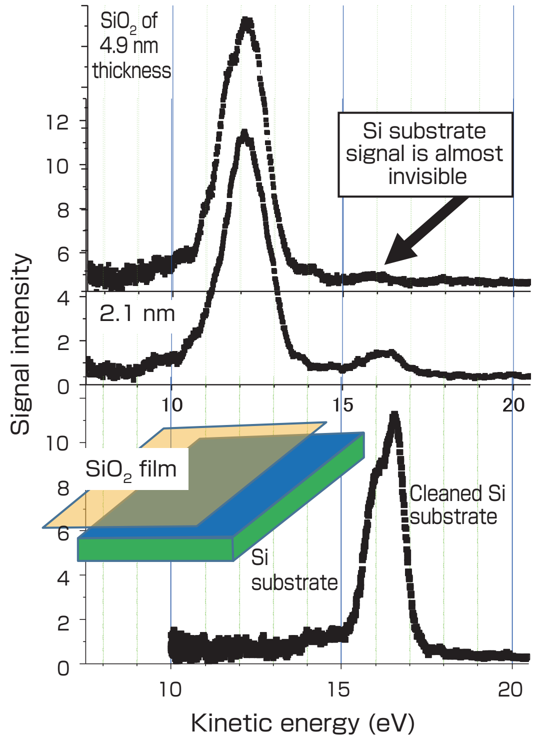


Fig. 11 Depth resolution of EUPS was evaluated to be about 0.5 nm, from the size of the background signal seen through a SiO_2 film of thickness of 2.1 nm

Single atom layer is about 0.3 nm, and information for the topmost atom layer can be obtained.

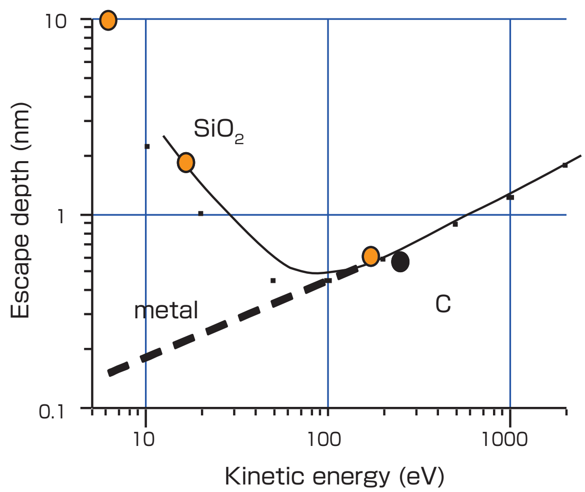


Fig. 12 Kinetic energy dependence of escape depth of electrons was investigated by EUPS[28]

Material dependence is large at the energy of 100-eV or less, and there is no “universal” curve.

when the irradiation intensities were changed. It took a few years for us to understand that the observed phenomenon was related with band bending.Term 6 This phenomenon occurred by the flattening of the bent band. With other sources bent band cannot be flattened, but it is possible with EUPS where ultrahigh density pulse excitation is used.

When an HfO_2 film with thickness of 12-nm formed on a Si wafer was measured by EUPS, the position of the Hf4f peak was dependent on the excitation intensity of EUV. When the measurement at strong excitation was continued, the peak position shifted to the low kinetic energy side. After the shift was saturated and the measurement was taken at different intensity, the intensity dependent peak shift increased further.[29]

When positive charge is captured in an insulating film, the potential of the electrons on the Si surface lowers because of the electric field generated by holes, and the peak of the photoelectron spectrum shifts to the low kinetic energy side. The captured positive charge increases for larger EUV irradiation, and then, the charge shift increases. When all the charge trapping centers are filled with the charges, the charge shift stops.

When the sample is photo-excited, the electron-hole pair is formed in the sample. Due to the electric field created by the charges in the insulation film, the electrons flow to the insulation film/Si interface while the holes flow to the Si interior. The electric field created by the separated electrons and holes offsets the electric field created by the charges captured in the insulating film on the surface. If a sufficiently large number of electron-hole pairs is created by increasing the excitation intensity, it is possible to completely cancel out the electric field created by the charges in the insulating film. That is, the bent band can be totally flattened.

When the surface density of the charges captured in the insulation film is set as $10^{12}/cm^2$, the electron-hole pair of a similar density or higher is needed for a flat band. Assuming the lifetime of an electron-hole pair as 10-ns, it is necessary to create an electron-hole pair at $10^{20}/(cm^2\ sec)$. However, the most powerful light emission facility in Japan, that is the hard X-ray undulator of SPring-8 in Himeji, has a photon flux of only 2×10^{16} photons/(cm^2 sec), and cannot affect band bending to a detectable level. On the other hand, in EUPS, the photon density on a sample per pulse is estimated to be $3 \times 10^{12}/cm^2$, and if 10 or more electron-hole pairs are produced for one 255-eV photon, it is possible to flatten the band bending produced by a charge of about $3 \times 10^{13}/cm^2$.

Evaluation of band bending is an important analysis characterizing semiconductors, and it is possible with an ultrahigh intensity pulse source as explained above. We could never have thought of this when we invented the EUPS.

5.3 Insulating film is not charged

In XPS, the electrostatic charge in the insulating material is a major problem. We knew that it was almost mandatory to install a neutralizer gun to neutralize the charge in photoelectron spectroscopy, but a neutralizer is not installed in the EUPS Unit 2. As the electrostatic charge in the insulating material was our concern, we studied SiO_2 ultrathin films and found they were not highly charged.

When a thermal oxide film of thickness of 100-nm was observed, the Si2p shifted with time, but it saturated with a shift of about 3-eV. It was then fully charged by irradiating the ion beam with a very small current. When this charged sample was EUPS measured, the peak shift was about 25-eV initially but peak shift decreased rapidly and the decrease of the shift stopped at a shift of about 10-eV. Next, after heating the sample to nearly 1000 ºC by using infrared rays, EUPS measurements were done and it was found that the charge had almost entirely disappeared. As the EUPS measurement was continued the charge shift was noticed, but it was only 0.1-eV.[30]

Textbooks of photoelectron spectroscopy explain that an insulating film is charged by photoexcitation because holes left after electrons are released into the vacuum do not flow through the insulating film to the substrate metal. We know this explanation is not correct from the result seen in Fig. 13. The result in Fig. 13 can be explained as follows.

Since a charge shift of 25-eV was observed with ion irradiation, we know that the SiO_2 used in the experiment had a high dielectric strength (>4 MV/cm). Since the charge shift decreased by EUPS measurement, it can be understood that EUV irradiation decreased the dielectric strength of SiO_2. It can be explained that the electron-hole pairs are created by EUV irradiation, this gives conductivity to the insulating film,

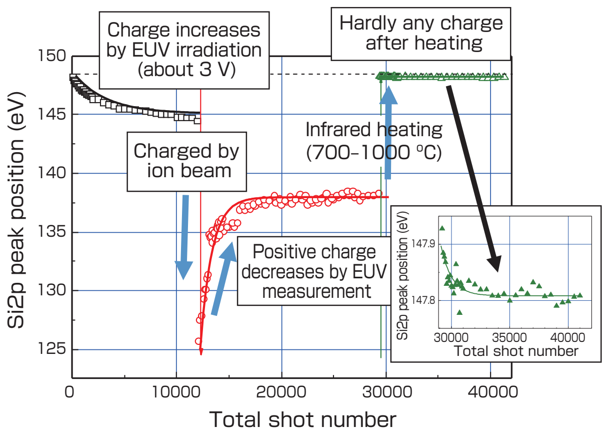


Fig. 13 Experiment that broke the “common sense” of photoelectron spectroscopy world that the thin insulator becomes charged because electrons go out of a sample
The amount of shift for SiO_2 of thickness of 100 nm charged positively with ion gun decreased by EUV measurement. This is thought to occur as the sample gained photoconductivity through EUV irradiation.

and the stored charges are released. It is thought that a similar phenomenon occurs in the XPS and the electron microscope, but we believe we are the first to make the argument that the insulating film becomes conductive during actinic excitation.

From fig.13, we can say that the charge shift of an insulating film is determined by the number of charge traps from the following three reasons: first, the large charge shifts generated by ion irradiation decreased rapidly by EUV irradiation but stopped at about 10-eV shift; second, the charge shift disappeared completely by infrared heating and annealing; and third, while the charge shift by EUV irradiation was 3-eV initially, it was only 0.1-eV after high-temperature annealing. The number of charge traps can be said to be as follows: after ion irradiation > initial stage > after high-temperature annealing.

We can say that the size of the charge shift is very important information of an insulator thin film, that is, the number of charge traps, and it should not be lost by neutralization.

5.4 Evaluation of electric conductivity from the charge shift

The insulating material with thickness over 1-μm such as a glass plate does not allow EUV light to pass through, therefore, will not become conductive with EUV irradiation. A thick insulator is charged as the electrons are released in the vacuum by EUV irradiation.

One user once asked us to analyze catalysts. When powder catalysts were measured at room temperature, there were absolutely no spectrum structure due to a very large charge shift, and the spectrum structure gradually appeared when the temperature was increased. When the temperature was increased further, the peak position shifted to the high kinetic energy side. This told us that the charge shift was smaller for higher temperature. The temperature dependence of the charge shift was different for different catalysts. The results are shown in Fig. 14.

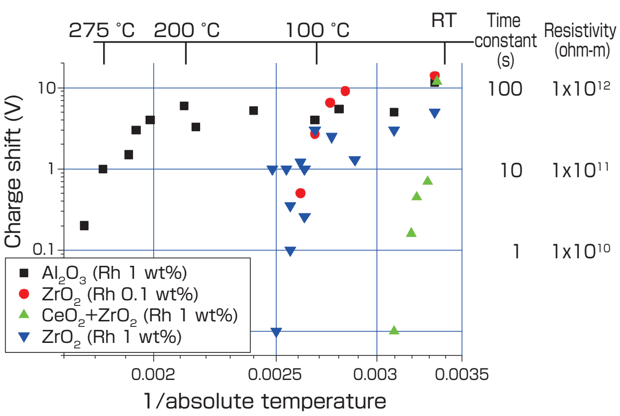


Fig. 14 Correlation was seen in the catalyst activity and the electric conductivity estimated from charge shift

The charge shift can be related to electric conductivity.

When the sample with thickness d and a surface area S is replaced with an electric circuit with capacitance C and leak resistance R, the charge Q on the surface decreases with time constant $\tau = CR$. Since C and R can be expressed by $C = \varepsilon S/d$ and $R = \rho d/S$, using electric conductivity ρ and permittivity ε, it is $\tau = CR = \varepsilon\rho$. That is, time constant τ of discharge is not dependent on the shape of the sample (S and d). The permittivity is only several times different for different materials, and it does not differ greatly in the same material at different temperatures. On the other hand, conductivity changes by several orders of magnitude. Therefore, the discharge time constant gives electric conductivity.

In photoelectron spectroscopy, the peak shift occurs (Fig. 15) by the positive charge remaining in a sample after electrons are released into the vacuum. The positive charge is accumulated when the excitation continues. But accumulated charges leak with time constant τ. Then, the amount of accumulated charge becomes saturated with time constant τ. If the size of the charge shift in one shot is set as V_0 and the pulse interval as ΔT, the shift V after n shots is given by

$$V = V_0(1-\exp(-\Delta T/\Delta)^n/(1-\exp(-\Delta T/\tau))$$

and the saturation value ($\fallingdotseq V_0(\tau/\Delta T)$) is proportional to the time constant τ. The size of the charge shift V_0 in one shot is dependent on the pulse energy and C that is determined by the shape of the sample. The time constant is given from the charge shift when calibration is done. Then, when the permittivity is known, the absolute value of the electric conductivity is given.

This method has the advantage that it can be applied to materials with all sorts of shapes including fine particles. It is now used as a method in the research of correlation between catalyst activity and conductivity of powder catalysts.[31]

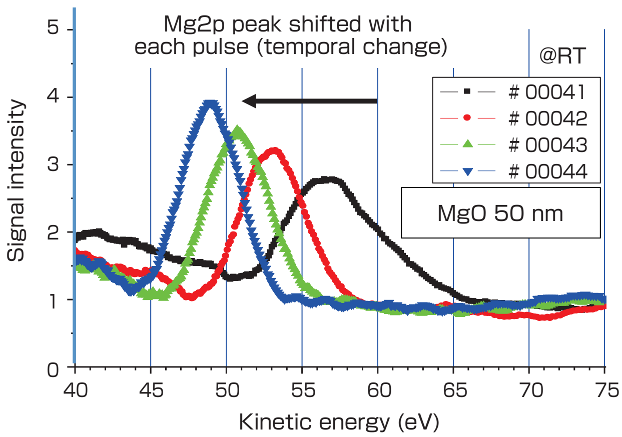


Fig. 15 Mg2p peak of MgO of thickness 50-nm shifted with every measurement
Electric conductivity is calculated from the time constant of shift.

5.5 Measurement of secondary electrons started by request from users

The measurement of secondary electrons was started by the request from the researchers of Panasonic Corporation who conducted the analysis of protection insulating film of electrodes in a plasma display using EUPS. They wanted to see the change of the vacuum level of the film by contamination. To answer their request, we improved the structures of the manipulator and the sample holder of EUPS so that we could control the potential of the sample.

5.5.1 Evaluation of the vacuum level (work function)

Primary electrons ejected out of atoms after gaining energy from photons collide with surrounding atoms, kick out new electrons (secondary electrons) from these atoms, and slightly lose energy. The primary electrons continue to collide with the surrounding atoms, kick out more secondary electrons, and continue to lose energy. Likewise, the secondary electrons collide with surrounding atoms and produce new secondary electrons. A large number of secondary electrons are produced like an avalanche, and the sea of secondary electrons is formed in the sample. The secondary electrons with kinetic energy surpassing the vacuum level are detected. Therefore, the vacuum level is obtained from the cutoff position of the energy spectrum of the secondary electrons.

An example of secondary electron spectra for TaN and W films with thickness of 10-nm formed on a Si wafer are shown in Fig. 16. In the case of TaN film, there is a sharp edge at 2.6-eV, and in the case of W, the sharp edge is at 3.5-eV. From these, it is found that the vacuum level of W was 0.9-eV higher than the vacuum level of TaN, and that the presence or absence of SiO_2 between the substrates did not change the vacuum level.[32]

Trajectories of electrons with small kinetic energy are easily affected by a very small floating electric field or magnetic field in the space around a sample, and the fraction arriving at the detector can be affected. To avoid this, the measurement of secondary electrons is performed by applying a negative potential bias to the sample holder to accelerate the electrons. In Fig. 16, bias of −2 V was applied to the sample holder in the measurement.

In the measurement of secondary electron spectra, it is often necessary to measure a wide range of several tens of eV with a very high energy resolution of several tens of meV. The TOF method is most powerful for such ultra-multi-channel measurements.

5.5.2 Evaluation of ultra-cleanliness of metal surface and evaluation of carrier density from secondary electron intensity

The above vacuum level observation was a response to the users' demand, but we realized that the intensity of the secondary electron was different for various samples (Fig. 17). Signals were small in metals and large in insulators. We interpreted this as meaning that the difference in the signal intensity of secondary electrons was due to the difference in the escape depth of low energy electrons.

In the case of insulating samples, new excitation of electrons cannot occur when the energy of the electrons becomes smaller than the band gap energy, and the electrons can reach the sample surface without losing any more energy and can escape into the vacuum.

On the other hand, for metals in which the Fermi level is in the continuous band, the electrons can be excited no matter how low the energy of the secondary electrons is, and electrons keep losing energy. Only the secondary electron that was created in the atom very close to the sample surface can escape to the vacuum. The escape depth of low energy electrons in metal is very small, and the secondary electron

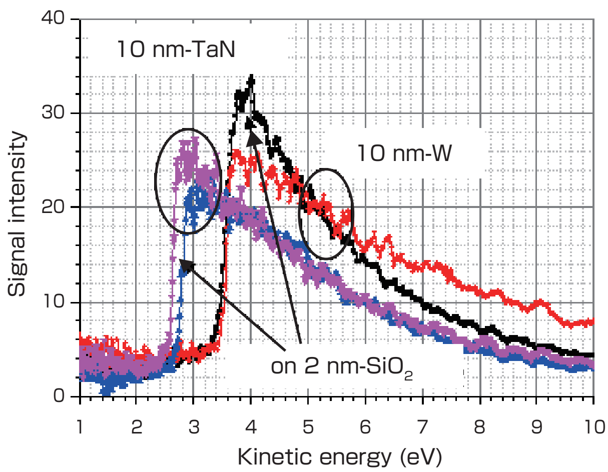


Fig. 16 Secondary electron spectra of TaN and W of thickness 10 nm

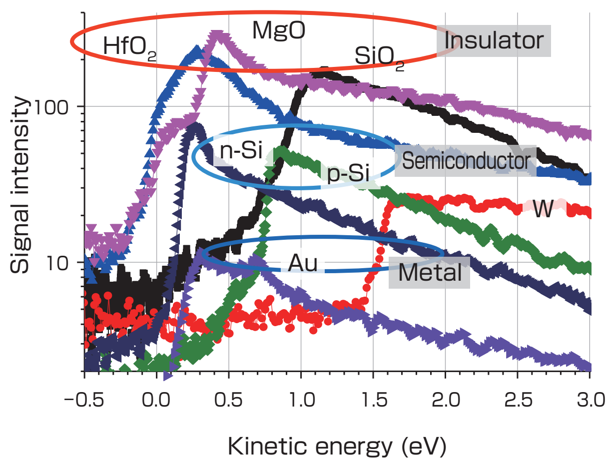


Fig. 17 Intensities of the secondary electron signals are in order of: insulator > semiconductor > metal

intensity of metal is small.

By knowing this principle, the metallic property of an ultrathin film or nm size particles can be evaluated, as shown in Fig. 18. Samples were TaN films with a different thickness formed on a W film of 100-nm thickness. Metal W has a weak secondary electron intensity and the effect of the transmission electrons from the underlying W layer is small. The secondary electron signal intensity was the smallest for a 10-nm TaN film, increased as the thickness decreased, and it was four times the intensity of 10-nm for TaN thickness of 1-nm. This result showed that the metallic property weakened as the film thickness decreased.[33]

This analysis method allowed a qualitative discussion of carrier density of organic semiconductor laser materials. By doping, the carrier density of organic semiconductor materials for optical devices is expected to increase, the resistance reduces, and enables injected current density to a higher level. The secondary electron spectrum of n-type AC5-CF_3 with 2 % Cs_2CO_3 as a dopant showed 0.9-eV lower cutoff position, and the signal intensity was half of that of a non-doped sample. This showed that doping lifts the Fermi level up by 0.9-eV against the vacuum level, and increases the carrier density. Incidentally, the secondary electron intensity of AC5 that had 1-eV greater work function compared to AC5-CF_3 was twice that of AC5-CF_3.[34][35]

5.6 Correlation between the inclination angle of electron clouds and catalyst activities

Emission from LPP is non-polarized, but in EUPS Unit 2, the measurement is the same as the polarized excitation owing to its configuration. EUV light irradiates sample horizontally and the electrons are detected at the vertical direction, and therefore, the electrons shaken in the electric field that oscillates horizontally (s polarization) are not detected, and only the electrons vertically shaken by p polarization are detected. Therefore, the angular distribution of the electron cloud can be observed from the angular dependence of the spectrum intensity by changing the angle of the sample (Fig. 19).

The angular distribution of the electron cloud was detected for the first time in a Si wafer. The direction of Si3p differed for different crystal orientation.

We were greatly surprised when we observed the angular dependence in the photoelectron spectrum of a powder sample that will be mentioned later. The direction of the electron cloud is usually considered to be determined by the crystal plane. If that is the case, the angular dependence should disappear in a powder sample in which crystal planes of individual particles are randomly oriented. Yet, there was angular dependence in powder samples. This means that the direction of the electron cloud is determined by the exterior form rather than the crystal plane, and this is extremely interesting. This phenomenon occurs only for the topmost atoms, and cannot be observed by XPS which gives information averaged over several nm depth.

ZrO_2 powder is employed as a catalyst for cleaning automobile exhaust gas. We measured the Zr3d photoelectron peaks of seven types of ZrO_2 powder samples with different additive precious metals. We found that the angles at the maximum intensity were different for different catalysts, and extremely good correlation was seen between the angle and the catalyst activity of water gas shift reaction of $CO + H_2O \leftrightarrows CO_2 + H_2$.[36] It is probably the first time that the correlation between the efficiency of dissolution of gas molecules and the inclination angle of the electron clouds of catalyst atoms was found. It is expected to provide important information for the understanding of the catalyst mechanism. This analysis is unique to EUPS which sees the topmost atomic layer.

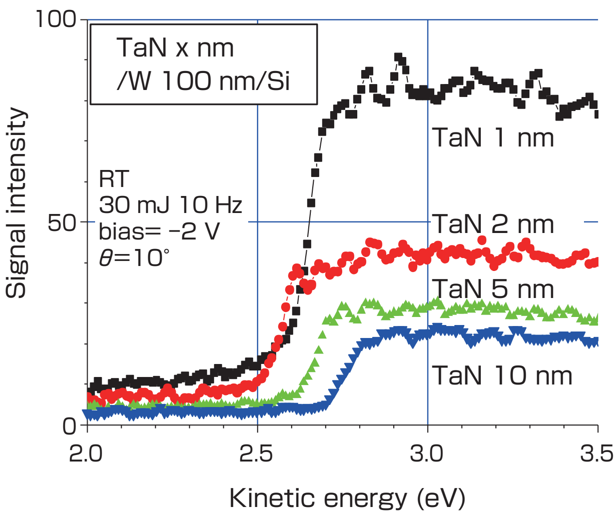


Fig. 18 Secondary electron spectrum of the TaN ultrathin film[32]

Since the signal is larger for thinner TaN, it can be said that the TaN film has lower electron density as it gets thinner.

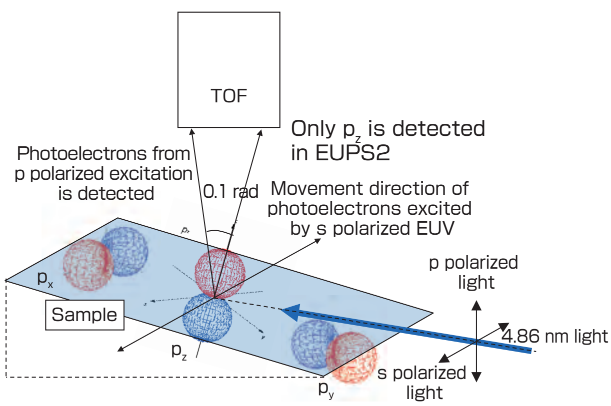


Fig. 19 In EUPS Unit 2, the electrons excited by p polarization of horizontal irradiation are detected by the detector installed vertically

5.7 Observation of the conduction band and the intra-gap states[Term 7]

Figure 20 shows the photoelectron spectrum near the Fermi level for four samples. As mentioned in Subchapter 4.6, we conducted improvements recently to enable obtaining event count data using the oscilloscope in the sequential mode. Therefore, it became possible to observe an extremely weak structure. Thus, it became possible to remove the effect of continuous spectra in the light source. By looking at Fig. 4 carefully, readers will notice that the 4.86-nm line emission lies on a weak continuous spectrum. The contribution of the continuum component was ignored until now because it was very small and was not easy to estimate. The contribution can be estimated by observing the sharp structure of the Fermi edge. Figure 20 is the spectrum after removing the effect of the continuous spectra. The signal intensity of four samples were different, and spectra were normalized at the peak.

As seen in the left figure of Fig. 20, sharp Fermi edges can be seen in the d band for Pt and Pd. As seen in the graph on the right that is the expanded vertical axis of the graph on the left, signals of 1–2 % intensity of the peak were seen at the energy above the Fermi level.

In the SiO_2 with thickness of 100-nm, there was a flat structure of about 2 % of the peak in the energy region between the Fermi level and the valence band top that dropped sharply at 5-eV below the Fermi level which lies at the 22-eV position of the horizontal axis. In the case of HfO_2 formed on 100-nm SiO_2, the signal near the Fermi level was large to the extent that the valence band top was not clear.

Since the size and spectral form were different for samples, the observed conduction band electron signal is considered real, but since the signal is weak, further research is necessary to be confident that it is real.

The lifetime of conduction band electrons in bulk metal is reported to be <1 fs–100 fs.[37] However, the lifetime of the surface states can be fairly long. In EUPS in which a sample is excited by ultrahigh density excitation of 3-ns pulse, it is no mystery if the transient states is observed. It is thought that the conduction band states and the intra-gap states at the surface-interface have large effect on properties of materials, and their analyses are important.

6 Summary

The characteristics of an LPP source are a narrow bandwidth line emission, a short pulse, and high brilliance. EUPS is an application taking full advantage of LPP. This is probably the first commercially viable technology of LPP application. The basic important synthesis technology is to obtain narrowband light without using a spectrometer and conducting the electron energy analysis by the TOF method. However, development of many elemental technologies was necessary before materialization of the basic idea. In this paper, we explained the process of synthesis to make the spectroscopy system as a working apparatus.

The shielding of contaminants, the greatest issue of LPP source applications, was solved using a BN rod as an LPP production target and an inexpensive Mylar film as the filter to block contamination. The frequent exchange of the shielding filter was reduced greatly by the introduction of an elliptical mirror. By not inserting a diffraction grating, the time averaged flux on a sample of EUPS is comparable to that using synchrotron radiation. The TOF method was employed as the electron energy analysis by taking advantage of the pulse nature of an LPP source. Problems of TOF not imagined when the basic principle was born arose when the actual measurements were conducted. The problems were solved by improving the structure of the flight tube.

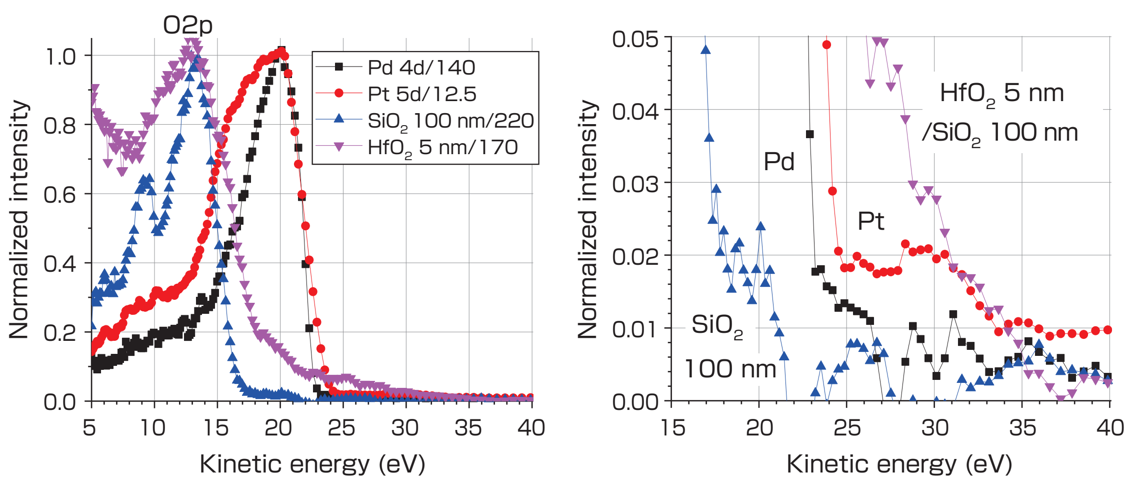


Fig. 20 Photoelectron spectra near the Fermi level for Pd, Pt, SiO_2, and HfO_2

Normalized by the peak signal. In the right graph that is the enlarged portion of the vertical axis of the left graph, the conduction band signals with intensity around 1 % of the peak are observed.

When the actual measurements of samples began, unexpected phenomena occurred one after another. By understanding them one by one, new analysis methods that could not be done with the conventional photoelectron spectroscopy were born. These include the detection of less than one monolayer contamination on material surface, detection of defects, measurement of insulator thin films without charge neutralization, evaluation of electric conductivity using the charge shift, observation of inclination angle of the electron cloud of the topmost atom, evaluation of electron density near the Fermi level from the secondary electron signal intensity, evaluation of the ultra-cleanliness of the surface of nm -size metal particles, and others.

The EUPS system was completed when the authors belonged to the Advanced Semiconductor Research Center, and since we know many researchers studying semiconductors, measurements were done mainly for semiconductor samples at the time. Due to the appearance of enthusiastic external users, the majority of the measurements are now for samples other than semiconductors. Since EUPS provides various findings for the topmost atom, it is expected to be most productive in the analysis of the catalytic phenomenon for which reactions occur at the topmost layer of atoms. In fields other than catalysts, if it is known that the topmost atom plays a critical role, EUPS will also be effective in that field. To expand to such new fields, participation of diverse users is necessary. Many of the new analysis methods unique to EUPS were born from the analyses of samples brought in by users. The author who started EUPS development without experience in photoelectron spectroscopy could not imagine the new ways of applying photoelectron spectroscopy and the wonderful potential of EUPS was made clear by enthusiastic users. I think “users’ demand advances the device” is a principle that is widely applicable.

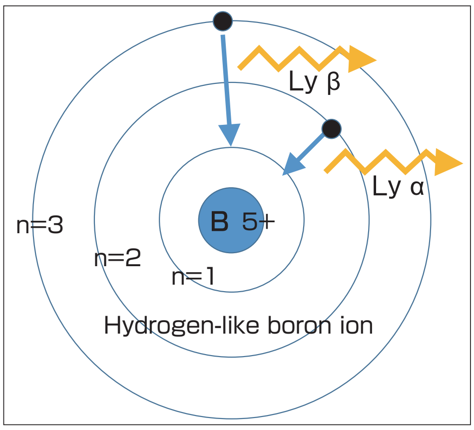
In terms of user-friendliness, EUPS does not compare with the XPS or UPS that are widely employed photoelectron spectroscopy. Drastic improvement of user-friendliness cannot be resolved by the author alone, and wide diffusion of EUPS analysis is necessary upon product realization. However, I think it is the responsibility and obligation of the inventor to bring EUPS to product realization.

Acknowledgements

The development of EUPS would not have started without the words, “Can we use LPP in industrial applications?” spoken by Dr. Yoichi Kimura, Manager of Extreme Technology Division, when the author was, at the time, conducting the research of the X-ray microscopy. Mr. Hiroyuki Kondo, who came to do research at the Electrotechnical Lab for a very long term from Nikon Corporation, built EUPS that was merely an idea on paper to a real device (Unit 1) with which we obtained the Si2p photoelectron spectrum. Therefore, the EUPS research would not have taken off without Dr. Hiroshi Nagata and Mr. Kiyoshi Iizuka of Nikon Corporation, as they dispatched Mr. Kondo to the Electrotechnical Lab. Without the strong support of Dr. Hideaki Shimizu, the joint researcher of X-ray microscopy research, we would not have obtained enough results to apply to the Special Coordination Funds for Promoting Science and Technology that gave us a large research budget to go forth on the device development. Without this, we could not have been selected by Project Leader Prof. Hirose to the MIRAI Project that started after AIST came to being, and the development of the current Unit 2 would not have happened. We have arrived at this place through the support of many, many people including the post docs, sample suppliers, and users. EUPS continues to grow, and there are people who support us. We are thankful to the generosity and kindness of all the people involved.

Footnotes

Footnote 1. Mechanism of X-ray production from LPP: When free electrons are captured by fully ionized boron ion, and when the main quantum number transition from n=2 to n=1 occurs, 4.86-nm Lyman α ray is emitted. In EUPS, the laser focusing density is adjusted to get an appropriate temperature at which the fully ionized boron ions will be present in sufficient amount, and at which the free electrons recombine at sufficiently high speed. The emission intensity is proportional to the product of ion density and electron density, that is, the square of plasma density, and the emission intensity increases as the plasma density is higher. On the other hand, if the density is too high, self-absorption increases, intensity is saturated, and the width of the line spectrum broadens. The limit intensity is the intensity of “blackbody radiation,” and at extremely high density, the spectral structures disappear and a spectral structure becomes flat. In EUPS, since the spectral width of excited EUV is determined by the emission spectrum of LPP, the plasma density should not be too



high. On the other hand, since strong emission is desired, some degree of density is wanted. The density of LPP depends on the wavelength of the excitation laser. The second harmonic of YAG laser with wavelength of 0.53-μm is an appropriate wavelength for producing narrow bandwidth of 4.86-nm light.

Footnote 2. Difference between LPPs for lithography and photoelectron spectroscopy: The requirements for light sources differ greatly for photoelectron spectroscopy and lithography. For the EUVL source, the requirements include high EUV power, low fluctuation of power, high conversion efficiency from lasers to EUV power, and being maintenance free for a long period such as one year. On the other hand, for the photoelectron spectroscopy source, narrow spectral width is the most important. Small power fluctuation, high conversion efficiency, and maintenance free characteristics are desirable, but they are not critical requirements. In EUPS, narrow bandwidth is a critical requirement to the LPP emission since a spectrometer is not used. For the EUVL source, wide emission that covers all widths (about 2 %) of the multilayer film reflection spectrum is needed to increase the usable power, but for EUPS, narrow bandwidth emission of about 0.1 % is required. In the TOF method, since the energy resolution is determined by time resolution, the emission time of the source is desired to be as short as possible, and it is 3-ns in the present EUPS. On the other hand, for the lithography source, the pulse width should be as wide as possible to have large power.[38] Pulse width of several tens ns is used. Therefore, all things are different for lithography and EUPS sources, including the laser selection, irradiation conditions, target materials, structures, and supply methods. For the target material, Sn is used in EUVL, while BN is used for EUPS. Rod target is used in EUPS while liquid droplets are used in EUVL. In EUPS, a YAG laser with pulse width of 3-ns, a wavelength of 0.53-μm, and repetition rate of 10 Hz is used, while for EUVL, a YAG laser with a wavelength of 1-μm or a CO_2 laser with a wavelength of 10.6-μm is used. EUPS uses single pulses, but for the EUVL source, use of prepulses is mandatory. Totally different LPP sources are used in these two applications, and the development of light source technology in one area does not benefit the other.

Footnote 3. Comparison of EUPS and XPS: While the XPS analysis is essential in today’s material development, taking a rather self-centered view, it is stuck in element analysis. An analysis that captures the attractive topmost phenomenon that delves into the essence of material functionality as described in this paper is not being done with XPS. However, since XPS can excite the deep inner shell and is highly effective for element analysis, EUPS will never replace XPS in that area. As the product realization technology for EUPS progresses, it is thought that the price of EUPS will become less expensive.

Terminologies

Term 1. Magnetic bottle: The arrangement of a magnetic force line in the form of a bottle. It is an idea that when a sample is placed at the mouth of the bottle with magnetic flux density of 1 tesla or more, and the electron is wound around the magnetic line and the line is adiabatically expanded, only the direction of motion is changed without changing the kinetic energy of the electron, and therefore the enlarged image of the electron can be obtained in the electron detector placed at the bottom position of the bottle. The reason we were unable to observe the expected performance might possibly be because the magnetic bottle used in Unit 3 did not have a sufficient performance level, and there is room for success in future development. However, to raise the magnetic flux density to 1 tesla or more, there is only 1 mm or less space available for the sample. Since we realized that this lack of space severely limited the samples that could be measured, we stopped the use of the magnetic bottle.

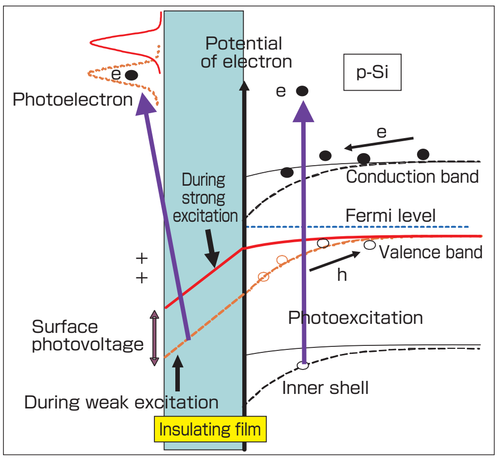
Term 2. Schwarzschild optics: To focus light onto minute diameters, it is necessary to decrease optical aberration, and near normal incidence is required. A Schwarzschild optics is a near normal incidence optical system that combines concave and convex mirrors, and submicron resolution can be achieved. However, it is not easy to obtain large vertical reflectivity in X-ray multilayer films, and the high normal incidence reflectivity of about 70 % has been obtained only for Mo/Si multilayer films. This is the reason 13.5 nm was selected in EUVL that uses more than ten multilayer mirrors.

Term 3. Microchannel plate (MCP): High-speed particle detector. About 2-kV of voltage is applied between both sides of a circular or rectangular glass plate where many microchannels of inner diameter of around 10-μm are bundled together. The secondary electrons produced at the MCP surface due to particle collision are led to the microchannels to achieve many orders of magnitude amplification. With a multiplication factor of 10^7 or more, the detection of a single electron becomes possible. It is characterized by high-speed response where the start-up time is 1-ns or less, and this is essential in TOF electron spectroscopy.

Term 4. Stimulated Brillouin scattering (SBS) mirror: When reflected wave having reversed phase with the incidence wave (phase conjugation) is generated, the diffraction-grating-like sound waves are produced in a substance due to the interference of the two waves. In stimulated Brillouin scattering that is the back scatter of the laser light by a sound wave, the wave front distortion of the incident wave that experiences as it passes through a distorted laser gain medium and the wave front distortion of the reflected waves are in antiphase. Therefore, the wave front distortion of the returning laser beam does not increase when the laser beam goes back and forth in the laser medium having a large optical distortion caused by strong excitation. Thus, good quality laser beam is obtained with a SBS mirror.

Term 5. Semiconductor doping: The Fermi level, which is the energy level at which the probability that electron exists is 50 %, is the center of the band gap in non-doped semiconductors. Impurities are introduced (doping) to adjust the position of the Fermi level to control the height of the Schottky barrier. When doping is done with impurities that supply electrons, the Fermi level approaches the bottom of the conduction band and an n-type semiconductor is formed, and when doping is done with impurities that receive electrons, a p-type semiconductor is formed.

Term 6. Band bending: Very often, the charge is captured in the interface, or the charge is captured in the insulating film that is sandwiched between the metal and the semiconductor, and the band position at the interface of the conduction band bottom of the semiconductor may be different from the position in the bulk. Then, the control of expected carrier transportation will be lost, and the device performance decreases. It is very important in device development to know the size of band bending and how to decrease unintended band bending. Since the flattening of the band is accomplished by strong excitation in EUPS, band bending can be evaluated from the difference of the positions of photoelectron spectra by changing excitation intensity.



Term 7. Intra-gap level: In crystals, energy gap is formed as the energy level separates into the valance band and the conduction band by periodicity. However, in surface interfaces, periodicity in the depth direction is lost, band boundaries become unclear, and electron levels occur in the energy position that corresponds to the energy gap of the bulk. Since this is thought to greatly affect the transportation property of the carrier, the evaluation of the position and the amount of intra-gap levels is important. Particularly, in catalysts where the reactions occur on the topmost atom, it is possible that the intra-gap level plays a definitive role in catalyst activity.

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on EUV source and the research on the carrier dynamics in the Au-TiO_2 nanoparticle system using the photoelectron microscope PEEM and femto-second laser. Continues the EUPS research as a Visiting Researcher at AIST. In this paper, he was in charge of the invention of EUPS, the development of technologies for practical device, and the pioneering of new applications for photoelectron spectroscopy.

Tomoaki ISHITSUKA

Completed the master's course at the Graduate School of Science and Engineering, Ibaraki University in 1992. Joined Hitachi Maxell, Ltd. in 1992. Technical staff at AIST in 2008. Since then, has continuously worked on EUPS measurement. Currently engages in the R&D for EUPS and other devices and the support of projects using such devices (including the Microstructural Characterization Platform Project). In this paper, he was in charge of the measurement of most of data presented in Chapter 5.

Discussions with Reviewers

1 Overall

Comment (Naoto Kobayashi, Waseda University)

This is a paper on the development of the photoelectron spectroscopy method for the practical use of laser-produced plasma (LPP) and its various applications, for which the authors have engaged passionately in R&D. It explains the detailed course in which the initial objective for the LPP source was set, how the new photoelectron spectroscopy was gradually established, and how the technologies were put to various advanced applications, and I think the paper is valuable from scientific and engineering perspectives. This was a typical "seeds-driven" or potential-driven research that utilized the characteristics and advantages of the technological potential of the LPP source and achieved practical use, and you describe that the voices of various users played an important role in exploring various applications. The unique technological synthesis method and process make this a paper appropriate for publication in *Synthesiology*.

Comment (Ken'ichi Fujii, AIST)

While the X-ray photoelectron spectroscopy (XPS) is used widely for the analysis of the surface and its contamination state, the authors looked at the pulse laser produced plasma as the source for the XPS and went on to develop the extreme ultraviolet excited photoelectron spectroscopy (EUPS) along with its effective applications. I think the paper that records this process is extremely valuable.

Answer (Toshihisa Tomie)

We received your useful comments on this paper after you read it carefully. I believe your comments are appropriate. I also made some improvements on the structure of the paper, as you proposed, like adding explanations of terminologies at the end. The subtitle you proposed is attractive, and I shall use it.

2 LPP source

Comment (Ken'ichi Fujii)

To make this technology be known widely by general engineers and researchers, I think you need to offer a more understandable explanation on the production principle of the EUV light source. You have some explanation in Subchapter 1.2 "History of the research of laser-produced plasma source," but I think the reader's understanding will deepen if you explain, along with figures, what is the excitation laser used in LPP, and by what mechanism extreme ultraviolet light is produced.

Answer (Toshihisa Tomie)

The selection of the laser is important, and I listed the following requirements in Subchapter 4.4 "Selection of a laser." That is, the pulse waveform needs to be fine Gaussian with pulse width of 3 ns using stimulated Brillouin scattering (SBS) mirrors, the spatial pattern ought to be TEM00 mode, the good beam pattern and the pulse waveform ought to be obtained independent of the pulse energy and the repetition rate up to the repetition rate of 100 Hz, and others. As you proposed, I added some footnotes at the end of the text.

3 Future prospect

Comment (Naoto Kobayashi)

It can be seen that the newly pioneered EUPS has provided new observation methods for various substance surfaces. Particularly, new findings never seen before have been obtained for the behavior of electrons on the topmost surface. To which science and technology fields do you expect this technology will be applied in the future?

Answer (Toshihisa Tomie)

I added the following expression in Chapter 6 "Summary."

"Since EUPS provides various findings pertaining to the topmost atom, we believe it will be most effective in analyzing the catalyst phenomenon where the reaction occurs at the topmost surface of atoms."

4 Commercial device

Comment (Naoto Kobayashi)

The technology was advanced through many users, and I feel that some manufacturers will be interested in making a commercial device. Was there any such plan? If the prospect for that is not good, what are the barriers that stand before commercialization?

Answer (Toshihisa Tomie)

Ten years ago, I visited many analysis companies but their responses were not very good. Recently, many users have asked how much would it cost to manufacture a device, and some have commented that 50 million yen isn't that expensive, but no actual project has taken off.

Since the Panasonic Corporation, who improved the manufacturing process by EUPS analysis, withdrew from the plasma display panel business, we can no longer ask them to advertise our work. However, if we gain more case studies where people have been able to advance the business because they used the EUPS analysis, stories will circulate, and I think the demand for product realization will be strengthened. Before talking about product realization, the greatest reason the product has not gained awareness is the lack of basic data published in the scientific journals, and for this, we are totally guilty. Recently, there are papers written by users, and I hope the knowledge of this technology will gradually increase. I hope the commercialization will become more specific. As I wrote in Chapter 6 "Summary," it is my responsibility to work on EUPS until product realization.

A revolutionary technical development to revitalize Japanese forestry

— A proposal for a portable tree felling manipulator to address specific properties of Japanese forestry —

Yuko SHIRAI

[Translation from *Synthesiology*, Vol.9, No.4, p.235–251 (2016)]

First, we present issues in Japanese forestry based on an explanation of the specific properties of Japanese forestry. Then, taking the revitalization of Japanese forestry as a goal, we present a scenario for the achievement of that goal and comment on the type of research that is needed for it. This includes descriptions of the positioning and role of the technical development currently undertaken by the authors. As a concrete example of machine development, we report on the details of a manipulator for cutting down trees, "TATSUMI," and the results of verification tests. The TATSUMI manipulator is a machine that is compact and lightweight enough to be carried by a single worker. This machine cuts down trees using mountable/dismountable chainsaws that are commonly available at forestry sites. We also discuss new design methods for machine development that were identified as suitable for the mountain forests of Japan.

Keywords **:** Japanese forestry, design methods, manipulator, tree felling operation, mobility

1 Introduction

We are developing two types of manipulators that are capable of felling standing trees, and a transport mechanisms that can travel and carry heavy loads over uneven terrain of the mountain forests. We are also working on a system to measure and visualize forest information,[1] and a system to automatically create a road network for forestry operation in mountain forests, from various geographical conditions.[2]

In this paper, first, we explain the specific properties of the Japanese forestry. Then, we set the revitalization of Japanese forestry as the goal of research, present the scenario, and select the research topics that are necessary for achieving the goal. For the issues of safety and productivity of forestry, we have proposed and are developing some novel machines. In this paper, the "portable tree-felling manipulator TATSUMI" that is capable of cutting down trees in mountain forests will be discussed as our case study.

Each work environment and each target tree in mountain forests are unique, and the differences in environment and individual trees are great. The methods for felling trees rely heavily on experience, and there has been no numerical evidence. It is difficult to develop machines that work in the Japanese mountain forests, using the conventional design methods such as the V model or the agile model. Therefore, we sought a new design development method. This design method will also be explained in this paper.

2 Specific properties of Japanese forestry

The specific properties of Japanese forestry will be explained (Fig. 1). Compared to the United States or Europe, one reason that makes Japanese forestry difficult is the Japanese natural environment. Forestry is done in the "forests" in Europe, while forestry is done in the "mountains" in Japan. This is a fundamental difference. In the following sections, the uniqueness of Japanese forestry will be spelled out one by one to extract the research topics.

2.1 Type, quantity, and location of the forest resources

In Japan, forest covers 67 % of the land, and the absolute quantity of surface area (2.5 million ha) and growing stock volume (4.9 billion m^3) are high. What should be particularly

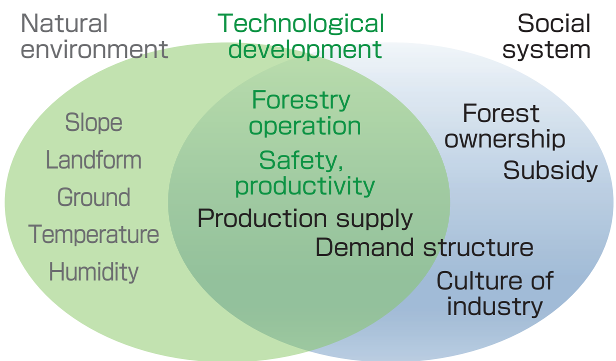

Fig. 1 Specific properties of Japanese forestry

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noted is that the growing stock per unit surface area is outstanding in Japan.[3] The growing stock volume moreover continues to increase. The increase is caused by artificial forests that were planted by humans. The amount a tree grows in one year is called the annual increment, and it is said that 70 % is allowable for harvest. About 70 % of the mean annual increment is harvested every year in Sweden, Finland, and Austria.[4][5] However, only about 20 % of the mean annual increment is harvested in Japan.[5][6] We should utilize the forest resources not only for forestry, but also for the preservation of the natural environment.

The above numbers are estimated values, and accurate understanding of where, what, and how much there is have not been obtained for this continuously growing forest resource. There are many cases where the owners and property boundaries are unknown which makes it even worse. These are also the points that complicate Japanese forestry.

The measurement of forest resources is mainly conducted for the purpose of grasping national resources and for the purpose of conducting forestry. It should also be pointed out that most of the measurements are done by hand.

2.2 Forest ownership

Europe and Japan take very contrasting stances on how the forest ownership is handled. The Japanese administration tends to work on protecting the ownership rights of personal property, while Europe has a system of calling owners to account along with the rights to ownership. Also, in the forest laws of Germany and Austria, the right to enter forests is given to all people.[7] In the survey done by the author in Germany, the burden on the owners that arises from other people entering privately-owned forests was estimated. One administrator replied that the owners sacrifice themselves for the sake of public good, therefore they are given subsidy money. On the other hand, the Japanese administration spends most of its time and money to find the owners to take care of the mountain forests.

2.3 Subsidy for forestry in Japan

The administrative investment, or subsidy, for building forestry roads and for forestation is 296.2 billion yen/year, and this surpasses the amount of lumber production of 214.3 billion yen/year (Fig. 2).[8][9] Of the forestry production amount, the production of mushrooms is 203.7 billion yen/year, and this is almost the same as the lumber production.[9]

In current Japanese forestry, many forestry businesses and unions engage in work as instructed by the administration to receive the subsidy money available at the moment, and the amount of subsidy is based on whatever the trend is at the time, and there are no definite rules. Innovation should originate from the sites. However, the current system does not provide an environment where people on site can engage in trial-and-error, originality or ingenuity.

2.4 Safety and productivity in forestry operation

2.4.1 Industrial accidents

In the 1970s, the accidents involving death and injuries in forestry surpassed 16,000 cases per year, and the number of deaths was nearly 250 people.[10] Comparing the occurrence of industrial accidents in forestry with other industries by "rate per 1,000 persons" that expresses the degree of occurrence of accidents, it is currently still the highest among all industries.

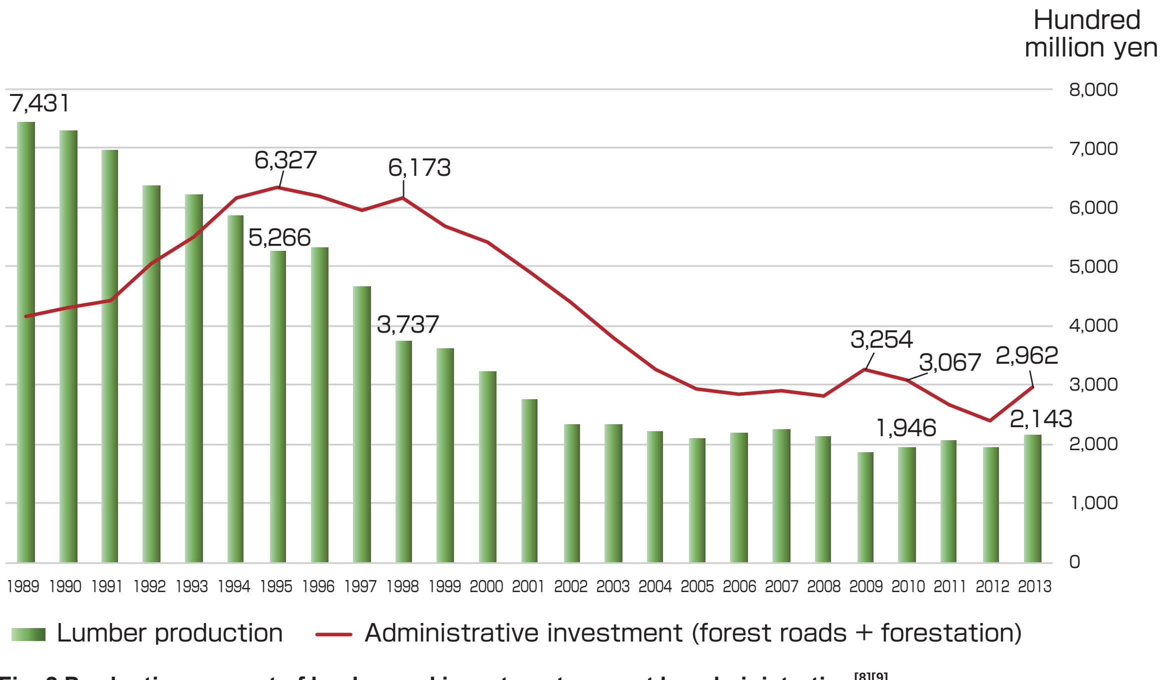

Fig. 2 Production amount of lumber and investment amount by administration[8][9]

While the average for all industries is 2.1, forestry stands out at 27.7 (2011).[11] Figure 3 shows the amount of industrial accidents in forestry. The serious accidents involving death and injuries were never less than 2,000 cases up to 2011, and most recently, there were 59 deaths in 2010.[10] Though the productivity of timber harvesting is low, before anything else, there is a major safety problem.

The tree-felling manipulator we are developing concentrates on the work of cutting trees. The tree-felling operation is the most dangerous operation in forestry, and the majority of industrial accidents occur during tree felling.

A worker holds a chainsaw with a blade that rotates at high speed (7,000–10,000 rpm), and stands right beneath the tree that he is cutting. The tree is 15 to 16 m high and weighs several hundred kg. Tree felling by humans has low accuracy and is unstable. Fatal accidents occur as the worker is struck by a high-speed rotating blade or is crushed by the tree that may fall in an unexpected direction.

2.4.2 Forestry machines

Along with the conventional forestry machines such as chainsaws, there are large heavy forestry machines.[Term 1] Moreover, there are two ways of collecting lumber: carrying the cut lumber on vehicles (the vehicle system) or hanging the trees on wire suspended in air (the wire system). Forestry machines of the vehicle system were developed in the US and Europe, and many are large heavy machines with multiple functions. In Japan, the attachments are imported from overseas, or even copied. And they adapt to the vehicle-type construction machines that are made in Japan. In some cases, the whole base is imported.

The large heavy forestry machines have increased from 23 machines in 1988 to 7,089 in 2014.[12] Subsidies are given to such machines, and the subsidy rate is one-half. However, even with this rapid increase, the production volume has not increased and the number of serious accidents have not decreased. Forestry in the US and Europe is conducted in rolling hill areas, and the large heavy forestry machines are made to work in such environments. It is difficult for such machines to fully show their capacity and performance in Japan.

Compared to the forests in the US and Europe, the Japanese mountain forests have steeper slopes, more complex landforms, and soft-ground terrains. Moreover, they have high temperature and high humidity, and the organization and maintenance/management of road networks are not easy as in the US and Europe. The road network density in mountain forests is low at 19.5 m/ha (2013).[13] Large heavy forestry machines that cut trees include the harvester and the fellerbuncher. These forestry machines cannot be driven on Japanese public roads, and must be transported on a trailer to the points where public roads meet forestry road networks. These forestry machines can enter the areas only where drivable roads are organized. Also, the work range is limited to the reach of the boom or arm.

Figure 4 shows the distribution of slopes in Shizuoka Prefecture. The percentage of forest coverage in Shizuoka is 64 %, and the forest covers the entire prefecture except for the urbanized area along the coast. The figure shows that the forestry is conducted in steep slope areas. It is dangerous to build roads on which large heavy forestry machines can be driven in the steep slope areas. To drive and to do forestry in

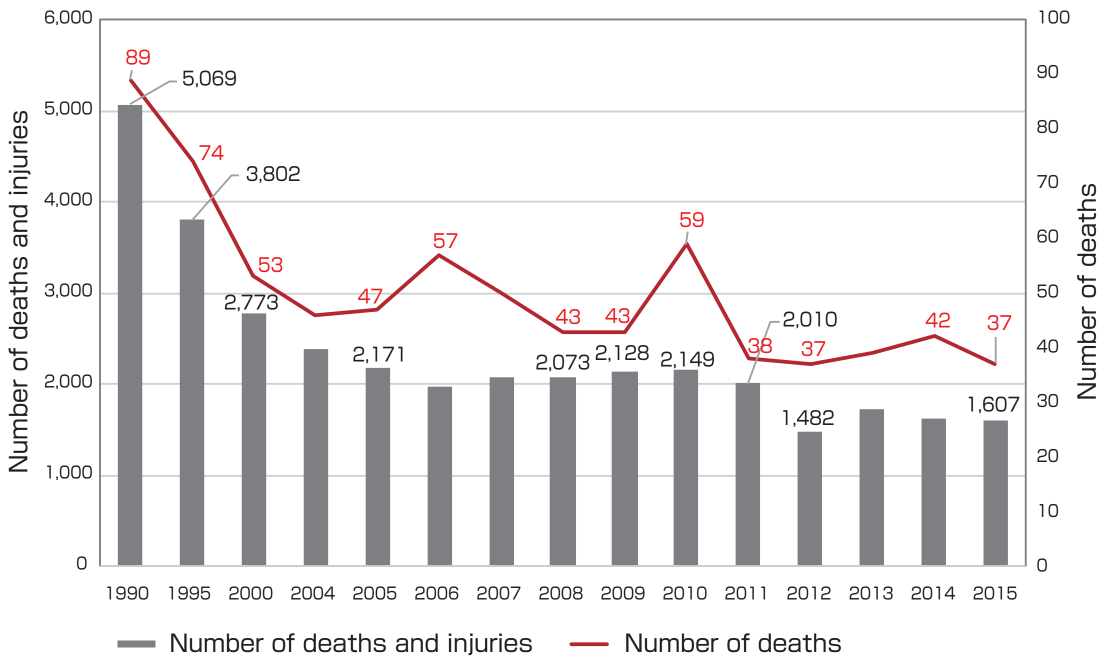

Fig. 3 Industrial accidents in forestry[10]

that area is even more dangerous.

Most of the tree-felling work in Japan is done by chainsaws even to this day. In a field survey by the author, a researcher of the University of Natural Resources and Life Sciences in Vienna answered that although the conditions are not as severe as Japanese mountain forests, Austria has relatively steep slopes within Europe, and 89 % of the felling work is done by chainsaws. In Japanese forestry, the "portable machines" that can be carried by humans such as chainsaws or a device proposed herein and the wire-type machines where the trees are hung on suspended wires continue to be used today, and improvement and development of such machines are necessary. Without these devices, timber cannot be harvested in many regions of Japan.

2.5 Production supply and demand structure

Another uniqueness of Japanese forestry is that the production and supply are done without a clear picture of outlets, quantity, or prices. If all the regions around Japan produce timber of similar quality at once only by inducement of subsidies, without knowing who will buy, how much, or at what price, the timber prices will decline. The price of Japanese timber continues to decrease and have become less expensive than imported timber.[14][15]

2.6 Culture of the industry

Chips and pulp are originally made from broad-leaved trees. Moreover, chips and pulp have low commercial prices, and the reproduction cost cannot be returned to the mountain forests if one produces only chips and pulp.

The Japanese cedar and Japanese cypress are planted to be harvested as construction materials. However, the domestic production volume has dropped, and the lumber material declined to 35 % (as of 2013) of the 1960s.[15] Even if one wishes to use solid natural wood that makes use of the property of wood itself rather than laminated wood or veneer, it is difficult to obtain such a material. The distribution is better for wood that is imported from abroad as lumber industrial products.

Particularly, the lumber needed for traditional Japanese wooden architecture is valuable, and the amount of wood used per unit surface area is high. However, traditional wooden architecture is difficult to build, even at a residential level, due to current laws as well as time and cost needed for construction permits.

Japan has had a culture of wood throughout its history, but the industry and culture that handle wood are declining. The industry and culture that raised the value of wood and contributed to the integrity of Japanese forestry are disappearing.

3 Objective and scenario

Many current artificial forests were planted during the forestation expansion period after the World War II, and they are ready for harvesting. The areas that were natural forests before forestation expansion have been planted and nurtured only by the instruction of the Government after the War. Therefore, people have very little experience in forestry. Yet, there are forests where forestry has been done since the medieval era in Japan.[16] Against the specific properties of Japanese forestry as mentioned before, importation or copying of the machines and extrinsic development using foreign systems will not endure. The revitalization

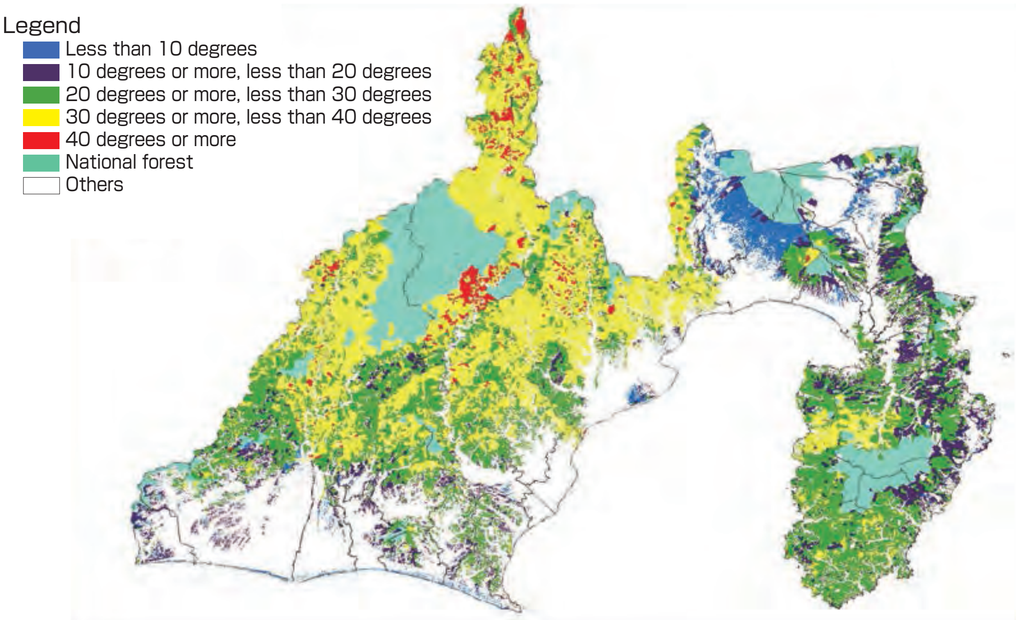

Fig. 4 Distribution of slopes in Shizuoka Prefecture (source: Shizuoka Prefectural Government)

of Japanese forestry must be based on Japanese nature, industrial culture, history, and local communities. Here, we set the revitalization of Japanese forestry as an industry as our goal, and we shall describe the elements and the scenario to achieve this goal.

As elements that must be studied for achieving the goal, other than technological issues, there are problems of inability to determine the owners or property boundaries of forests, difficulty of conducting efficient forestry due to strong rights of ownership, and the problem of the subsidy system, production supply, and demand structure.

First, innovation occurs on site. The ingenuity of the people on site, the ability to make decisions and carry responsibility, and the autonomy and sustainability of the industry must not be taken away. The damaging subsidy system, where the independence of private companies and on-site workers are destroyed, is contrary to the original objective. The subsidy money is not a tool to make people do what the administration wants, but it should be designed so the subsidized businesses and industries will take off and become independent of the administration.

One characteristic of forestry that should be pointed out is that the public function of forests contributes to the external effect that does not go through the market economy. In Europe, the maintenance of the public function of forests is perceived as "responsibility" that is attached to the "right of ownership by the owners. In Japan, it is the opposite, and there is a tendency to protect forests with public funds and systems because forests are considered having a public role even if they are privately owned.

Rather than designing a system where the person is compelled to do something using subsidies, which are our taxes, it is important for each person to play a steering role in a place where he/she is the main player. There is expectation for the administration to do things that can be done only by the administration, such as figuring out how to incorporate public and private roles of forests into the social system or how to create a system to recover and enhance the independence and sustainability of forestry as an industry.

Pertaining to the production supply and demand structure, it is necessary to revive distribution in our modern times by returning to the original purpose of tree planting, and by processing and high added value lumber that will bring back the reproduction cost to the mountains. This is the recovery of industry and culture that sees wood as wood. By reviewing the social mechanism that we have created, the technological development that aims at ensuring safety and productivity will become useful in severe natural environments. The vitalization of Japanese forestry will be limited with technological development alone.

3.1 Positioning and role of technological development of this research

Technological development should be inspired by the specific properties of Japanese forestry, rather than relying on foreign innovations. It is also necessary to "engage in technological development through 'Japanese' ideas when dealing with Japanese mountain forests." Therefore, we propose technological development that is related to neither forestry machines from overseas nor conventional forestry machines, but somewhere in between.

Forestry operation begins from *jigoshirae*[Term 2] or preparation, goes through processes of planting, weeding, pruning, clearing and thinning, and then final harvesting. Harvesting starts with complete enumeration[Term 3] where standing trees are surveyed one by one, work of felling the trees, log-making where the branches are removed to form logs, yarding: collection where the timber is taken to the timber yard, and of transporting the timber to the market. Other than the trucks running on public roads, there must be some kind of technological development. According to the survey done by the author, it was found that technological development was necessary for felling trees in terms of safety, yarding for productivity, and weeding in terms of labor intensiveness.[17] In the scenario for technological development, the most important point is to decrease or eliminate the chances that people who work in forestry may die or get injured. The causes of industrial accidents must be removed from individual operations. Next, the productivity needs to be improved. To do this, it is necessary to develop an overall system to increase the efficiency of work by advancing each operation and linking the individual operations without any delay. It is necessary to build a production and distribution system that links the forests to consumers, from the timber yard in the mountains to the log market without any delay. It is also necessary to consider demand and supply, production, and mountain management in the future. Figure 5 shows the elements extracted for the goal and the scenario, and the positioning of our development.

3.2 Survey before starting the development of the machine

The author started a survey by visiting the tree-felling sites in mountain forests, forestry cooperatives, log and product markets, lumber mills, and others to collate needs with seeds. Then, application was submitted to the "New Interdisciplinary Field Research Strategy Access Survey" (FY 2006) of the Ministry of Agriculture, Forestry and Fisheries (MAFF), and the application was selected. In this project, the problems were organized, the technological demands were extracted, and the objective and topic settings were done. This information was presented in papers at the Robotics Society of Japan.[17][18] The findings up to this time of research from forests to timber to wooden architecture were reported to the general public in a book.[19] Afterward, on-site surveys in Japan and overseas were continued, the project was selected by MAFF in 2011, and the actual development of the machine started.

3.3 Approach taken to start development without precedence

As a technology for felling trees, there was an attempt at automation of chainsaws as a specialized vibration proof measure.[20] However, it did not reach practical use, and there was no more development after 1970. We were unable to start the development based on prior research. There were many given conditions that had to be considered in the design. Therefore, rather than starting by gathering representative values that would serve as design indices through simulation, we started by fabricating a simple machine and actually using it on site. Why did we start on site? That is because what can be expressed as numbers and words, and what can be determined based on them are limited. If a phenomenon is represented by a certain index, the phenomenon that is left outside the number is ignored. In machine development, it is necessary to nurture the experience and insight on site, very much like forestry technicians. Conversely, if the work does not require general perception or decision of a living human being, there wouldn't be so many industrial accidents, and a machine that can replace human workers would have been already developed.

3.4 Induce an optimal solution rather than one solution

The forestry worker looks at the diameter of the tree and determines the size of cut that will be made to the tree. This size is based on experience, and there is no numerical basis. In the work environment of mountain forests, no two conditions are the same including branches and leaves on the soil or forest floor, stumps, and position of the surrounding trees. Moreover, the differences are significant. Taking the example of water content in Japanese cedar, the individual differences among trees that are the target of felling work can range from 50 to 250 %. Even in one single tree, the direction of grain may differ depending on where the cut is made. Therefore, simulation that fits work environment and subjects into a single model may bring about one solution, but it is not known whether that solution is applicable to all other work environments or subjects. Therefore, we thought it was necessary to obtain an optimal solution rather than one solution. To do so, we created a prototype that realized the minimum movements, conducted actual operation on site, and went on to the next development from the results obtained. This process was repeated to approach the optimal solution model. As a result, we created a new design theory as presented below.

4 Creation of a new design theory

4.1 Design theory for a machine for Japanese mountain forest

Before starting the machine development, we found a new design development method, and by employing this method, we succeeded in demonstrating the effectiveness of the machine created in two years since the start of the development. It was a design theory that was inevitably reached after considering a machine that can be actually used on site, after surveying the sites of Japanese forestry.

There are design theories such as the V model[21] (Fig. 6). However, in developing machines for the Japanese mountain forests and assuming actual practical use, it was difficult to develop something that can be used on site, using the design method for machines that are generally imaged as robots. Machine that may be able to do everything may turn out to be useless in practice.

Taking the V model as an example, the process goes from the left-top of Fig. 6, then down, and reaches the right-top.[21] From the left-top to the bottom, all the requirements for design that

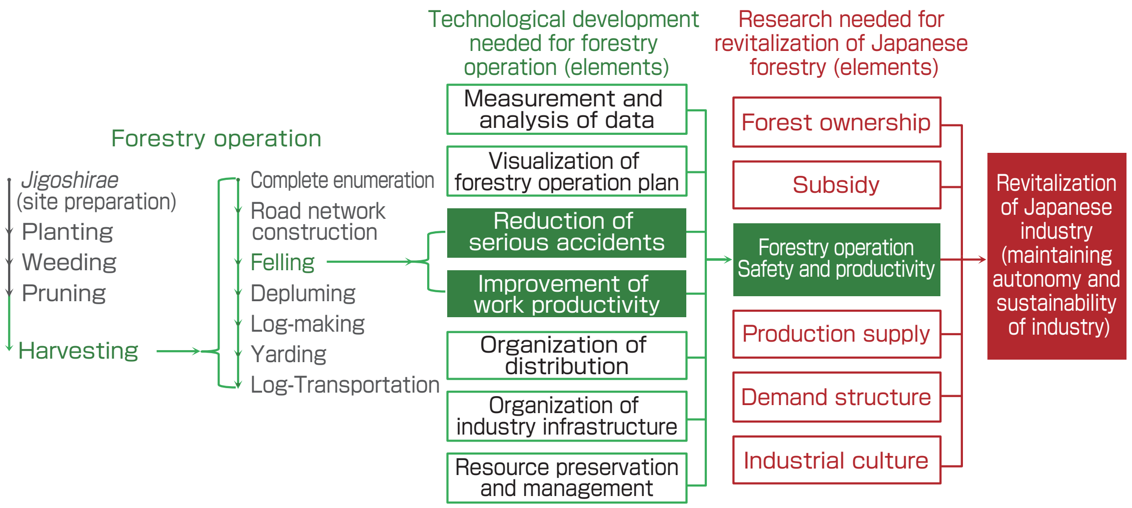

Fig. 5 Positioning of the technical development for the goal and scenario

one initially thinks of are thought out, the limiting conditions are considered, the specifications are carefully reviewed, and the design is narrowed down. A machine is developed, experiments are done from the bottom to the right-top, and the process finally reaches the demonstration experiment on site. In the design theory discussed in this paper, contrary to the flow, the process starts from the right-top, and a machine is operated on site to do the actual work. A machine with the minimum necessary moves is created, and is set to work at actual work sites. Rather than replacing all thinkable moves with machines, first, it is narrowed down to "this much must be done by machines" when working in the Japanese mountain forests. Those include heavy labor or maneuvers for which many serious accidents occur. In a natural environment, nothing is better than human sensing and control. It is not necessary to take these away from the workers as that may lead to danger. If all replaceable work is done by machine, the superiority of machines doing the work is lost. Cases of failure of surpassing the threshold are seen in robots that were developed for on site use. As a reaction, there is a tendency in robot development to realize robots with a single function only.

As the demonstration experiment on site of the right-top is started and the specifications become clear, the findings are incorporated into indoor experiments. It goes from the right-top to the bottom. The necessary functional requirements are added to the minimum motion needed for the machine, and the specifications are listed as a result. Finally, an overall system is created, and the flow goes from the bottom to the left-top.

The design theory of the development of our machine is summarized in Fig. 7. The objective of TATSUMI reported here is to ensure safety of tree-felling. To achieve this objective, first we aimed to have the most important "movements that must be done by a machine" with the ultimate simplification of the shape and mechanism of the hardware. There are no two same conditions for the environment and target for using the manipulator, and the differences are great. Therefore, if

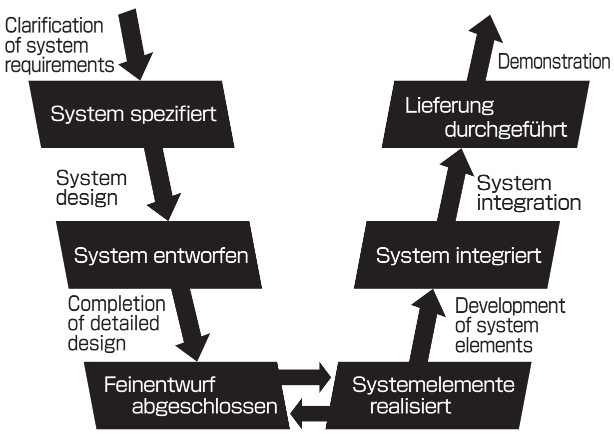

Fig. 6 V model[21]

specifications are clearly set at first and designed, the machine may become unusable in practice, and the specifications that have been once set may have to be changed. Therefore, it is necessary to rotate the "design spec determination loop" shown in red in Fig. 7. TATSUMI I and II were developed using this loop. This is similar to the design theory for the web, since it is similar to the characteristics of web technology where "one does not know who will use it, how he/she will use it, and in what kind of connection environment it will be used" and where "one does not know what will happen unless it is made, implemented, and run in the real world." In both cases, unless one experiences the operating environment including humans and the interaction, the specifications will not be usable in real society.

Next, we entered the development of TATSUMI III, and utilized the green "system integration loop." The development of this machine was the integration of the design spec determination loop and the system integration loop, and particularly, the development by the design spec determination loop was important.

4.2 Machine conceived from the sites of Japanese forestry

In the process of going from the right-top to the bottom in the design development process, we decided to use the commercial chainsaw in the tree-felling portable manipulator TATSUMI. If we had started from the clarification of system requirements in the left-top, we would have not used the chainsaw but started by designing the cutting function. Because we were creating a machine that could be used on site, we decided it was more realistic to mount/dismount an already available chainsaw.

A large heavy forestry machine harvester is also equipped with a saw chain. There is no better blade for cutting down

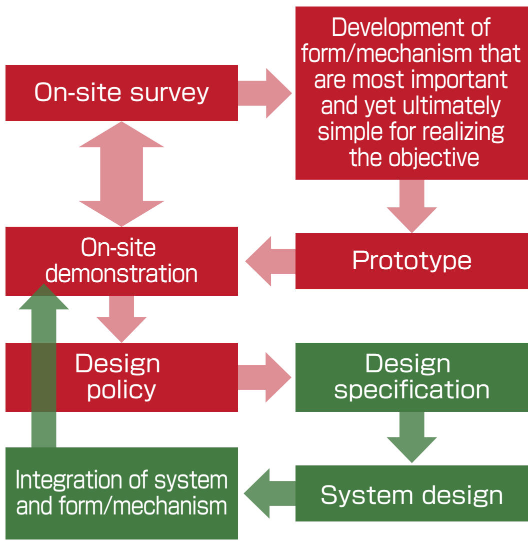

Fig. 7 Design spec determination loop and system integration loop

Table 1. Outline of developed machines

Name	Characteristic	Development period
TATSUMI I	Composed only of rectilinear pair,[Term 5] 3 degrees of freedom ※Feasibility of proposed machine investigated	2011-2012
TATSUMI II	Composed only of turning pair, 3 degrees of freedom	2012
TATSUMI III	1 rectilinear pair, 3 turning pairs, 4 degrees of freedom ※One manipulator model (optimal solution) presented	2012-2013
TATSUMI IV	1 rectilinear pair, 3 turning pairs, 4 degrees of freedom ※Joint improved, turn control function for chainsaw added	2013-2014
TATSUMI V	1 rectilinear pair, 3 turning pairs, 4 degrees of freedom ※Operability of installation, removal, setting and carrying improved by separating the fixture and working part of manipulator	2014-
TENRYU I	Composed only of rectilinear pair ※Feasibility of proposed machine investigated	2011-2012
TENRYU II	Develop and install drill/end mill with diameter of cut surface ※Main difference between II and III is blade length and cutting orbit.	2012
TENRYU III	Develop and install drill/end mill with half diameter of cut surface ※One manipulator model (optimal solution) presented	2012-
MOBILITY I	Composed only of crawler[Term 6]	2012-2013
MOBILITY IR	Composed of crawler and tow arm ※Hill climbing performance by tow arm improved	2013-2014
MOBILITY II	Composed only of newly developed crawler ※Response to load shift and position shift by unevenness of slope and ground improved	2014-

trees than saw chains. Chainsaws are widely used, as can be seen in the numbers: There are about 180,000 chainsaws being used (in 2014) according to official figures for about 50,000 forestry workers (numbers from 2010).[12] There is no forestry worker who does not own or use this tool. It is present at all sites. The road network density in forests is low at 19.5 m/ha (2013),[13] and most workers must reach the tree-felling site on foot. The Japanese chainsaws have evolved into a compact form. We aimed at downsizing and weight reduction of the manipulator by employing the chainsaw that is continuously being improved for small size and light weight.

Among accidents during tree-felling, a tree that a worker is cutting may fall in an unexpected direction and crush the worker. Also, a tree may fall in an unexpected direction and become a hanging tree,[Term 4] and this hanging tree may fall and crush the worker as the worker cuts the trees around the hanging tree. Deaths occur in such accidents. The worker concentrates on operating a chainsaw at the foot of a tree, and it is difficult to detect changes of movement of the nearby branches and changes in the situation. He may not notice when the tree starts to fall, and it may be too late to get out of the way leading to a serious accident. If the person is not holding and operating a chainsaw at the foot of the tree, such danger can be avoided. One of the reasons that trees fall in directions unexpected by the workers is because the accuracy cannot be obtained by human work. If chainsaws can be operated by machines, it will be possible to stabilize work accuracy, trees can be fell in the intended directions, and this can prevent workers being crushed by trees or hanging trees.

Moreover, the rotation of the chainsaw engine surpasses 10,000 rpm at full throttle. To maintain distance from this blade rotating at high speed during tree-felling not only will prevent serious accidents, but also will reduce vibration disorder.

By creating a distance between the worker and the trees and the chainsaw, industrial accidents that arise from these two danger factors are removed, and by stabilizing the cutting accuracy of trees through machine work, trees can be fell in intended directions. These were set as the objective of machine development.

5 Case studies

In this chapter, the results of the actual development will be described. Table 1 shows the outline of the developed machines. The machines include the manipulators to cut down trees named TATSUMI[22][23] and TENRYU[24] (Fig. 8), as well as the transport mechanism[25] (Fig. 9) that carries heavy items and follows the worker through forests where road networks are not available. In this paper, TATSUMI will be described. Details of the design and experiments on the tree-felling portable manipulator TATSUMI III and IV are given in the patent[23] and the paper[22] presented at the Japan Society of Mechanical Engineers.

5.1 TATSUMI I

TATSUMI I (Fig. 10) was composed only of a rectilinear pair[Term 5] that linearly moves the chainsaw in three axes. TATSUMI II (Fig. 11) that was developed next was composed only of a turning pair. In TATSUMI III (Fig. 19), the rectilinear pair and the turning pair were combined from the findings obtained from TATSUMI I and II.

The engine chainsaw was automatically operated by TASUMI I, and it was confirmed that the cutting operation could be accomplished normally. There were no looseness of chainsaw fixture or effect on the encoder by vibration, no leakage of fuel due to changes in machine positions that do not occur in normal use, no interruption of fuel supply, and it was possible to cut into trees in constant speed.

5.2 TATSUMI II

TATSUMI II was composed only of a turning pair, whereas TATSUMI I was composed only of a rectilinear pair. From the findings from I and II, the optimal type and arrangements of joints were investigated for TATSUMI III.

5.3 Design policy

The design policy obtained through the survey on site and TATSUMI I and II will be explained.

(1) The device must be the simplest possible mechanical apparatus resulting from the pursuit of ultimate simplification of the shape and mechanism of the hardware. Also requires minimal external recognition and control.

(2) Respond to diverse environment and diverse work with simple motion created by the machine. That is, the aim is a mechanical device that creates simple motion that can handle all patterns of work in all kinds of environment. The machine itself does not realize all patterns of work.

(3) For ideas of mechanism, *kata* (form) and *shosa* (gesture) that were handed down traditionally are valued.

(4) The aim is a mechanical device that can work with nimbleness and mechanism that utilizes the form and weight of the target, like in judo. Downsizing and weight reduction are achieved.

(5) The most important components in the work process are people.

In (1), a simplified mechanical device is aimed at by the pursuit of ultimate simplification of the shape and mechanism of

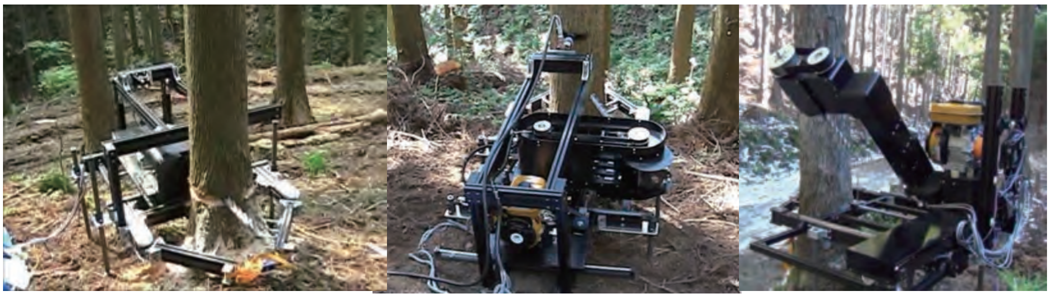

Fig. 8 TENRYU: From left I, II, and III

Fig. 9 MOBILITY: From left I, IR (improved I), and II

Fig. 10 TATSUMII: Composed only of rectilinear pair

Fig. 11 TATSUMI II: Composed only of turning pair

the hardware, and minimal external recognition and control methods are wanted. The reason is because the machines must endure the temperature differences, heavy rain, and heavy humidity in the Japanese mountain environment. The overseas forestry machines have corrosion problems of the electric circuits when used in Japan. The natural environment is composed of elements that are difficult to identify, and the control of a mechanical device by automatic sensing tends to cause malfunction, and that is dangerous in work involving a blade rotating at high speed. Also, daily maintenance and repair must be done by the on site workers. Therefore, the mechanical device must have a simple and easy to understand structure.

There is a machine with a similar concept as (2). This is the “yarder” that is a typical Japanese forestry machine. The yarder is a device that merely rolls out and rolls in wire. It can be used in any mountain forests. A chainsaw itself is also simply a rotating saw. However, it is possible to cut down almost any tree with a chainsaw. On the opposite side stands a complex machine called a humanoid robot. Various findings can be obtained in its development process. However, if a humanoid is to be used on site, it will be a different story. A machine that seems to be able to do everything like a human does makes the significance of people doing the work and the purpose for machines replacing humans ambiguous. Therefore the superiority of machines may be lost.

For the work itself, there are many operations that are perceived similarly in forestry. The most common tree-felling method is the *ukekuchi-oikuchi-giri* (undercutting and backcutting method).[Term 7] Basically, most trees can be fell by cutting three planes with a chainsaw. The TENRYU, which we are developing separately from TATSUMI, employs the *mitsuhimo-giri* (three-hinge tree felling method) that has 1,300 years of history in Japan.[26] The *mitsuhimo-giri* is the method for cutting down the sacred trees used to build the Ise Shrine. The tree is notched in three places, and almost any giant tree can be cut down safety. While the *yoki* (Japanese style axe) is used in the ceremony, TENRYU reenacts this operation by a manipulator equipped with a special blade that has functions of a drill and an end mill.[24]

In (3), the work method handed down traditionally is used in considering the mechanism. In the *ukekuchi-oikuchi-giri* (Fig. 12) employed in TATSUMI, the size of the *tsuru* (hingewood)[Term 7] (Fig. 13) is handed down based on experience and there is no scientific basis. However, in many cases, these traditional methods employ the most reasonable form and gesture as the technique has evolved over a long period of time.

In (4), the aim is to create a device that works with adeptness and mechanism that uses the form and weight of the target, or neutralizes the opponent like in judo, rather than taking on force by force like the large Western machines against tall, heavy targets (trees). In other words, the technological development is conducted using the Japanese concept.

In (5), people are the most important components in the work process, and the system we aim for is not totally automatic, but a system where workers can enter correction and support of the work as well as cognition and decisions. Therefore, the goal is a machine that can be used easily and intuitively by the workers.

5.4 TATSUMI III

5.4.1 Specifications

The specifications of TATSUMI III was derived from the results of TATSUMI I and II.

(1) Weight that can be carried by a single worker in mountain forests away from forestry roads: 18 kg or less

(2) Ensured self-sustaining operations: Uses an engine chainsaw (an electric chainsaw is not suitable for felling work in mountain forests)

(3) Workability: The chainsaw can be mounted and dismounted (the chainsaw is used also before and after the tree felling)

(4) Selection of a tree-felling method that optimally uses the chainsaw’s tree felling capability: Use of *ukekuchi-oikuchi-giri*

(5) Warrant the safety of workers during tree felling: Automatic operation (Trees, which are mostly cedar and cypress planted after WWII, are 15~16 m high and weighs several 100 kg. The work is done right beneath such trees, and the worker must handle a blade rotating at high speed. If the worker can keep a distance from these two elements, accidents can be prevented.)

(6) Target diameter and tree species: Up to 350 mm for thinning purposes; artificially planted cedar and cypress (*Cryptomeria japonica* and *Chamaecyparis obtusa*)

5.4.2 Cutting process

The sawing orbit by which a tree is cut is shown in Figs. 12 and 13. The *ukekuchi-oikuchi-giri* method described above is used. The tree-felling direction is to the left for both Figs. 12 and 13. The *ukekuchi* (undercut) is the triangular space that is made in the direction of tree fall, and the *oikuchi* (backcut) is a single planar cut made on the opposite side of the undercut. As it can be seen, the undercuttig and backcutting method is suitable for chainsaws that are good at making planar cuts, and just three planar cuts come to fell the tree. The part remaining between the undercut and the backcut is the *tsuru* (hingewood), and this is C in the figure. When the notch is made followed by the backcut, the hingewood acts as the hinge, and the tree falls in the direction of the undercut (to the left in Figs. 12 and 13).

5.4.3 Functional requirements

Based on the findings obtained from TATSUMI I and II, functional requirements were derived to approach the optimal solution.

(1) Lower the sawing plane: When the harvested timber is used as lumber, the area near the root where the maximum diameter can be obtained is the most valuable, and the height of the stump (sawing plane) should be lowered. (In III, the L-shaped arm was devised to eliminate the intermediate turning joint in II. As a result, the height of the sawing plane was lowered 150 mm in III compared to II.)

(2) The undercut is made by turning the chainsaw: As shown in the left side of Fig. 13, if the tree is cut by turning, the chainsaw does not have to be moved out of the tree, the movement range of the manipulator can be reduced, and the device can be reduced in size and weight.

(3) Backcut is made by moving the chainsaw parallel, rather than turning the chainsaw: If the backcut is made by turning like the undercut, the width of the remaining hingewood may become uneven, and there is danger that the tree fall may start unexpectedly while cutting. To avoid this, the backcut is made by maintaining the end line of the undercut (line where top and bottom notch cuts meet) and the direction of the guide bar is kept parallel.

(4) The arm itself is moved to widen the applicable diameter: If one tries to widen the diameter of the wood to which the device can be applied or the work area, under the condition that only the base of the arm turns and the movement of the arm itself is fixed, it is necessary to lengthen the arm. If this is done, the torque at the base joint increases, and the weight and size of the device increases. Therefore, by moving the arm parallel, the work area is widened while maintaining the small size and the light weight. III was designed with this method, and its maximum sawing diameter increased to 350 mm, and it could fell larger trees than II.

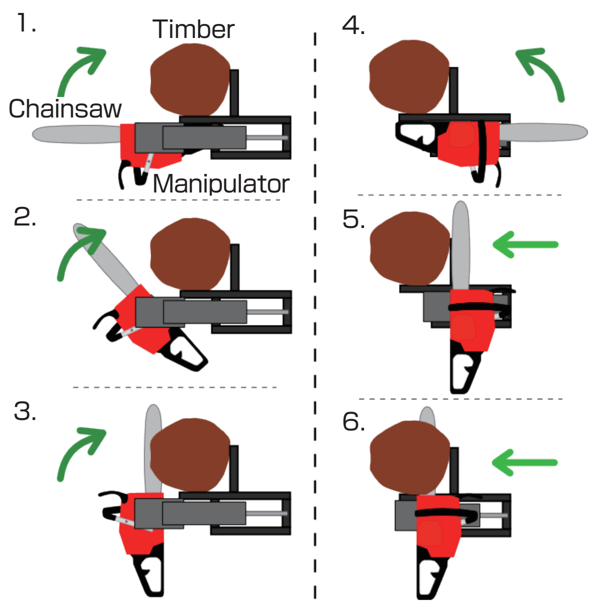

Fig. 12 *Ukekuchi-oikuchi-giri* method

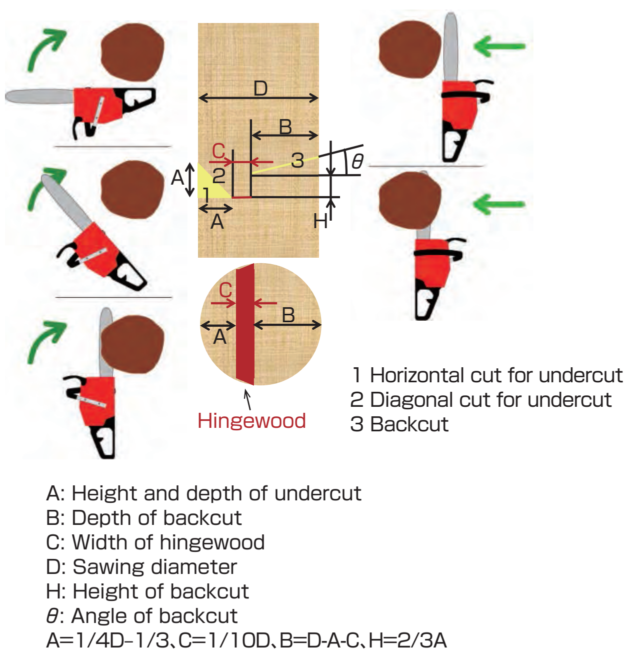

Fig. 13 Cut dimension and order of cuts

The undercut is cut by two turning moves, and the backcut is made with one horizontal movement.

5.4.4 Determination of major dimension—L-shaped arm

The overall form and dimensions (Fig. 14), the degree of freedom of arrangement (Fig. 15), and the rectilinear and turning allotment of each pair are shown. The first condition was to lower the height of the sawing plane. Therefore, an arm form where the height from the base, or the fixture position on the tree, to the chainsaw blade would be the lowest was considered. The mechanism that results in increasing the height was removed from between the base and the chainsaw. The chainsaw would not interfere with the arm when turning to undercut. The mechanism and form

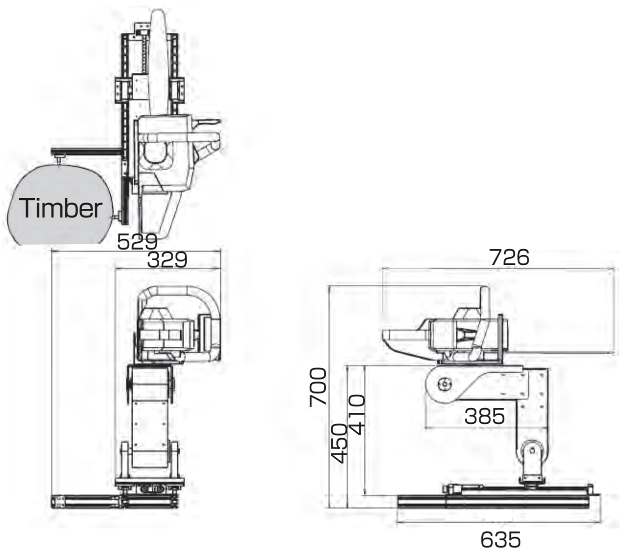

Fig. 14 Overall form and dimension

where the chainsaw came closest to the base were sought when the position for undercutting was taken. Based on these conditions, the design was done with the minimal dimensions that allowed holding the chainsaw, and we devised the L-shaped arm. The arm dimensions are shown in Figs. 16, 17, and 18. L1 and L2 are dimensions of the L-shaped arm, and L3 and L4 show the offset from the rotation axis at the tip to the sawing position when the chainsaw is mounted on the arm. The length of the linear motion part of the base was derived by calculating the amount of movement of the arm necessary for undercutting and backcutting.

The total weight of the manipulator is 18 kg or less, including the 6 kg for the chainsaw. The chainsaw used is ZENOAH (G3711EZ) with a 40 cc engine. The dimension in height is 700 mm, the longer direction size is about 725 mm, and shorter direction size is 529 mm, and it fits a space of 329 mm excluding the base bar.

5.4.5 Configuration of the joints

In this section, the number of joints and their arrangements are explained. To realize the aforementioned cutting process, a minimum of four degrees of freedom are desirable. Four joints are set: the lowermost joint for linear motion (Joint A, rectilinear pair), the joint to turn the chainsaw by the rotation of one axis (Joint D, turning pair), the joint to turn the L-shaped arm (Joint B, turning pair), and the joint to turn the chainsaw in the opposite direction (Joint C, turning pair). The range of motion of each joint is shown in Table 2.

The designs of each joint are explained. The chainsaw is made to cut forward due to the form of the saw chain. Therefore, the reaction force when cutting is at maximum when it first contacts with the tree and starts the cut. The reaction force was measured to be about 20 N. Other than using this value for the design of Joints A and D, the mechanisms of other joints were designed from the rotation moment in a stationary condition (Tables 3, 4, and 5).

In considering the mechanism, the method handed down traditionally is utilized. The undercutting and backcutting is done by workers using the chainsaw, and this is taught as one of the tree-felling methods with the least burden on the workers. This proposal substitutes the handwork with a manipulator, and it was confirmed that the manipulator would not be subject to excessive reaction force that surpasses human power in both TATSUMI I and II.

5.4.6 Control system

A program to numerically control the work was developed. With this algorism, the sawing orbit is automatically calculated from the timber size, and this is sent to the control program to control the manipulator. The machine language used is C++ considering its universality.

Table 2. Range of motion of the joints

Joint	Range of motion
Joint A [mm]	0.0-320.0
Joint B [deg]	0.0-127.4
Joint C [deg]	-180.0-10.0
Joint D [deg]	32.0-45.0

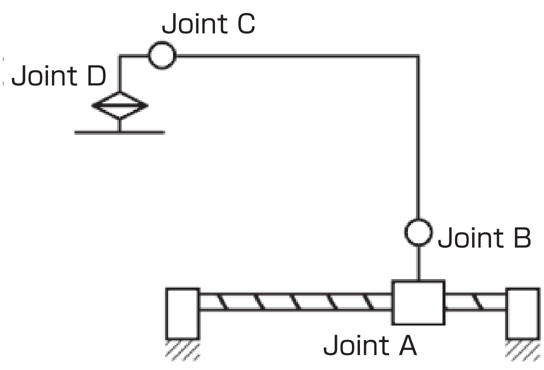

Fig. 15 Arrangement of degrees of freedom

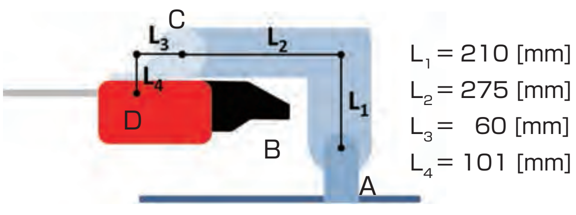

Fig. 16 Dimension of arm

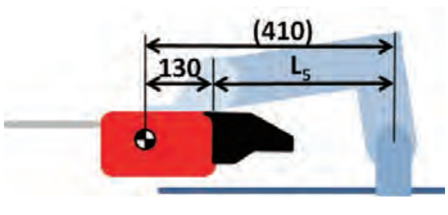

Fig. 17 Maximum extended size of Joint B

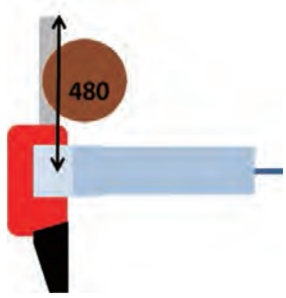

Fig. 18 Turn radius of chainsaw

Table 3. Reduction ratio

Joint	Name	Reduction ratio
Joint A	Gearhead	139:1
Joint B	Timing Pulley	3:1
	Hamonic Drive	160:1
	Total	480:1
Joint C	Timing Pulley	4:1
	Hamonic Drive	100:1
	Total	400:1
Joint D	Timing Pulley	4:1
	Hamonic Drive	100:1
	Total	400:1

Table 4. Specification of motor

	Joint A	Joint B	Joint C	Joint D
Maker	Maxon	Maxon	Maxon	Maxon
Model	RE25	EC60flat	RE30	RE30
Weight of motor [g]	130	470	260	260
Nominal voltage [V]	24	24	24	24
Assigned power rating [W]	20	100	60	60
Nominal torque (max. continuous torque) [mNm]	26.3	221	85.6	85.6
Max. speed [rpm]	14000	6000	12000	12000

Table 5. Specification of gear

	Joint A	Joint B	Joint C	Joint D
Maker	Maxon	Harmonic Drive Systems		
Model	GP26A	CSD-20-160-2UH	CSD-17-100-2UH	
Weight [g]	93	650	460	460
Reduction ratio	139:1	160:1	100:1	100:1
Permissible max. input rotational speed [rpm]	8000	6500	7300	7300
Permissible max. value of ave. load torque [Nm]	4.5	34	27	27

5.5 Verification Test

5.5.1 Demonstration experiment for TATSUMI III

A demonstration experiment to cut down trees in an actual mountain forest using TATSUMI III was conducted (Fig. 19). The location of the experiment was the Tenryu region of Hamamatsu City, Shizuoka Prefecture, and the subject was cedar of an artificial forest. The diameter at chest height was 240 mm, the sawing plane diameter was 265 mm, and the tree height was about 15 m.

The accuracy of the tree-felling operation is evaluated according to the size of the undercut and the backcut, the shape of the hingewood, and the direction to which the tree actually fell against the scheduled felling direction (actual tree-felling direction). The performance of manipulator III was evaluated according to this method.

It was verified that the operation from the horizontal cut of the undercut, the diagonal cut of the undercut, the removal of the undercut piece, the backcut after changing the position, and then the felling the tree could be accomplished with TATSUMI III.

One issue was that the diagonal cut for the undercut was lower than the targeted line, and the undercut became smaller. This was because the shaft that transferred the torque was not sufficiently stabilized in the joints at the base and the tip of the manipulator.

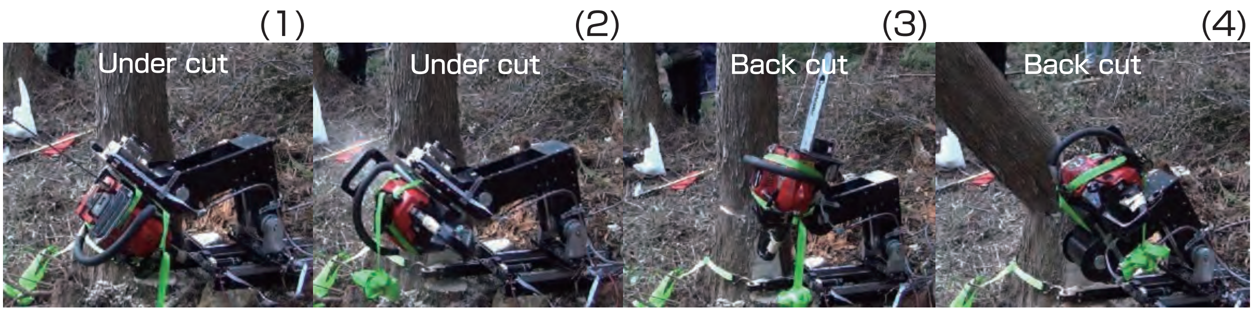

Fig. 19 TATSUMI III
(1) and (2) are diagonal cuts of the undercuts, (3) and (4) are backcuts.

5.5.2 Demonstration experiment for TATSUMI IV

In TATSUMI IV, the joint area of TATSUMI III was improved, the fixture base could be removed from the manipulator, and the rotation frequency of the chainsaw could be controlled remotely. In the four experiments conducted with TATSUMI IV (Fig. 20), it was verified that the operation from the horizontal cut of the undercut, the diagonal cut of the undercut, the removal of the undercut piece, the backcut after changing the position, and then the felling the tree could be accomplished. The sawing diameter of the four trees shown in Figs. 21 and 22 were D = 260 mm, 250 mm, 230 mm, and 270 mm, respectively. The times required from undercutting to felling were 278 s, 330 s, 219 s, and 267 s, respectively. The chainsaw made the cut into the tree at full throttle, and the manipulator moved the chainsaw at 5 mm/s.

5.5.3 Cutting line of the undercut, the backcut, and the hingewood

Fig. 20 TATSUMI IV

In forestry, the differences in skill of tree-felling are determined by the cutting line of the undercut, the backcut, and the hingewood. These were used for evaluation. The operation by humans is not stable, and the undercut and the backcut are not made accurately, and the hingewood does not turn out as intended. Therefore, in the work by humans, the tree may fall at unexpected timing in unexpected directions during the operation, and serious accidents may occur. The stability of work and sawing accuracy can be evaluated by the cutting line of the undercut, the backcut, and the hingewood that remain after the operation.

Figure 21 is a view of the cross-sections of the stumps of the fallen trees. Blue is the target line and red is the actual cutting line. In all experiments, the diagonal of the cutting line almost completely matched the target line. To control the felling direction of the trees, it is necessary that the undercut made on the tree side, to which it is expected to fall, is precisely cut out according to plan. In the experiment, the end line of the horizontal cut and the diagonal cut of the undercut matched. The operation by the proposed mechanical system was stable, and it was confirmed that the sawing accuracy was high.

Figure 22 shows the shape of the hingewood remaining on the stump. The target value of the program was that the hingewood width should be 1/10 of the tree diameter. Considering the phenomenon where the hingewood is torn when the log separates from the stump, the hingewood was made according to the program instructions. The guide bar of the chainsaw does not have constant width over its entire

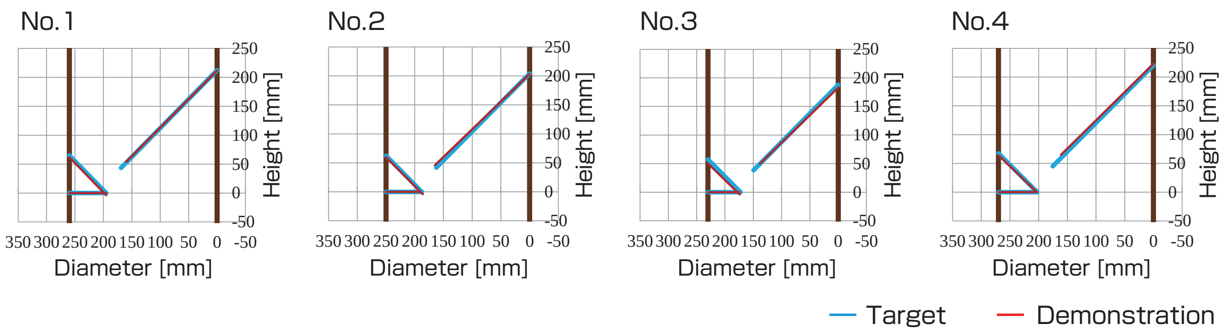

Fig. 21 Cut line of the undercut and backcut

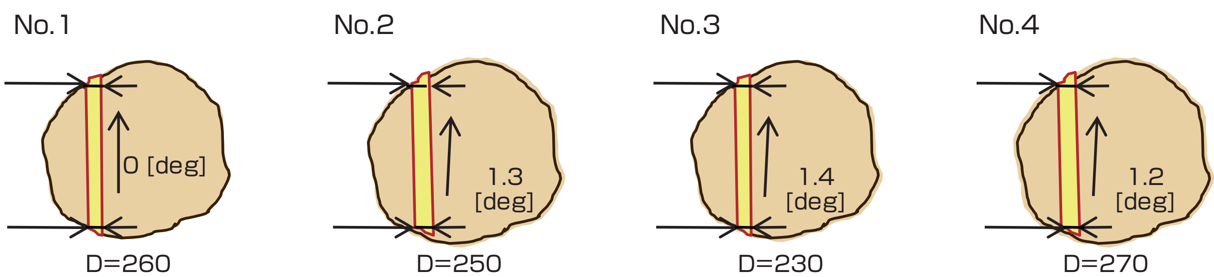

Fig. 22 Shape of hingewood

length, but becomes narrow at the tip like a rugby ball. It was predicted that when the sawing diameter approaches the effective length of the guide bar, the hingewood on the other side of the manipulator followed the shape of the guided bar and would be left thick. It was also predicted that the hingewood on the deeper side will be left thick due to the shape of the guide bar, it would be difficult to cut the hingewood, and the felling direction might be disturbed. However in the experiment, the hingewood width did not become uneven so as to affect the felling direction, and the error of felling direction was 1.4 deg or less, which was hardly significant.

Demonstration was done for the manipulator operation with the worker standing away from the tall, heavy tree and the chainsaw rotating at high speed that are often causes of serious accidents. The operation itself was stable, the sawing accuracy was evaluated, and we were able to demonstrate the effectiveness of the proposed manipulator.

5.6 TENRYU and MOBILITY

TENRYU realizes the *mitsuhimo-giri* using a special blade that has the functions of a drill and an end mill, rather than an axe. There were two and four blades in the end mill of the special blade for TENRYU I and II, and there were three in TENRYU III. The blade length that was the same as the sawing diameter in I and II was reduced to half in III. The tree-felling operation of trees was demonstrated with TENRYU III. Since then, improvements were made to increase the stiffness of the manipulator. For MOBILITY, after the development of I and the IR transport mechanism, II was developed after devising a new suspension mechanism based on the findings of the first two. The II transport mechanism demonstrated performance needed on uneven ground such as driving over the barrier of 245 mm only with a crawler[Term 6] on one side, assuming irregular ground in mountain forests.

6 Prospects for the revitalization of Japanese forestry

6.1 Prospect for TATSUMI

One of the roles of universities is to study the topics that are problems in society, think about how to solve the problems, and to propose new ways of thinking, mechanisms, and methods unseen before. We were able to present a certain optimal solution for maintaining safety in tree-felling operation through our research. In the future, we plan to hand over this new idea to a private company for realization and engage in development together. Since companies have different objectives than university research, development will take place with participation of corporate experts on sales and diffusion etc. Currently, the issues for TATSUMI are further downsizing and weight reduction, as well as improvement of ease of portability and installation on the tree. Another future prospect is the investigation of the chainsaw engine. The displacement of the engine chainsaw used in Japanese forestry is 40–60 cc, and for a person or a manipulator this is like having to hold up a motorcycle engine. Therefore, consideration is made for motors other than the engine and the separation of the blade and the power source.

The issue other than technology for TATSUMI is the diffusion method. From the manufacturing cost of the manipulator, we feel there is no particular problem for cost. The prices of large heavy forestry machines such as harvesters or fellerbunchers are tens of millions of yen just for the attachments.

6.2 Prospect for technological development

Of the elements that require research for the revitalization of Japanese forestry, the maintenance of safety and improvement of productivity in forestry are important for technological development. The order of technological development is first, to eliminate serious accidents from individual operations, next to improve productivity, and then, to construct the overall operation system that smoothly links one operation to the next. Even if the efficiency of one operation improves, if the operations before and after do not change, the operation stalls there. One harvesting operation may involve surveying the mountain forest, determining the target of harvest, cutting the trees down, gathering, and transporting, and the whole series of tasks must be improved to improve the overall productivity.

As described in Subchapter 2.1, the advancement in measurement of resource and data analysis for forestry is essential. The technological development is being started, but most of the tasks are currently still done by hand. Sophistication of this enormous amount of work is necessary. Also, how to set wires and road networks in mountain forests relies on experience and insight of the on site workers. Even if a landslide occurs, feedback cannot be done. At this moment, it is impossible to create a vision on how road networks are laid out in mountain forests and throughout the whole regions. The technology to visualize the management plan and how to carry on forestry from there are necessary. The technology to “make visible” the forest and the operation, or to be able to grasp the operations in real time, is desired.

6.3 Prospects other than technological development

In Chapter 2, the specific properties of Japanese forestry was explained, and it was pointed out that it was necessary to review the social system and establish a new plan, not only technological development. Here, the revitalization of Japanese forestry will be discussed from a different perspective.

In Germany, there are many people who engage in both farming and forestry, and there are some farming machines equipped with winches to pull the logs for forestry use. In Japan, there is almost no doubling with agriculture, and some people who work in other industries have even forgotten that they own mountains. With the appearance of machines like TATSUMI, I hope that people who do not specialize in forestry will become interested in the new Japanese forestry, and start taking care of our mountains.

Industrial products needs to be produced quickly and inexpensively, and the product value is highest when they are shipped and then tends to decline rapidly. Lumber, however, has the characteristic of gaining value over time through careful production, from growing the trees in the forests, processing including drying, and use in wooden architecture. It is necessary to regain a society that approves the production and distribution of such products and that appreciates the value. From the author’s local surveys overseas, in Europe, the workers, the craftsmanship, and their work are valued in the community, and considerations are made for their coexistence in the industrialized society. Thinking of how to revitalize and to make independent industries such as forestry that concern mountain forests that cover 67 % of our land may give us a clue to revitalizing the local communities.

7 Conclusion

The TATSUMI project presented in this paper obtained development funds in 2011, the first machine was developed in FY 2012, and a manipulator model was proposed as III in the same year. III was able to conduct a series of maneuvers from undercutting to backcutting, and successfully cutting down trees while workers maintained a distance. In the following IV, the sawing accuracy was evaluated, and it was demonstrated that it could cut down trees in targeted felling directions. It was confirmed that there were no errors in the sawing accuracy and the operation was stable. Improvements are being made thereafter. The development aims for practical use from the beginning, and we believe our design theory is valid.

The author has engaged in research as a researcher and a consultant in a private company. There, research was required, and there were “partners” and “sites” with whom and at which problems had to be solved using the results of research. The partners and sites were never the same, the given conditions were diverse, and the clients’ demands could not be fulfilled if one single value was used to fulfill a given condition. Therefore, rather than one solution, the optimal solution that resulted from the expert’s experience and insight, or a mixture of senses and decisions, would be sought. What is important is to go to the site, experience, and understand, in order not to make mistakes in making decisions. In various fields, there is a sense of *déjà vu* at the destination reached. I think it is important in all fields to go to the site, to gain experience there, and to understand things.

Acknowledgements

I have obtained support from many researchers at the Forestry and Forest Products Research Institute (FFPRI). I have also received cooperation of the people of Tenryu, Shizuoka Prefecture for the demonstration experiments. Currently, I am continuing the research with funding from the Canon Foundation. I am grateful to all people who support this research and development.

Terminologies

Term 1. Large heavy forestry machine[*1]: These include the harvesting and forestation machines with high work performance and multiple functions, as described in the Basic Policy for the Promotion of High-Performance Forestry Machine that was announced by the MAFF in 1991.

Term 2. *Jigoshirae*[*1]: Site preparation work conducted prior to planting the saplings, so the saplings will take root in the forestation area.

Term 3. Complete enumeration[*1]: Basic survey method to clarify the forest composition, including the measurement of tree stand growth.

Term 4. Hanging tree: The tree that is cut may become caught by the surrounding standing trees. Such hanging trees are unpredictable as to when they will fall, and accidents occur in the maneuver to dislodge the hanging trees.

Term 5. Pair[*2]: When two or more joints engage in limited motion under a mutual bond, the joints are in “pairs.”

Term 6. Crawler: Caterpillar. A crawler has a belt with wider surface area that touches the ground compared to tires, and therefore has an advantage on weak or irregular ground. However, it is slower than tires.

Term 7. *Ukekuchi-oikuchi-giri* and *tsuru*:Undercutting and backcutting method is one of the methods of cutting down trees. In this method, first the *ukekuchi* (undercut) is made in the direction one wishes to fell the tree. The undercut consists of horizontal and diagonal cuts, and in Fig. 13, dimension A is 1/3 to 1/4 the diameter of the tree. Then, the *oikuchi* (backcut) is made from the opposite side, and dimension C of *tsuru* (hingewood) will be 1/10 of the diameter. The hingewood is the part that acts as a hinge when cutting down the tree. Normally, the backcut is placed horizontally. If the tree does not start to fall, a wedge is placed in the backcut and hit to make the tree fall. In this manipulator, the backcut is made diagonally rather than horizontally. This is to avoid interference of the hardware, and placing the cut diagonally will ensure that the tree will start falling on its own.

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Graduated from the Department of Architecture, Faculty of Science and Engineering, Waseda University; received Award of the Alumni Association of Waseda Architecture (Toumon Kenchiku Kai). Studied abroad at Bauhaus Universität, Germany. Completed the master's course at the Graduate School of Science and Engineering, Waseda University; received Sanae Award. Worked for the Nomura Research Institute, Ltd. Doctor (Engineering), Waseda University; Postdoctoral Fellow, Japan Society for the Promotion of Science; Researcher, Centre national de la recherche scientifique, France; and currently, Associate

Professor, Faculty of Science and Engineering, Waseda University. First class registered architect. Has authored *Shinrin No Hokai* (Collapse of Forest), Shincho Shinsho. Engages in research on the social infrastructure and its systems such as forest and water that are deeply intertwined with people's lives from a science and engineering perspective, and conducts technological development based on the findings. Wrote all parts of the paper.

Discussions with Reviewers

1 Overall

Comment (Naoto Kobayashi, Waseda University)

In this research, upon overviewing the current problems of Japanese forestry, an extremely ambitious and important goal is set, that is, to propose new technological development, as a way to solve the biggest problem that prevents forestry from becoming autonomous as an industry. The scenario to achieve the goal is built, and the specific method selected is to develop a tree-felling manipulator that is safe and easy to use. In this paper, the details of technological development are discussed, and at the same time, a new design method discovered in the process is introduced. The structure of the paper is clear and sound, and I believe this paper is suitable for publication in *Synthesiology*.

Comment (Yuki Imatomi, Tokyo University of Agriculture)

This is a commendable research paper on the R&D of new machines directed at ensuring safety that is a very important issue in forestry. In this paper, the goal is set as the revitalization of Japanese forestry as an industry, while considering the uniqueness of forestry in Japan, and the research elements for achieving the goals are presented. The research elements for achieving the goals and the positioning of the technological development elements can be understood well. Please consider reviewing the final draft so it can be easily understood by readers who are outside of this field.

2 Design theory

Comment (Naoto Kobayashi)

In this paper, the author states a unique design theory. Comparing it with the V model that was born in Germany, you state that your model starts from the right-top and goes to the right-bottom, that is totally different from the V model that starts from the left-top of V and ends at the right-top. The V model was born as a system design theory for software development, and it starts by clearly defining the requirements and specifications of the total system from the beginning. On the other hand, in a case where the environment in which the machine is used is extremely complex like the manipulator development in this research, the initial design spec may be unclear, and it must be changed along the way, even if it is defined at the beginning. Also, the author makes a comparison with the web design theory, and states that the theory of this paper is similar to that design theory. I think what is common is that the design spec of the system cannot be determined unless there are multiple interactions with the environment (including people). Even if the design spec is determined and a certain prototype development is assumed, further revisions will be necessary. Therefore, I think the process will be as follows for this design. First, there is a loop that determines the design spec, and one can determine the design spec by turning this loop around several times (TATSUMI I, II). Next, the system design and developments are done along a larger loop based on this decided design spec, and then the demonstration is done. I think this latter loop (TATSUMI III and others) is basically close to the V model. The design theory of this research is an integration of the "one design spec determination loop" and the "two system integration demonstration loop," and I understood that the former "design spec determination loop" is extremely important. Is my way of thinking in the right line?

Answer (Yuko Shirai)

I added a figure that shows the design spec determination loop and system integration loop in Subchapter 4.1, and also added a text explanation.

3 Need for using a portable tree-felling machine in the future

Comment (Yuki Imatomi)

Other than chainsaws, the forestry machines that can cut down trees currently include the fellerbuncher (that can cut down and gather timber) and the harvester (that has functions of cutting as well as log-making). In this research, you developed a tree-felling manipulator using the chainsaw. I think people's understanding will deepen if you clarify why people must continue using chainsaws or portable machines like your R&D machine in Japan in the future, by presenting data for the slope distribution in the forest area.

Answer (Yuko Shirai)

I added Section 2.4.2 "Forestry machines" and discussed the harvester and the fellerbuncher, and stated why it is necessary to continue using "portable machines" like the chainsaw or the R&D machines in Japan. Also, I presented the slope distribution map of Shizuoka Prefecture as an example.

4 Diffusion of the R&D machine

Comment (Yuki Imatomi)

In Japan, normally, the chainsaw is used for felling trees. Downsizing, weight reduction, and achievement of high performance have been done for the chainsaw as a portable machine, and this tool is essential in Japanese forestry. You are developing the tree-felling manipulator in this research from safety considerations, but I think the diffusion of a new machine is not easy because high-performance forestry equipment like the chainsaw already exists. I think you should mention the diffusion method for your machine as it nears completion.

Answer (Yuko Shirai)

I added the description about the prospects for diffusion in Subchapter 6.1 "Prospect for TATSUMI."

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
Revised August 5, 2010
Revised February 16, 2012
Revised April 17, 2013
Revised May 9, 2014
Revised April 1, 2015
Revised October 1, 2015

1 Types of articles submitted and their explanations

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

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

② Commentaries
Commentaries describe the thoughts, statements, or trends and analyses on how to utilize or spread the results of R&D to society. Although the originality of the statements is not required, the commentaries should not be the same or similar to any articles published in the past. The submitted manuscripts will be reviewed by the Editorial Board. The authors will be contacted if corrections or revisions are necessary, and the authors complete the final draft based on the Board members' comments.

③ Roundtable talks
Roundtable talks are articles of the discussions or interviews that are organized by the Editorial Board. The manuscripts are written from the transcripts of statements and discussions of the roundtable participants. Supplementary comments may be added after the roundtable talks, if necessary.

④ Readers' forums
The readers' forums include the readers' comments or thoughts on the articles published in *Synthesiology*, or articles containing information useful to the readers in line with the intent of the journal. The forum articles may be in free format, with 1,200 Japanese characters or less. The Editorial Board will decide whether the articles will be published.

2 Qualification of contributors

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

3 Manuscripts

3.1 General

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

research ethics

3.2 Structure

3.2.1 The manuscript should include a title (including subtitle), abstract, the name(s) of author(s), institution/contact, main text, and keywords (about 5 words).
3.2.2 Title, abstract, name of author(s), keywords, and institution/contact shall be provided in Japanese and English.
3.2.3 The manuscript shall be prepared using word processors or similar devices, and printed on A4-size portrait (vertical) sheets of paper. The length of the manuscript shall be, about 6 printed pages including figures, tables, and photographs.
3.2.4 Research papers 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, for subsections, 1.1.1.1, 1.1.1.2, 1.1.1.3.
3.3.2 The chapters, subchapters, and sections should be enumerated. There should be one line space before each paragraph.
3.3.3 Figures, tables, and photographs should be enumerated. They should each have a title and an explanation (about 20–40 Japanese characters or 10–20 English words), and their positions in the text should be clearly indicated.
3.3.4 For figures, image files (resolution 350 dpi or higher) should be submitted. In principle, the final print will be in black and white.
3.3.5 For photographs, image files (resolution 350 dpi or higher) should be submitted. In principle, the final print will be in black and white.
3.3.6 References should be listed in order of citation in the main text.

Journal—[No.] Author(s): Title of article, Title of journal (italic), Volume (Issue), Starting page–Ending page (Year of publication).

Book—[No.] Author(s): Title of book (italic), Starting page–Ending page, Publisher, Place of Publication (Year of publication).

Website—[No.] Author(s) name (updating year): Title of a web page, Name of a website (The name of a website is possible to be omitted when it is the same as an author name), URL, Access date.

4 Submission

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

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

5 Proofreading

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

6 Responsibility

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

7 Copyright

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

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Vol.9 table of contents (2016)

Vol.9 No.1

Research papers

Developing a leading practical application for 3D IC chip stacking technology
— *How to progress from fundamental technology to application technology* —
- - - M. Aoyagi, F. Imura, F. Kato, K. Kikuchi, N. Watanabe, M. Suzuki, H. Nakagawa, Y. Okada, T. Yokoshima, Y. Yamaji, S. Nemoto, T. T. Bui and S. Melamed 1 - 15

Securing a stable supply of critical raw metals
— *Efforts and issues for the securement of rare-earth resources* —
- - - T. Takagi 16 - 26

Scenario in synthetic-type research: its role and description
— *An investigation from Synthesiology papers* —
- - - A. Ono, M. Akamatsu and N. Kobayashi 27 - 41

Article

Consortium style study on the development of highly reliable photovoltaic modules and acceleration test methods
— *Management of the "Consortium Study on Fabrication and Characterization of Solar Cell Modules with Long Life and High Reliability"* —
- - - A. Masuda and N. Igawa 42 - 54

Vol.9 No.2
Special Issue of Geological Surveys

Research papers

Development and utilization of geochemical reference materials
— *Reliability improvement in the analysis of geological materials* —
- - - T. Okai 60 - 73

Three-dimensional urban geological map
— *New style of geoinformation in an urban area* —
- - - T. Nakazawa, S. Nonogaki and Y. Miyachi 74 - 86

Constructing a system to explore shallow velocity structures using a miniature microtremor array
— *Accumulating and utilizing large microtremor datasets* —
- - - I. Cho and S. Senna 87 - 98

Development of rock deformation techniques under high-pressure and high-temperature conditions
— *Evaluation of long-term geological processes by a compressed timescale process model* —
- - - K. Masuda 99 - 111

Vol.9 No.3
Special Issue of Nanomaterials

Research papers

Development of human-friendly polymeric actuators based on nano-carbon electrodes
— *Toward the practical realization of artificial muscles* —
- - - K. Asaka 117 - 123

High quality and large-area graphene synthesis with a high growth rate using plasma-enhanced CVD
— *Toward a high throughput process* —
- - - M. Hasegawa, K. Tsugawa, R. Kato, Y. Koga, M. Ishihara, T. Yamada and Y. Okigawa 124 - 138

Radioactive cesium decontamination technology for ash
— *Utilization and application of nanoparticles as an adsorbent* —
- - - T. Kawamoto, H. Tanaka, Y. Hakuta, A. Takahashi, D. Parajuli,
K. Minami, T. Yasutaka and T. Uchida 139 - 154

Development of HASClay® as a high-performance adsorption material
— *Developing adsorbents for energy conservation systems from a kind of clay nanoparticle* —
- - - M. Suzuki, M. Maeda and K. Inukai 155 - 166

A super-growth method for single-walled carbon nanotube synthesis
— *Development of a mass production technique for industrial application* —
- - - K. Hata 167 - 179

Vol.9 No.4

Research papers

Development of a framework for risk tradeoff analysis of chemical substance substitution
— *An approach to risk assessment using relative comparison* —
- - - M. Gamo and J. Takeshita 185 - 196

A first empirical analysis of JIS lifespan
— *Implications for the review system of de jure standards* —
- - - S. Tamura 197 - 212

Development of EUPS for analyzing electronic states of topmost atomic layer
— *Materialization of laser-produced plasma source application and EUPS observed fascinating surface* —
- - - T. Tomie and T.Ishitsuka 213 - 233

A revolutionary technical development to revitalize Japanese forestry
— *A proposal for a portable tree felling manipulator to address specific properties of Japanese forestry* —
- - - Y. Shirai 234 - 251

Letter from the editor

This is the last issue of Volume 9. There are four papers published in this issue two of which were submitted from outside AIST, and I think this is greatly appreciated from the point of view of how widely *Synthesiology* is known.

The common theme of this issue is that all the papers are “highly useful.” Particularly, the first paper, “Development of a framework for risk tradeoff analysis of chemical substance substitution,” is about deriving a method for quantitatively comparing the effects of different risks, applying it to actual regulatory cases, and then validating the usefulness of the method. The method is not limited to chemical substances, and can be applied to other substances, pharmaceutical products, and medical devices, and its further development is highly anticipated.

The second paper, “A first empirical analysis of JIS lifespan,” focuses on the JIS standards that were not taken up as academic subjects until now, and discusses the appropriate timing for review and the improvement of efficiency so the standards will be useful for innovation. The characteristics of the standards are evaluated by a common statistical method from various data, and this method is beneficial for the discussion on reviewing the JIS standards in the future.

While the aforementioned two papers and the last paper are about research with clear objectives or demands, “Development of EUPS for analyzing electronic states of topmost atomic layer” is a paper on the development of a prospective research “seed (potential).” It studies various cases to which the technology can be applied, and clarifies the diversity and usefulness of the technology. It is a “seeds-driven” research. The method developed in this research has the advantage that it enables observation of the behavior of substances of the topmost atom layer that could not be achieved by other methods, and future development is expected. In this case, further acquisition of users is essential.

The final paper, “A revolutionary technical development to revitalize Japanese forestry,” is a technological development in the field of forestry, which is rarely taken up in *Synthesiology*, and contains several extremely interesting contents such as the research objective, scenario, and the design theory to achieve the goal. The author explains that for the Japanese forestry to become an autonomous industry, energy conservation and increased efficiency through safe, automated tree felling in severe forest environment are mandatory, and the result presented in the paper is a technology that is expected to offer a solution. It is hoped that this technology becomes practical soon.

Although I said that the research results are highly useful, large hurdles must be overcome for innovation to happen. Needless to say, the hurdles are not necessarily technological. I wish the research mentioned here will reach out further and lead to future innovation.

(Naoto KOBAYASHI, Executive Editor)

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

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

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

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Synthesiology-English edition 4 2016

Highlights of the Papers in *Synthesiology*

Research papers

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—Materialization of laser-produced plasma source application and EUPS observed fascinating surface—
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Y.Shirai

Editorial policy
Instructions for authors
Letter from the editor
Aim of *Synthesiology*

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