A social system for production and utilization of thermophysical quantity data

— Measurement technology, metrological standard, standardization of measurement method, and database for thermal diffusivity by laser flash method —

Tetsuya BABA * and Megumi AKOSHIMA

[Translation from *Synthesiology*, Vol.7, No.1, p.1-15 (2014)]

The National Metrology Institute of Japan designed a system to supply reliable thermal diffusivity data efficiently and quickly to society. The system was founded on the technology for thermal diffusivity measurement by using a laser flash method, by establishing a metrological standard and reference materials, and by standardization of measurement technology. Uncertainty in measurement of thermal diffusivity with practical apparatus by the laser flash method was reduced using elemental technologies to homogenize the laser beam, a fast response infrared thermometer, and a curve fitting method to analyze temperature response curves. JIS and ISO standards were established or revised based on the developed technologies. In addition, methods to evaluate uncertainty in measurement of thermal diffusivity and calibration procedure by reference materials are described in the latest update of the JIS standard for the laser flash method. A procedure to obtain intrinsic thermal diffusivity is also described. Traceable thermophysical quantity data produced by this social system can be accumulated in a database system developed and operated by the National Metrology Institute of Japan (NMIJ), which is accessible via the Internet.

Keywords : Thermophysical quantity data, thermal diffusivity, laser flash method, metrological standard, reference material, standardization of measurement method, database, traceability, uncertainty, intellectual infrastructure

1 Introduction

In advanced devices, equipment, or structures, it is often pointed out that their thermal properties are bottlenecks to their performance and reliability. For example, in order to have a highly integrated electronic device to function fully, a cooling mechanism that efficiently removes the massive amount of heat produced by multiple elements that are packed in a limited space is necessary. For space vehicles entering the atmosphere to withstand severe aerodynamic heating, a special material that possesses a heat insulation function under ultra high temperature is necessary for its exterior. When analyzing a severe accident at a nuclear power plant, it is necessary to accurately simulate the temperature behavior of the nuclear fuel and the core that surrounds it, all the way to ultra high temperature. It is necessary to improve the efficiency of thermal energy use of the whole society in order to reduce fossil fuel consumption and carbon dioxide gas emissions, and therefore, it is necessary to choose the appropriate materials with excellent insulation, conductance, and heat storage functions.

For a device, instrument, or structure to operate with full function safely and effectively, a highly reliable thermal design should be made in advance. In a thermal design, accurate thermophysical quantity data must be known for all related materials and components. However in reality, one often faces problems when trying to obtain such data from available information.[1]-[3] When there was need for thermophysical quantity data, it was a common practice for the researcher to look through data books or databases for the material that was the subject of interest.[4] However, the required thermophysical quantity data often could not be found. Even if some data was found, it was unclear whether the data could be assigned to the exact material of interest. This situation is not so much a problem if one simply needed the thermophysical quantity data as a rough guideline, but such data were insufficient for realizing the maximum function or guaranteeing safety, and the researcher him/herself had to measure the thermophysical quantities or subcontract such measurement to a specialized lab. If measurement is necessary, the next problem is how to choose the appropriate measurement method and how to evaluate reliability of thermophysical quantity data measured by the method. However, there is no such information or guideline yet based on agreement among the academic and technological community.[1][2]

While various advanced materials are being developed in the fields of electronics, precision optics, environment and energy, aerospace, and nuclear technologies, it is not easy to obtain highly reliable thermophysical quantity data of the materials. In order to solve this situation, the National Institute of Advanced Industrial Science and Technology (AIST) has been conducting research on the thermophysical quantity measurement and reference materials for the past 30 years. In setting the research scenario, the greatest

National Metrology Institute of Japan, AIST Tsukuba Central 3, 1-1-1 Umezono, Tsukuba 305-8563, Japan
＊ E-mail : t.baba@aist.go.jp

Original manuscript received January 10, 2012, Revisions received August 16, 2013, Accepted August 22, 2013

focus was placed on how AIST could play a public role in society. In other words, the production of highly precise thermophysical quantity data at AIST that was responsible for metrological standards should not be the endpoint, but the final goal of the research was to envision and construct the technological infrastructure, in which specialists of not only Japan but also of the world could efficiently produce highly reliable thermophysical quantity data, and these data could be effectively utilized in society. This can be called a "social system for efficient production and effective use of thermophysical quantity data."[3]

To achieve this goal, AIST engaged in various research activities on thermophysical quantities. In this paper, we shall focus on the thermal diffusivity of solid materials, and describe the research scenario of precise measurement technology, metrological standards, reference materials, standardization of practical measurement methods, and research results. AIST also conducted R&D for the thermophysical quantity database, but due to limitations of space in this paper, we shall only list them in the references.

2 Measurement and data of thermophysical quantities

2.1 Definition of thermophysical quantities

Thermophysical quantity values are numerical expressions for thermophysical properties of materials and substances. The functions pertaining to the transfer and storage of heat energy such as insulation, thermal conductance, or thermal storage are expressed as thermophysical quantity values such as thermal conductivity, specific heat capacity, thermal diffusivity, and thermal effusivity. These thermophysical quantity values are defined as follows.[4]

Thermal conductivity (λ): The phenomenon in which heat is transferred by conduction in a material is expressed as thermal conductivity, and is defined as the ratio of the heat flow density that passes through the material to the temperature gradient. The unit is $Wm^{-1}K^{-1}$.

Specific heat capacity (c): The amount of heat necessary to raise the temperature of a material 1 K per unit mass. The unit is $Jkg^{-1}K^{-1}$.

Thermal diffusivity (α): This is defined by the following equation based on thermal conductivity λ, specific heat capacity c, and density ρ.

$$\alpha = \lambda / (c\rho) \quad \dots (1)$$

The unit of thermal diffusivity is m^2s^{-1}. When part of a material insulated from the environment is heated in a short time to raise the temperature of that part only, the heat spreads throughout the material in time, and homogenous temperature is attained eventually. The speed at which the temperature converges to the final homogeneous temperature is proportional to the thermal diffusivity.

When it is difficult to directly measure the thermal conductivity λ, thermal conductivity measurement can be substituted by separate measurements of thermal diffusivity α, specific heat capacity c, and density ρ, and the thermal conductivity can be calculated from Equation (1).

Thermal effusivity (b): This is defined by the following equation calculated from thermal conductivity λ, specific heat capacity c, and density ρ.

$$b = \sqrt{\lambda c\rho} \quad \dots (2)$$

The unit of thermal effusivity is $Jm^{-2}s^{-1/2}K^{-1}$. It can be intuitively explained as the capability of the material of a sufficiently thick object to absorb heat when the surface is heated uniformly. Materials such as iron that has large thermal conductivity and density have large thermal effusivity, and the temperature rise of the heated surface is small. Inversely, insulating materials with low thermal conductivity and low density has small thermal effusivity, and therefore the temperature rise of the heated surface is large.

The four values, thermal conductivity λ, volume heat capacity C (= specific heat capacity × density), thermal diffusivity α, and thermal effusivity b are not mutually independent. If the arbitrary two quantities are known, the remaining two quantities are determined. Since these thermophysical quantity values are temperature dependent, they must be expressed as functions of temperature.

While the electric/electronic, mechanical, and optical quantities can be relatively easily measured in accordance with a defined condition, it is not easy to accurately measure thermophysical quantities according to the definitions explained above. The main reason is the difficulty of complete insulation and of accurately controlling heat flow in contrast to the situation of electricity which can be almost perfectly insulated by electrically insulating materials. Even in vacuum, heat energy can be transferred as thermal radiation. That is the reason why measurement methods for thermophysical quantities are still actively developed and improved, incorporating new technologies.

2.2 Traceability of measurement and reference materials

The National Bureau of Standards (NBS) developed reference materials[5] and organized standard data[6] in a wide range of fields including thermophysical quantities, in order to fulfill a national demand for achieving measurement traceability in the 1960s. NBS was reorganized into the National Institute of Standards and Technology (NIST) in 1988. NIST gradually lost its ability to develop new reference materials in the thermophysical quantity field, and is currently unable to replenish the out-of-stock reference materials of thermophysical quantities.

The development of thermophysical quantity reference materials was also done in Europe. For solid materials, the Institute for Reference Materials and Measurements (IRMM) started providing three types of reference materials for thermal conductivity and thermal diffusivity in the 1990s.[7]

In Japan, there is a well-organized traceability system, or a social system in which measurement results taken at any institution are guaranteed within a certain range of uncertainty, as long as the instruments used are traceable to the national standards for basic quantities such as length, mass, time, electricity, or temperature. One example of such a system is the Japan Calibration Service System (JCSS).[8] Although JCSS was established for the thermal conductivity measurement of insulation materials by contribution of the Japan Testing Center for Construction Materials (JTCCM), a traceability system for thermal diffusivity measurement or thermal conductivity measurement of dense materials has been established using the reference materials developed by AIST instead of JCSS.[2][3][9]

Efforts for “a social system for production and utilization of thermophysical quantity data” by the academic community in Japan, Comité International des Poids et Mesures (CIPM), and the former National Research Laboratory of Metrology (NRLM), Agency of Industrial Science and Technology, Ministry of International Trade and Industry are described in the appendices.

2.3 History of accumulation, evaluation and publication of thermophysical quantity data

There is a long history in Europe and the United States for collecting thermophysical quantity data of substances and materials, and then providing them systematically to a wide range of users. Germany has continuously worked on collecting and evaluating physical quantity data including thermophysical quantities since the end of the 19th century, and the results were compiled and published, for example, as the Landolt-Börnstein Data book. Today, the data is available online as Springer Materials (Landolt-Börnstein Database).[10]

In the United States, compilation of the international critical tables was started under the auspices of NBS in the 1920s.[11] The US Congress established the “standard reference data act” in 1968, and the US government decided to start the data evaluation program named the National Standard Reference Data System (NSRDS).[12] As a part of this program, the Thermophysical Property Research Center (TPRC) at Purdue University collected and evaluated a vast amount of thermophysical quantity data, and published a 14-volume thermophysical property data book, the TPRC Data Series.[13]

This data series collected the thermophysical quantity data of a wide range of materials and substances, mainly for practical materials in addition to basic substances such as elements. For some data, the reliability was evaluated and recommended values were provided. However, the collected data included those obtained by measurement methods that were not well established. In some cases, the measurement apparatus were not correctly calibrated and the information to identify the measured substances and materials (characterization information) was limited.

The TPRC was reorganized into and the activities continued at the Center for Information and Numerical Data Analysis and Synthesis (CINDAS) at Purdue University over the 1980s. Currently, the data has been succeeded by the CINDAS Limited Liability Company, and is available online as a database.[14]

In the field of thermophysical quantities for solid materials, the notable accomplishments of NSRDS were the thermophysical quantity reference materials of NBS and the Purdue University TPRC database, but the cooperation between the two activities was not so effective. It was not common practice for apparatus of thermophysical quantity measurements to be calibrated or verified by the reference materials provided by NBS when TPRC data series were published.

Although the demands for thermophysical quantity data for electronics, environment and energy, and life science materials increased in recent decades, there has been no compilation or publication of a comprehensive data book comparable to the TPRC Data Series to the present.

2.4 Reliability of the thermophysical quantity data and the issues of measurement technology

Even for measurement methods that were used popularly to obtain thermophysical quantity data, there were examples in which handling of the measuring apparatus was too complicated, the reliability (precision and accuracy) of the measurement was not enough, or the evaluation of reliability was not easy. There were fairly large variations in the thermophysical quantity data obtained for the same specimen even based on the same measurement method, and the variations increased further when the measurement methods differed.[3][15][16]

It would be useful if thermophysical quantity data became universal information generalized from an individual measurement apparatus or a laboratory. To realize such a situation, the triangle set of tasks was planned, including the establishment of national standards and reference materials, standardization of the measurement methods, procedures of measurement, and specifications of apparatus (JIS and ISO standards), and development of practical measurement apparatus that fits the standard.[17]

3 Scenario setting

In the initial stages of research, we envisioned a social system where engineers and researchers could easily and speedily access thermophysical quantity data with sufficiently small uncertainty. Hence, we set the research scenario as shown in Fig. 1.[3][15]

In the scenario of Fig. 1, first, improvement of precision and accuracy for the measurement technology of corresponding thermophysical quantities and the establishment of national standards and reference materials based on this technology were set. Second, the conditions required for reference materials were specified that they should be homogenous and their properties should not change over a long period of time. Then, the reference values of the reference materials should be determined by the national standard, the uncertainty of reference values should be evaluated, and the related information should be provided to society.

Third, the standardization of measurement technology or the organization of standards was planned. If the measurement methods and procedures for thermophysical quantities were standardized by JIS or ISO, it would be possible to evaluate measurement uncertainty as well as to realize thermophysical quantity measurement traceable to the national standard, by calibrating/validating the measurement apparatus using the aforementioned reference materials according to the document standard such as JIS or ISO.

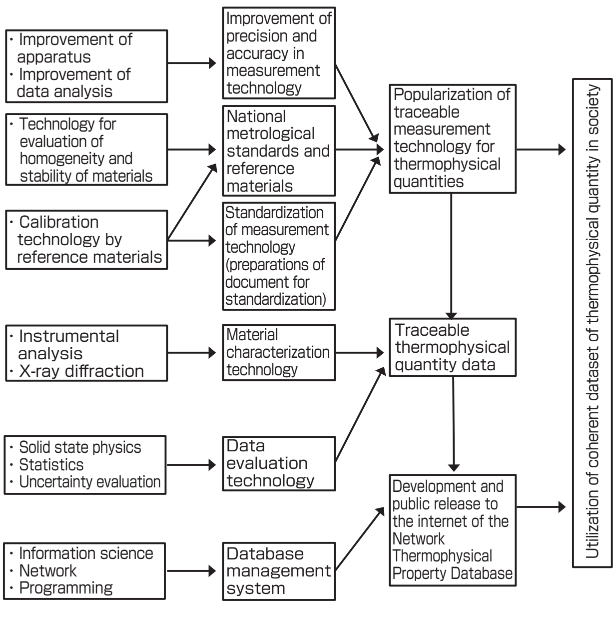

Fig. 1 Scenario for constructing a social system for the production and use of thermophysical quantity data

Fourth, the material characterization technology was addressed. Physical properties of a material are universal information applicable to materials with the same characteristics (composition, structure, etc.) at the same conditions (temperature, pressure, etc.), in contrast to the measurement data for physical variables such as temperature and pressure which are only applicable to each measurement. However, contrary to a physical quantity of a liquid which is uniquely determined by composition, a thermophysical quantity value of a solid may change depending on characters, even if the material has the same composition. Particularly, the transfer quantities such as thermal conductivity and thermal diffusivity are sensitively dependent on the characters such as crystal structures and grain boundaries.

Fifth, if the measured thermophysical quantity data could be successfully assigned to the information of characters such as compositions and structures of materials, the combination of measurement values of thermophysical quantities and material character could be called a “Coherent Dataset.” which would be widely useful in society.

Moreover, if data evaluation technology and data management system would be implemented to the database, the thermophysical quantity data with high reliability and universal value would be produced continuously and systematically, according to the scenario shown in Fig. 2.

In the field of physical measurement of basic quantities such as length, mass, time, electricity, and temperature, the common practice is to maintain traceability of the measured value by conducting calibration using a higher-level standard by transporting the standard artifact. In the field of chemical analysis, the traceability of measurement value is maintained by transporting the reference material.

There is a special approach specific to the field of material measurement including thermophysical quantities where traceability can be proved by presenting the set of information for “standard data” and “material character”

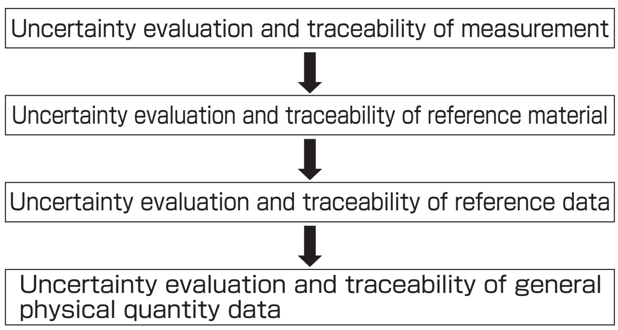

Fig. 2 Process of generalizing concept of uncertainty evaluation and traceability from “measurement of physical quantities” to “physical quantity data”

(that is, it is not necessary to transport any artifact or material), in addition to the conventional traceability system of transporting the standard artifact or reference materials.[16][18]

In this scenario, the concept of uncertainty evaluation and traceability was consecutively expanded from measurement to reference materials, to standard data, and then to general physical quantity data. Eventually, the reliability of the general thermophysical quantity data would be proved. In order to realize this process, new R&D that allows quantitative expression of material character and physical quantity value is necessary as shown in Fig. 2.

4 Measurement of thermal diffusivity by a laser flash method

4.1 Selection of the laser flash method

Since thermal conductivity and thermal diffusivity are proportional to each other as indicated by Equation (1) when specific heat capacity and density are fixed, measurements of thermal conductivity and thermal diffusivity are complementary to each other and each can be chosen to be measured according to the situation.

Thermal conductivity is generally measured by a steady-state method. In the steady method, a certain amount of constant heat flow passes through a specimen, and the thermal conductivity is calculated by measuring the temperature gradient that occurs in the specimen. On the other hand, thermal diffusivity is generally measured by a transient method. In the transient method, the thermal diffusivity is calculated by measuring the relaxation time where the temperature distribution becomes homogenous over time in a specimen in which the temperature was initially inhomogeneous.

In thermal conductivity measurement by a steady-state method, it is necessary to measure the absolute value of the temperature by attaching thermometers to the specimen. Therefore, this method is suitable for materials where a specimen of a large size can be obtained. Since the heat source, heat sink, and thermometers should be in contact with the specimen, various conditions must be considered such as selecting the materials for heat source and heat sink, selection of the thermometers, and correction for radiation heat loss, particularly in conducting the measurement at high temperature.

For measurement technology of thermal diffusivity by a transient method, a laser flash method had already been developed in advance before the start of our research. It was widely used as a standard method for measuring thermal diffusivity of dense solid materials such as metals, alloys, ceramics, semiconductors, and graphite.[19]-[22] Reflecting such wide-spread use, most of the thermal diffusivity values listed in product catalogs and those in the data books and database for dense solid materials have been measured using the laser flash method.

The measurement principle of the laser flash method is shown in Fig. 3. With this method, the front face of a planar specimen that has been maintained at steady temperature is pulsewise heated. Then, the heat diffuses one-dimensionally from the front face of this specimen to the inside of the specimen, and finally, the entire specimen reaches homogeneous temperature.[19]-[22] The thermal diffusivity of the material is calculated from the rate of temperature rise at the rear face of the specimen.

As a measurement method of thermal diffusivity, other than pulse heating, cyclical heating methods and step-function heating methods have been developed. Thermal effusivity can also be calculated from the temperature change at the location where the specimen surface is heated.[4]

Advantages that the laser flash method possesses are as follows :

- Quality of the measurement can be evaluated by fitting the analytically calculated theoretical curve to the temperature response curve at the rear face of the specimen under one-dimensional heat diffusion after pulse heating.
- Typical shape and size of a specimen is a disk of 10 mm in diameter and 1 -2 mm in thickness and one measurement can be completed within a few seconds.
- Impulse heating and measurement can be done without contact by using a pulsed laser and a radiation detector for measuring the temperature changes, respectively.
- Measurement can be made at a wide temperature range from lower than room temperature to higher than 2,000 °C.
- Since the method using a flash lamp was invented in 1961, many researchers have contributed to improving the method, and its validity, reliability and superiority are commonly recognized. As a result, practical measurement apparatus are widely used.

From the above background, we decided to start research of the laser flash method in order to establish the national

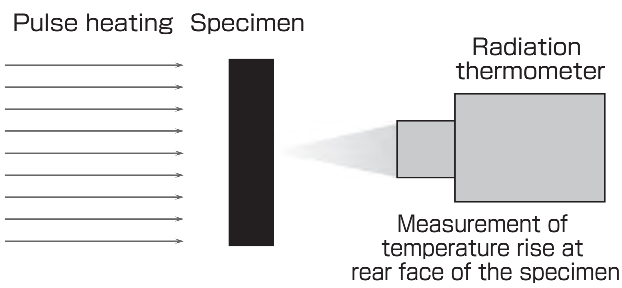

Fig. 3 Measurement principle of the laser flash method

standard for thermal diffusivity measurement since it had potential for achieving precise and accurate thermal diffusivity measurement. On the other hand, we noticed that the laser flash method had not matured enough for the national metrological standard of thermal diffusivity, and the uncertainty evaluation of measurement results had not been done sufficiently when we decided to start the research of the laser flash method in 1983. Therefore, the first priority was set to improve the measurement technology. In order to provide highly reliable thermal diffusivity data to society efficiently and quickly, development of the reference materials and standardization of the measurement technology (organization of the JIS and ISO standards) were also set as research topics.

4.2 Improvement of measurement technology

Figure 4 shows a non-dimensional temperature rise (temperature rise normalized by the maximum temperature rise) at the rear face of a specimen under the ideal initial conditions and boundary conditions. It is expressed as the function of non-dimensional time (time normalized by the characteristic time $\tau = d^2/\alpha$ of heat diffusion across the thickness). The rate of temperature rise at the rear face of the specimen is proportional to the thermal diffusivity α, and is inversely proportional to the square of the specimen thickness. In ideal conditions, the thermal diffusivity can be calculated from the specimen thickness d and the half rise time $t_{1/2}$ required to reach half of the maximum temperature rise $\Delta T/2$, by the following equation.[19][20]

$$\alpha = \frac{0.1388\, d^2}{t_{1/2}} \qquad \ldots (3)$$

Figure 5 shows the time change of the temperature at the rear face of the specimen observed for glass like carbon by the laser flash method.

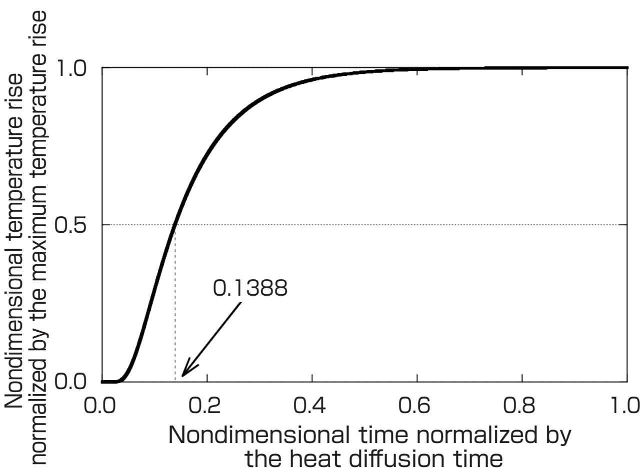

Fig. 4 Temperature response curve at rear face of the specimen by the laser flash method under ideal initial and boundary conditions

AIST developed several new elemental technologies of the laser flash method. First, the spatial energy distribution of the laser beam was homogenized to realize one-dimensional heat flow within the specimen by passing the pulse laser beam through optical fibers.[22][23] Second, the temperature change at the rear face of the specimen was accurately measured by using the high speed infrared radiation thermometer.[22][24] Third, the thermal diffusivity was calculated considering the heat loss from the specimen surface to the environment through radiation.[20][21] Fourth, the intrinsic thermal diffusivity unique to the material was calculated by extrapolation when the heating laser energy was zero (Fig. 6).[22][25]-[27]

Figure 7 shows the photograph of the national standard apparatus of the laser flash method.

4.3 Development of the national standard and reference materials

The National Metrology Institute of Japan (NMIJ), AIST accomplished the development plan of the national

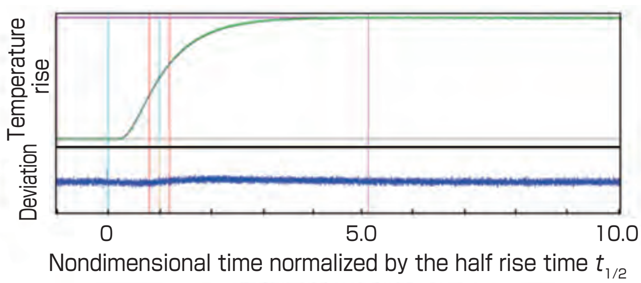

Fig. 5 Temperature response curve (green) at rear face of the specimen of glass like carbon measured by the laser flash method, ideal curve (black), and the deviation between the two curves (shown in blue at bottom part of graph, magnified 10 times)

The measured curve (green) and ideal curve (black) are agree with each other and systematic deviation could not be observed.

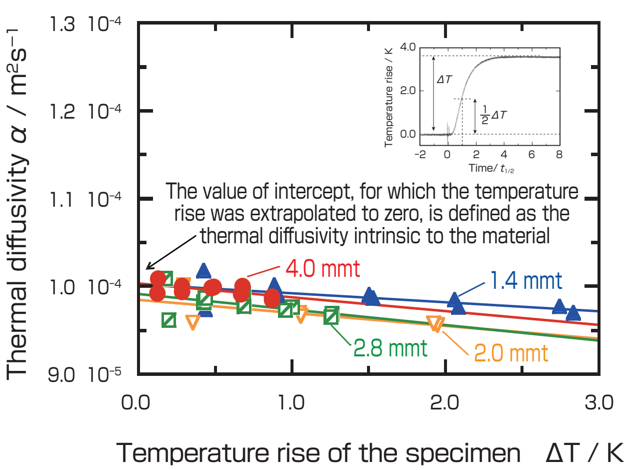

Fig. 6 Data plot to calculate the thermal diffusivity intrinsic to the material (example where the high-density isotropic graphite is measured at room temperature)[26]

Table 1. Items for which standards are provided in the solid thermophysical quantity field based on the national metrological standard development plan (~2010)

Development plan Number	Quantity (item of calibration service)	Range (range of calibration temperature for service)	Extended (relative) uncertainty of coverage factor k=2	Type of service	Remarks
240-00 241-00	Thermal diffusivity	1×10^{-6}m^{2}s^{-1}~5×10^{-4}m^{2}s^{-1} (297 K~1500 K)	< 3.4 %	Calibration service (general)	Calibration specified for isotropic graphite
		(300 K~1500 K)	5 %~7 %	Certified reference material NMIJ CRM5804-a	Isotropic graphite supplied as NMIJ RM 1201-a before 2010
242-00	Thermophysical quantities of thin films (heat diffusion time across thickness of the specimen)	100 ps~6500 ps	4.2 % (Mo thin film of 400 nm thick)	Calibration service	Thin film of from 100 nm to 400 nm thick
242-10		40 ns~1000 ns	3.6 %	Calibration service	Calibration specified for titanium nitride thin film on transparent glass substrate
		150 ns (Room temperature)	4.9 %	Certified reference material NMIJ RM1301-a	Titanium nitride thin film of 700 nm thick on transparent glass substrate
244-00	Thermal conductivity	(300 K~900 K)	7.4 %~9.8 %	Certified reference material NMIJ RM1401-a	Isotropic graphite
245-00	Specific heat capacity	(50 K~350 K)	1.3×10^{-3}JK^{-1}g^{-1} ~6.1×10^{-3}JK^{-1}g^{-1}	Calibration service	Adiabatic calorimetry
246-00		(300 K~900 K)	1.9×10^{-2}JK^{-1}g^{-1} ~3.1×10^{-2}JK^{-1}g^{-1}	Calibration service	Differential scanning calorimetry

metrological standards, with the goal of establishing 250 physical metrological standards and 250 chemical metrological standards by 2010.[28] The thermophysical property standard section, physical property statistics division (currently material physical property division), NMIJ AIST was in charge of the solid thermophysical quantity field and has established the metrological standards for thermal diffusivity, thermal conductivity, and specific heat capacity of the bulk materials, as shown in Table 1. Also, reference materials for thermal conductivity, thermal diffusivity, and specific heat capacity have been developed and supplied.[9][29][30] Responding to the needs from advanced industry, AIST has also engaged in the development of metrological standards for thermal diffusivity and heat diffusion time of thin films, and started measurement service and supply of a reference material.[31][32]

Fig. 7 The apparatus of national standard for thermal diffusivity

NMIJ established the SI traceable thermal diffusivity measurements and completed uncertainty evaluation according to GUM. A quality management system based on ISO 17025,[33] the international requirement for calibration laboratories, has been established and is under operation at NMIJ. The calibration service of thermal diffusivity started in 2004. High-density isotropic graphite was selected as the distributed thermal diffusivity reference material because of its homogeneity, stability, and black surface without a coating process.[34][35] Its reference values were determined using the national standard of thermal diffusivity, and distribution was started in 2006.The quality management system of thermophysical quantity reference material is operated in accordance with ISO Guide 34[36] that is an international requirement for manufacturers of reference materials since FY 2010. It has been provided as a certified reference material called NMIJ CRM-5804 since 2010.

As the next step after the establishment of the national standard, it is important to verify whether the Japanese and overseas national standards for thermophysical quantity measurement are equivalent.[37] To do so, the homogenous specimens cut from the same lot are distributed to several national metrology institutes (NMIs), and the measurement values at each NMI are compared to see whether they agree within the range of uncertainty. This is called an international comparison of national metrological standards. The NMIs with which the measurement results agreed mutually recognize their calibration and measurement capabilities (CMCs), and the results are entered into the BIPM Key Comparison Database (KCDB)[38] that can be viewed by everyone.

International comparisons for thermophysical quantities have been conducted by the Working Group 9 (WG9) for thermophysical quantities of the CIPM Consultative Committee for Thermal Metrology (CIPM CCT)[39] and the Technical Committee for Thermometry (TCT) of the Asia Pacific Metrology Programme (APMP).[40][41]

The pilot study of thermal diffusivity measurements of solid materials by the laser flash method was conducted from 2008 to 2011.[42][43] This was the first international comparison of thermal diffusivity measurement by the laser flash method among the NMIs, and the objective was to survey the level of the latest measurement technology, as well as to obtain common understanding of the measurement procedure, data analysis, and the uncertainty evaluation method. One of the authors (Akoshima) was the organizer of this pilot study, and four NMIs participated, including the Laboratoire National de Metrologie et d'Essais (LNE) of France, the National Institute of Metrology (NIM) of China, the NMIJ AIST of Japan, and the National Physical Laboratory (NPL) of the UK.

Armco iron and high-density isotropic graphite were selected because they were readily available on the market, and have been confirmed to be stable based on sufficient evaluations in the past. Each set of disk specimens of Armco iron and high-density isotropic graphite with a diameter of 10 mm and thickness of 1.0, 1.4, 2.0, 2.8, and 4.0 mm were prepared for each participating NMIs. After checking the homogeneity, one set each was sent to the participating NMIs for measurement.[43] For the measurement procedure, a method for obtaining intrinsic thermal diffusivity that was proposed by AIST was used,. The results reported by all NMIs are shown in Fig. 8 and Fig. 9. The figures show a set of data by an NMI systematically deviates from those of the other NMIs. This systematic deviation was attributed to slow response of the detector for the rear face temperature change used by the apparatus. After excluding this set of data, the other data agreed within the uncertainty range, where the standard deviation of variation among the three NMIs were 7 % or less in the temperature range from 300 K to 1200 K. From this international comparison, it was confirmed that the thermal diffusivity measurement by the laser flash method had sufficient equivalency among the NMIs. The result of the comparison has been published already.[42][43] The pilot comparisons of CCT-WG9 were registered as supplementary comparisons of key comparison database of BIPM, KCDB, in 2012.

At the meeting of CCT in 2010, an agreement was reached to incorporate thermal conductivity, thermal diffusivity, specific heat capacity, and thermal expansion of the thermophysical quantity field into the service category of thermal metrology registered in the Key Comparison Database (KCDB). Protocol for reviewing the CMC for thermal diffusivity has been submitted to the Working Group 8 of CCT.

In APMP, the Regional Metrological Organization (RMO) of which members are NMIs in the Asia-Pacific region, a working group for thermophysical properties was established in the Technical Committee covering Thermal Measurement (TCT) in 2010. In conjunction with the activities of CIPM CCT, two supplementary comparisons were organized in the Asia-Pacific region. Specifically, they were thermal diffusivity measurement by the flash method (APMP T.S-9) and thermal conductivity measurement of insulating

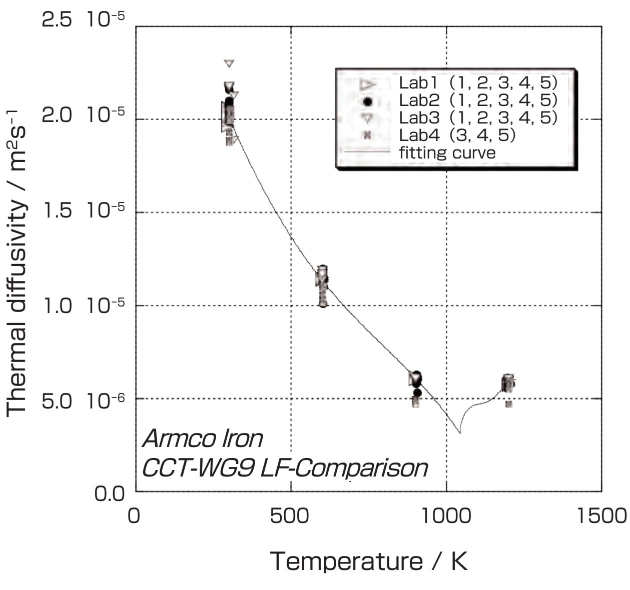

Fig. 8 Result of the pilot study for international comparison of the thermal diffusivity of Armco iron[43]

The numbers in parentheses [e.g. Lab1 (1, 2, 3, 4, 5)] indicate the specimen identification numbers.

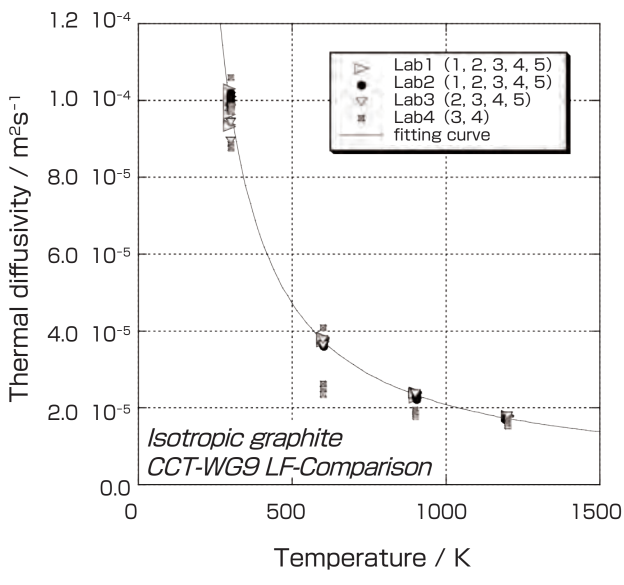

Fig. 9 Result of the pilot study for international comparison of the thermal diffusivity of high-density isotropic graphite[43]

The numbers in parentheses [e.g. Lab1 (1, 2, 3, 4, 5)] indicate the specimen identification numbers.

Table 2. ISO and JIS standards related to the laser flash method to which this study contributed

Category	Standard identification number.	Issued year	Japanese title	English title
ISO	18755	2005	—	Fine ceramics: Determination of thermal diffusivity of monolithic ceramics by laser flash method
JIS	R1611	1991 (Revised 2010)	ファインセラミックスのレーザフラッシュ法による熱拡散率・比熱容量・熱伝導率試験方法	Test methods of thermal diffusivity, specific heat capacity, and thermal conductivity for fine ceramics by flash method
JIS	H7801	2005	金属のレーザフラッシュ法による熱拡散率の測定方法	Method for measuring thermal diffusivity of metals by the laser flash method
JIS	R1667	2005	長繊維強化セラミックス複合材料のレーザフラッシュ法による熱拡散率測定方法	Determination of thermal diffusivity of continuous fiber-reinforced ceramic matrix composites by the laser flash method
JIS	H8453	2010	遮熱コーティングの熱伝導率測定方法	Measurement method for thermal conductivity of thermal barrier coatings

materials by the GHP method (APMP T.S-10). APMP T.S-9 is a joint project of TCT and the Technical Committee for Material Measurement (TCMM).

4.4 Standardization of the measurement technology

For the thermophysical quantity data to be utilized widely for design and performance evaluation of apparatus and instruments, evaluation of energy-saving performance, or guarantee of quality and safety, it is desirable that the measurement procedure and the specifications of the apparatus used to measure thermophysical quantities are standardized and operated by a standard guideline.

The list of the standards documents to which the results of this research and related activities contributed is shown in Table 2. One of the authors (Baba) worked as the chairman or project manager of the Draft Proposal Committee for the two JIS standards (JIS H7801 and R1667).[44][45] He also participated as the Japanese representative to WG15 of ISO/TC206 (Fine Ceramics) and contributed to the establishment of ISO 18755 "Fine Ceramics: Determination of thermal diffusivity of monolithic ceramics by laser flash method."[46]

The technologies of thermal diffusivity measurement can be roughly classified into the general technologies not specific to measured material, and the individual technologies specific to each material. The former are shared in society as universal technological standards through standardization. In the case of the thermal diffusivity measurement by the laser flash method, as shown in Fig. 10, the general technologies independent of the material can be classified into the specifications of the measurement apparatus (hardware technology) and data analysis (software technology).

The following technical requirements are listed in this standardization:

1) A specimen is maintained stably in a condition where contact thermal conductance with the specimen holder is minimized.
2) Temperature of the specimen is kept constant and is accurately measured.
3) Front face of the specimen is pulsewise heated by spatially uniform beam.
4) Duration of pulse heating should be sufficiently short compared with the heat diffusion time across the specimen.
5) Temperature rise at the rear face of the specimen after pulse heating is observed by a detector with sufficiently fast response.

The developed technologies which satisfy these conditions have been transferred to the manufacturers of the apparatus.

The authors (Akoshima and Baba) contributed to the revision of JIS R1611 in 2010.[47] In this revision, procedure for uncertainty evaluation of measurement data, calibration/

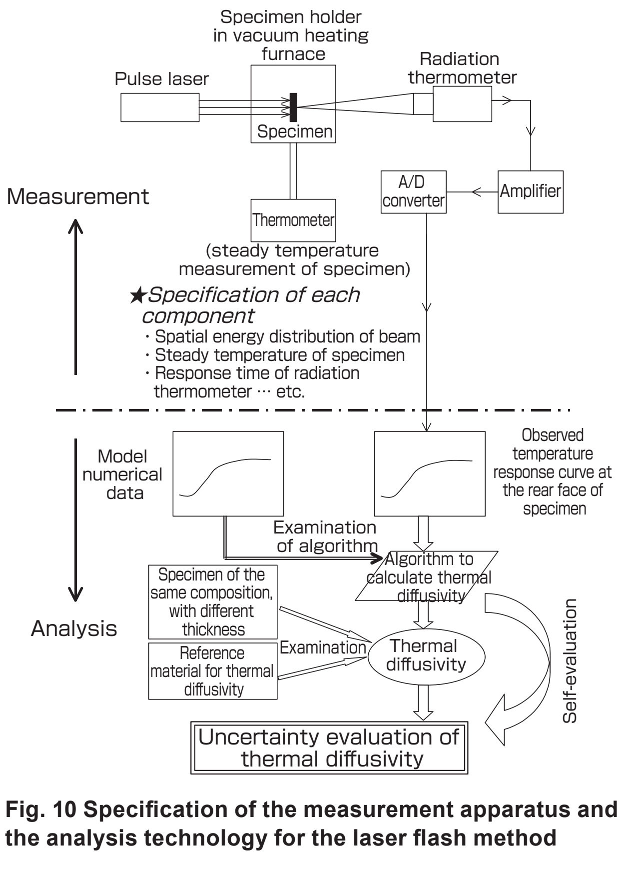

Fig. 10 Specification of the measurement apparatus and the analysis technology for the laser flash method

validation method using thermal diffusivity reference materials, estimation of intrinsic thermal diffusivity as physical properties of material, and quantitative evaluation method of the effect of black coating on surfaces of the specimen were added in the Appendices.

Our efforts in thermophysical quantity measurement were exerted in a triangle set of style as shown in Fig. 11 where the metrological standards, standardization, and practical measurement technologies contributed mutually to the overall improvement. Similar effort was made for the thermal diffusivity measurement of thin film.[17]

5 Current situation of developing a social system of thermophysical quantities

Figure 12 shows categories of technologies related to the thermal diffusivity measurement by the laser flash method and their relationships. As shown in the middle of the figure, thermal diffusivity is calculated by fitting the theoretical curve to the temperature response curve using a data analysis algorithm. In this case, there was a synergetic effect where the progress of data analysis technology was inspired by the improvement of measurement technology, and also the demand for an improved measurement apparatus was induced by the progress of the analysis technology. Therefore, the measurement technology and data analysis technology closely interacted with each other and evolved cyclically.

One specific example is that one-dimensional heat diffusion in a specimen was realized by homogenizing the spatial energy distribution of the pulse laser beam using optical fibers.[22][23] Stimulated by such advances in the measurement technology, universal data analysis technology for one-dimensional thermal diffusivity was developed as an application of a response function method.[48]-[50]

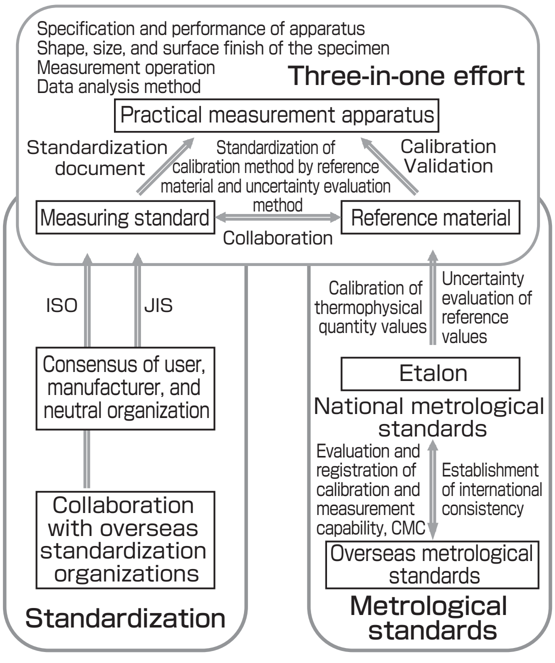

Fig. 11 Standardization of measurement technology and collaboration with metrological standard

6 Summary and future development

A triangle set of measurement technologies, metrological standards, and standardization for thermal diffusivity measurement by the laser flash method was developed synthetically in order to create a social system for efficiently and quickly providing highly reliable thermal diffusivity data. The national standards and reference materials were established based on the measurement and data analysis technologies developed in this research. These technologies have also been transferred to practical measurement apparatus. JIS and ISO standards were revised reflecting these new technologies. A database of thermophysical quantities was developed to accumulate traceable data produced from this social system, and was opened on the Internet.

Current approach for uncertainty evaluation and traceability will be extended to the general thermophysical quantity data, not just to the standard values and standard data of the reference materials. A method for quantitatively expressing the correlation between the material characters and quantity values is expected to be a key technology.

By actively collaborating with the overseas institutions working on the physical quantity data of substances and materials as well as Japanese institutions (such as universities, national laboratories, academic societies, companies, etc.), we aim to develop a system to continuously produce highly reliable thermophysical quantity data which is the basic information for science and technology and accumulate the produced data in the Network thermophysical property database.

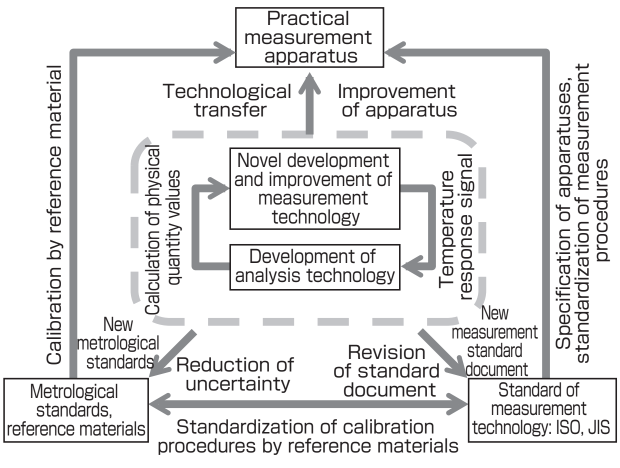

Fig. 12 Systematized techniques for thermal diffusivity measurement by the laser flash method

In the future, we hope to evolve this system into an international framework. The practical measurement apparatus of the laser flash method whose measurement target is bulk materials is commonly used among a wide range of users throughout the world. The international standard is set as ISO/TC206 (Fine Ceramics), and further efforts are in progress for the organization of the metrological standards under the Meter Convention in the CIPM CCT Task Group on thermophysical quantities. We aim to spread the standards to the manufacturers of the measurement apparatus, research institutes, energy related companies, electronic companies, and others that need the thermophysical quantity data, in collaboration with the overseas NMIs.

Appendix A Effort in the academic societies

In Japan, importance of thermophysical quantity data was strongly recognized in the 1970s when science, technology, and industry caught up with the developed countries. In 1980, the Japan Research Meeting for Thermophysical Properties was established, and this was renamed the Japan Society for Thermophysical Properties (JSTP) in 1990.[51]

The JSTP hosts the Japan Symposium on Thermophysical Properties every year and publishes the academic journal *Netsu Bussei (Japan Journal of Thermophysical Properties)*.[52] Also, the Asian Thermophysical Properties Conference (ATPC) was started in 1986, and the World Congress on Thermophysical Properties was formed in collaboration with the research institutions on thermophysical properties of the United States and Europe, and it provides a place for worldwide research exchange.

The JSTP published the *Netsu Bussei Handbook* (Thermophysical Property Handbook) in 1990 that compiled information on thermophysical properties in wide-ranging fields of science and technology.[53] The *Netsu Bussei Handbook* not only collected thermophysical quantity data, but also provided definitions and descriptions of thermophysical quantity values, how to search data, data availability, and measurement methods. It systematically described how to obtain and utilize thermophysical quantity data, including cases in which one must obtain thermophysical quantities through actual measurement by oneself. In 2008, it was revised to reflect the progress in science and technology and the increase in data from the first edition up to that time, and was published as the *Shinpen Netsu Bussei Handbook* (Thermophysical Property Handbook, New Edition).[4]

The importance of the scientific and technical infrastructure was pointed out in the Technology Basic Plan of Japan announced in 1996.[54] Metrological standards, safety management of chemical substances, human life and welfare, biological resource information, and materials were selected as the fields of focus where organization and R&D should be done actively to promote the creation of new industry by the "Special Committee on Measurement Standards and Intellectual Infrastructure" which was organized in 1998 as a joint effort of the Industrial Technology Council and the Japanese Industrial Standards Committee.[55] In linkage with this movement, the JSTP investigated the "Role of Thermophysical Properties in Intellectual infrastructure" in 1997, and this was reported as a proposal of JSTP in 1998.[2]

Appendix B Efforts at CIPM

For over 100 years since the establishment of the Meter Convention, national standards were mainly set for basic quantities such as length, mass, time, electricity, and temperature. However, in recent years, equivalence of analysis results for the content of elements and chemical substances became widely required in the tests for food safety and environmental pollutants, health check, and others. International consistency and traceability were demanded for chemical measurements.[56] In response to such demands, the Consultative Committee for Amount of Substance (CCQM) was established in CIPM within the framework of the Meter Convention in 1995.[57]

While the physical standards are provided through the calibration services with the national standards based on the calibration and measurement capabilities (CMCs) of the NMIs of each country, in general, the material quantity standards are mainly provided by the certified reference materials (CRMs) and thereby the traceability is maintained.

In the field of material measurement evaluation, the Versailles Projects on Advanced Materials and Standards (VAMAS) was organized "to support trade in high technology products, through international collaborative projects aimed at providing the technical basis for drafting codes of practice and specifications for advanced materials" among the participating countries, at the Versailles Summit in 1982.[58]

At the CIPM, considering the importance of the technological role and trade of "materials" that support modern society, it decided to establish the Ad Hoc Working Group of Material Metrology (WGMM) in 2005. This was composed of the NMIs of the world and the specialists from material research institution, with the purpose of studying general material quantities including mechanical and electric quantities of materials as well as thermophysical quantities. Discussions were started in 2006, and the final report was submitted to the CIPM in 2007.[59] The WGMM concluded that instead of establishing a new consultative committee for material measurement, the WG on materials in the existing CC should work in liaison with VAMAS. The details of the activities were published as a special edition of *Metrologia*.[60]

WG9 for Thermophysical Quantity was established in CCT in 2003.[61] One of the authors (Baba) was the chairman of WG9 from 2005 to May, 2014. WG9 decided to start three pilot studies for international comparisons of thermal conductivity of insulation materials by the guarded hot plate (GHP) method, thermal diffusivity of dense solid materials by the laser flash method, and thermal radiation of dense solid materials from 2008. There are comprehensive needs for their metrological standards among a wide range of fields of science and technology.[16] Thermal diffusivity of a dense solid material is intrinsic to the material with which the same value can be obtained independent of the measurement methods as described in chapter 1[27][62]. The measurement results of the participating NMIs were collected at the pilot institution and the final report is currently being drafted. Activities of WG9 were succeeded by the Task Group on thermophysical quantities in May, 2014.

Appendix C Effort of NRLM

The National Research Laboratory of Metrology (NRLM), Agency of Industrial Science and Technology, Ministry of International Trade and Industry was one of the predecessors of the National Metrology Institute of Japan, AIST. It conducted the “Survey Research for Physical Property Measurement” and the results were presented as a report in 1985.[1] The thermophysical property department was established based on this investigation, and the organizational effort for the research on the measurement technology and metrological standard for thermophysical quantities was started.

As part of the effort, within the Promotion System for Intellectual Infrastructure of Research and Development of the Special Coordination Funds for Promoting Science and Technology, the former NRLM acted as a core institution and instigated the five-year plan for “Research on the thermophysical property measurement technology and reference material for functional materials” from FY 1997. In this research topic, the following goals were set for density, thermal conductivity/diffusivity, specific heat capacity, thermal expansion, radiation, acoustic speed, and elasticity. These were conducted as a joint research project by over ten Japanese institutions.[3][15][63]

1) To establish primary standards of Japan by developing precision measurement technology for thermophysical property values.
2) To develop reference specimens and materials, to obtain standard data, and to widely distribute them to sites of research and production.
3) To develop state-of-the-art measurement methods capable of handling advanced functional materials, and to standardize practical measurement methods.
4) To identify material character for certain important materials, to create coherent datasets for thermophysical property values (set of data that includes the character for identifying the materials as well as the thermophysical property values), and to verify the efficacy of the approach of this research.
5) To create prototypes of thermophysical property database and then to verify the efficacy as a data diffusion tool.

A social system for production and utilization of thermophysical quantity data has been developed through the accumulation of projects including this Intellectual infrastructure Organization Promotion Project. The prototypes of thermophysical property database described in 5) has been evolved into the network thermophysical property database opened on the NMIJ web-site.[64]-[66]

References

[1] National Research Laboratory of Metrology AIST-MITI (ed.): Survey Research Report on Physical Property Measurement Technology, (1985) (in Japanese).
[2] S. Fukusako: What Thermophysical Properties Activities should be as a Technical Infrastructure, *Japan Journal of Thermophysical Properties*, 12 (3), 152-169 (1998) (in Japanese).
[3] A. Ono: Technical Infrastructure and Thermophysical Properties, *Japan Journal of Thermophysical Properties*, 12 (1), 20-30 (1998) (in Japanese).
[4] Japan Society of Thermophysical Properties (ed.): *Thermophysical Properties Handbook, New Edition*, 23-25, Yokendo (2008) (in Japanese).
[5] http://www.nist.gov/srm/
[6] http://www.nist.gov/srd/
[7] http://irmm.jrc.ec.europa.eu/reference_materials_catalogue/Pages/index.aspx
[8] http://www.iajapan.nite.go.jp/jcss/outline/index.html
[9] M. Akoshima: Development of thermal diffusivity standard of solid materials, *Measurement Standards and Metrology Management*, 59 (2), 23-29 (2009) (in Japanese).
[10] http://www.springermaterials.com/navigation/
[11] National Research Council: International Critical Tables of Numerical Data, *Physics, Chemistry and Technology Vol. 1-7*, U.S. (1929).
[12] D. R. Lide Jr.: Critical data for critical needs, Science, 212 (4501), 1343-1349 (1981). DOI:10.1126/science.212.4501.1343
[13] Y. S. Touloukian and C. Y. Ho eds.: *Thermophysical Properties of Matter; TPRC Data Series*, 1970-1972, IFI-Plenum, New York (1970).
[14] https://cindasdata.com/
[15] A. Ono, T. Baba and K. Fujii: Traceable measurements and data of thermophysical properties for solid materials: a review, *Meas. Sci. Technol.*, 12 (12), 2023-2030 (2001).
[16] T. Baba: Measurements and data of thermophysical properties traceable to a metrological standard, *Metrologia*, 47 (2), S143-S155 (2010). DOI:10.1088/0026-1394/47/2/S12
[17] Joint Meeting of the Industrial Structure Council and the Japanese Industrial Standards Committee: *Interim Report Compiled by the Special Committee on Measurement Standards and Intellectual Infrastructure –Promotion of the Program to Develop and Promote the Utilization of Intellectual Infrastructure–*, 49 (2012) (in Japanese). http://www.meti.go.jp/press/2012/08/20120815002/20120815002-3.pdf

[18] T. Baba and Y. Yamashita: Development of a network database system for thermophysical property data, *Measurement Standards and Metrology Management*, 59 (2), 49-57 (2009) (in Japanese).
[19] W. J. Parker, R. J. Jenkins, C. P. Butler and G. L. Abbott: Flash method of determining thermal diffusivity, heat capacity, and thermal conductivity, *J. Appl. Phys.*, 32 (9), 1679-1684 (1961).
[20] Y. Nagasaka and T. Baba: Optical measurement technology of solid thermophysical properties, *Progress in Heat Transfer, New Series*, 3, Chap. 3, 163-226, Yokendo, (2000) (in Japanese).
[21] A. Cezairliyan, T. Baba and R. Taylor: A high-temperature laser-pulse thermal diffusivity apparatus, *Int. J. Thermophys.*, 15 (2), 317-341 (1994).
[22] T. Baba and A. Ono: Improvement of the laser flash method to reduce uncertainty in thermal diffusivity measurements, *Meas. Sci. Technol.*, 12 (12), 2046-2057 (2001). PII: S0957-0233 (01) 27342-1
[23] T. Baba, M. Kobayashi, A. Ono, J. H. Hong and M. M. Suliyanti: Experimental investigation of the nonuniform heating effect in laser flash thermal diffusivity measurements, *Thermochim. Acta*, 218, 329-339 (1993).
[24] M. Kobayashi, T. Baba and A. Ono: Infrared radiation thermometer for laser flash thermal diffusivity measurements, *Japan Journal of Thermophysical Properties*, 8 (3), 143-148 (1994) (in Japanese).
[25] M. Ogawa, K. Mukai, T. Fukui and T. Baba: The development of a thermal diffusivity reference material using alumina, *Meas. Sci. Technol.*, 12 (12), 2058-2063 (2001). DOI:10.1088/0957-0233/12/12/305
[26] M. Akoshima and T. Baba: Study on a thermal-diffusivity standard for laser flash method measurements, *Int. J. Thermophys.*, 27 (4), 1189-1203 (2006).
[27] M. Akoshima, B. Hay, M. Neda and M. Grelard: Experimental verification to obtain intrinsic thermal diffusivity by laser-flash method, *Int. J. Thermophys.*, 34 (5), 778-791 (2013). DOI:10.1007/s10765-013-1470-7
[28] http://www.nmij.jp/service/
[29] T. Baba, K. Ehara, S. Yamada and K. Fujii: Achievement and future of physical property measurement and metrological statistics technology, *Measurement Standards and Metrology Management*, 60 (4), 2-9 (2011) (in Japanese).
[30] T. Baba, N. Yamada, N. Taketoshi, H. Watanabe, M. Akoshima, T. Yagi, H. Abe and Y. Yamashita: Research and development of metrological standards for thermophysical properties of solids in the National Metrology Institute of Japan, *High Temperatures-High Pressures*, 39 (4), 279-306 (2010).
[31] N. Taketoshi, T. Yagi and T. Baba: Current activity of thin film thermal diffusivity standard using pulsed light heating thermoreflectance method, *Measurement Standards and Metrology Management*, 59 (2), 36-41 (2009) (in Japanese).
[32] T. Yagi, N. Taketoshi and T. Baba: Thermophysical property measurement and standard of thin films: Standard thin film for thermal expansion time, *Measurement Standards and Metrology Management*, 59 (2), 42-48 (2009) (in Japanese).
[33] ISO/IEC 17025 1999 General requirements for the competence of testing and calibration laboratories.
[34] M. Akoshima and T. Baba: Thermal diffusivity measurements of candidate reference materials by the laser flash method, *Int. J. Thermophys.*, 26 (1), 151-163 (2005).
[35] M. Akoshima and T. Baba: Laser flash thermal diffusivity measurements of isotropic graphite and glass-like carbon, *Thermal conductivity*, 28, 497-506 (2006).
[36] ISO Guide 34:2009 General requirements for the competence of reference material producers.
[37] Mutual recognition of national measurement standards and of calibration and measurement certificates issued by national metrology institutes, BIPM (1999).
[38] http://kcdb.bipm.org/
[39] http://www.bipm.org/en/committees/cc/cct/
[40] http://www.apmpweb.org/
[41] http://www.bipm.org/en/committees/cc/cct/tor_wg9.html
[42] M. Akoshima, B. Hay, J. Zhang, L. Chapman and T. Baba: International comparison on thermal diffusivity measurement using laser flash method - preliminary results, *Thermal Conductivity*, 30, 367-377 (2010).
[43] M. Akoshima, B. Hay, J. Zhang, L. Chapman and T. Baba: International comparison on thermal-diffusivity measurements for iron and isotropic graphite using the laser flash method in CCT-WG9, *Int. J. Thermophys.*, 34 (5), 763-777 (2013). DOI:10.1007/s10765-012-1209-x
[44] JIS H7801 Method for measuring thermal diffusivity of metals by the laser flash method (established 2005).
[45] JIS R1667 Determination of thermal diffusivity of continuous fiber-reinforced ceramic matrix composites by the laser flash method (established 2005).
[46] ISO 18755:2005 Fine ceramics (advanced ceramics, advanced technical ceramics) - Determination of thermal diffusivity of monolithic ceramics by laser flash method.
[47] JIS R1611 Measurement methods of thermal diffusivity, specific heat capacity, and thermal conductivity of fine ceramics by flash method (established 1991, modified 2010).
[48] T. Baba: Introduction of a response function method for analysis of functionally gradient materials, *Japan Journal of Thermophysical Properties*, 7 (1), 14-19 (1993) (in Japanese).
[49] T. Baba and N. Taketoshi: Analysis of thermal diffusion in multi-layer thin films by a response function method, *Proc. of Eurotherm.*, 57, 285-292 (1999).
[50] T. Baba: Analysis of one-dimensional heat diffusion after light pulse heating by the response function method, *Jpn. J. Appl. Phys.*, 48, 05EB04. 1-05EB04.9 (2009).
[51] http://www.netsubussei.jp/
[52] http://www.jstage.jst.go.jp/browse/jjtp/-char/ja
[53] Japan Society of Thermophysical Properties (ed.): *Thermophysical Properties Handbook, First Edition*, Yokendo (1990) (in Japanese).
[54] http://www8.cao.go.jp/cstp/kihonkeikaku/index4.html
[55] http://www.meti.go.jp/report/committee/data/g_commi03_01.html
[56] Evolving Needs for Metrology in Trade, Industry and Society and the Role of BIPM, edited R. Kaarls, (2007) http://www. bipm.org/utils/en/pdf/Kaarls2007.pdf
[57] http://www.bipm.org/en/committees/cc/ccqm/
[58] http://www.vamas.org/index.html
[59] S. Bennett and G. D. Sims: Evolving Needs for Metrology in Material Property Measurements, *Report of the CIPM ad hoc Working Group on Materials Metrology (WGMM)*, (2007).
[60] S. Bennett and J. Valdés: Special issue for materials metrology: *Metrologia*, 47 (2) (2010). DOI:10.1088/0026-1394/47/2/E01
[61] http://www.bipm.org/en/committees/cc/cct/working_groups.html#wg9
[62] R. G. Munro: Data Evaluation Theory and Practice for Materials Properties: *NIST Recommended Practice Guide*, SP 960-11, U.S. Government Printing Office, Washington (2003).
[63] http://www.mext.go.jp/a_menu/kagaku/chousei/data/14/

hyoka030129/2/40.pdf
[64] http://tpds.db.aist.go.jp/
[65] T. Baba, Y. Yamashita and A. Nagashima: Function sharing and systematic collaboration between a networking database system and printed media on thermophysical properties data, *J. Chem. Eng. Data*, 54, 2745-57 (2009).
[66] Y. Yamashita, T. Yagi and T. Baba: Development of network database system for thermophysical property data of thin films, *Jpn. J. Appl. Phys.*, 50 (13) 11RH03 (2011).

Authors

Tetsuya BABA

Completed the doctorate course at Graduate School of Science of the University of Tokyo in 1979 (Doctor of Science). Joined the National Research Laboratory of Metrology, Agency of Industrial Science and Technology, Ministry of International Trade and Industry in 1980. Visiting Researcher, National Institute of Standards and Technology, USA from 1986 to 1988. Chief, Thermophysical Property Section, Physical Property Statistics Division, National Metrology Institute of Japan, AIST in 2001. Chairman, Working Group 9, Thermophysical Quantities, Consultative Committee for Thermal Metrology, Comité International des Poids et Mesures (CIPM) from 2005 to 2014. Prime Senior Researcher, National Metrology Institute of Japan, AIST from 2007 to 2012. President of Japan Society of Thermophysical Properties in 2011. Engaged in the research of measurement technology, metrological standards, and reference materials for thermal quantities and the development of thermophysical quantity database. Received the Thermal Conductivity Award in 1999; Ichimura Award for distinguished accomplishment in science in 2000; Commendation for Science and Technology in Research by the Minister of Education, Culture, Sports, Science and Technology in 2005; and Significant Contribution Award of the Asian Thermophysical Properties Conference in 2013. In this paper, led the entire project.

Megumi AKOSHIMA

Completed the doctoral course at the Department of Applied Physics, Graduate School of Engineering, Tohoku University in 2000. Ph.D.(Engineering). Research Fellow (DC) of the Japan Society for the Promotion of Science from January 1999 to March 2001. Joined AIST in 2001. Appointed to the Thermophysical Property Standard Lab, Physical Property Statistics Division (currently Material Physical Property Division), National Metrology Institute of Japan. Since joining AIST, engaged in the research for the thermal diffusivity measurement of solid materials. Established the method for determining the intrinsic thermal diffusivity of materials that are SI traceable using thermal diffusivity measurement using the laser flash method. Receives subcontract testing and provides reference materials. Recently, involved in the CCT WG9 and APMP TCT as the organizer for the international comparisons of thermal diffusivity. In this paper, was in charge of the development and standardization of the measurement technology, metrological standard, and reference materials of thermal diffusivity, as well as the international comparisons.

Discussions with Reviewers

1 Overall evaluation

Comment (Akira Ono, AIST)

This paper is a description of excellent synthetic research with a goal to build a whole system that allows efficient production and effective use of thermophysical properties of solid materials in society. I am impressed with the grand scale of the research goal and the practical scenario to realize such a goal that invites the involvement of multiple stakeholders.

2 Situation of thermophysical property research in the USA

Question (Akira Ono)

In subchapter 3.2, the authors look back at the thermophysical property research in the United States from the 1960s to 70s, and you mention that the collaboration between the thermophysical property reference materials of the National Bureau of Standards (NBS) and the data evaluation of the Thermophysical Property Research Center (TPRC) at Purdue University was not quite sufficient.

At the time, thermophysical property research was very active in the US, and the results contributed to various technological fields. Are the authors saying that even though the reference materials were used widely in society and consequently many good thermophysical property data were produced, there was not enough time for them to be reflected in the Purdue University data book? What were the points that constrained the thermophysical property research in the US?

Also, please give the authors' opinion on what motivation or scenario could have been shared between the NBS and Purdue University to encourage their better collaboration?

Answer (Tetsuya Baba)

The "data evaluation" at Purdue University TPRC was conducted before the concept of measurement traceability and uncertainty prevailed over science and technology. Therefore, rather than evaluating data from the traceability of measurement, the reported data were overviewed as a whole, and evaluation was done individually based on the knowledge of the evaluator on physical properties and the reputation of the institution that conducted the measurement.

Development of thermophysical property reference materials at NBS was started slightly later than the data evaluation at TPRC, and most data in TPRC data series have been published in the latter half of the 1970s. Unfortunately, there has been no comprehensive revision of the thermophysical property data book of TPRC or its succeeding institution CINDAS.

If Purdue University had enough time to recognize the importance of the "uncertainty and traceability of measurement" in its data evaluation, it could have created reference information from the data obtained using a measurement apparatus calibrated by NBS reference materials, and could have developed a general data evaluation method based on measurement uncertainty, by introducing the viewpoint of evaluating the reliability of other general data using the reference information.

3 Maintaining traceability by information

Question & Comment (Akira Ono)

In chapter 4, the authors mention that in the field of material standards, there may be possibility that the traceability can be maintained only by information, without depending on artifacts such as standards or reference materials. This is a flexible idea about traceability unheard of before, and I hope the authors proceed with it.

(1) Specifically, with which materials is it possible to maintain the traceability only by information? Do you think thermophysical property of single crystal silicon is a candidate?

(2) I think the idea of maintaining the traceability only by information is not widely accepted by the metrological community at the moment. Can the authors introduce typical examples, such as how the uncertainty (variation and bias) of measurement results is evaluated when the traceability is maintained only by information. If the authors have any thoughts on how this idea can be widely accepted, please state them.

Answer (Tetsuya Baba)

(1) The thermophysical quantity data of water in liquid and gas phase can be supplied as universal data with evaluated uncertainty as long as the compositions (amount of impurities and isotope composition ratio are considered as needed) are specified and the measurement traceability is maintained. For example, for temperature scale, the triple point of pure water with the isotope composition of Vienna Standard Mean Ocean Water (VSMOW), which is a reference material of the International Atomic Energy Agency (IAEA), can be realized universally without the calibration by a higher level standard. For the viscosity standard, the viscosity of distilled water at 20 °C at atmospheric pressure as designated by ISO/TR3666:1998(E) comprises the primary standard.

For specific heat capacity and thermal expansion, the silicon single crystal of a certain lot is stored at NMIJ, their homogeneities are evaluated, valuation is done using the national standard, and these are provided as reference materials. As the next step, the possibility of providing the standard data for specific heat capacity and thermal expansion of silicon single crystal with better purity than the designated one can be provided as information without depending on artifacts. Also, for silicon single crystal, we think it is possible to provide the standard data for thermal diffusivity without depending on artifacts, as described in this paper.

Complimentary to the abovementioned efforts on the provision of national metrological standards, aluminum oxide can be mentioned as a reference material for specific heat capacity, as the de facto standard born from the demand of the users and instrument manufacturers. As a reference material for enthalpy and specific heat capacity, NIST provides “Enthalpy and Heat-Capacity Standard Reference Material: Synthetic Sapphire” (Alpha-Al_2O_3) which is provided in a thin columnar shape. Since disk shaped specimens are commonly used for differential scanning calorimetry (DSC), this NIST CRM is not appropriate for DSC, and the general practice is to use the commercially available highly pure alumina of a disk shape. There has been no systematic investigation of the information about the variation in specific heat capacity of the alumina from a random lot produced by the same manufacturer or different manufactures yet. This would be of an issue for investigation in the future.

(2) In order for the idea that traceability can be maintained with information only to be accepted by the metrological community, first, it is necessary to prove that the values are consistent with the results of the measurements traceable to the national standard. Second, it is essential to clearly characterize materials and substances that correspond to the information. For gas and liquid, substances can be quantitatively defined by composition, but for solids, the methodology to quantitatively describe the structures of different hierarchy of micro, meso, and macro scales has not been established yet.

Today, a vast amount of digital information is produced, shared on the web, and analyzed and utilized. How to evaluate and guarantee the reliability and credibility of such data is the most basic and most difficult problem. Among various data, the reliability and credibility of the quantitative data of the measurement apparatus including sensors can be expressed as uncertainty, and we believe the realization of “traceability of information” is a promising approach.

4 Standardization of the uncertainty evaluation procedure

Question & Comment (Akira Ono)

In subchapter 5.4, the authors state that the calibration/validation method and uncertainty evaluation method of measurement data using thermal diffusivity reference materials were standardized in JIS R1611. I think this is a wonderful achievement and should be highly evaluated.

Now please explain briefly yet specifically, how the authors standardized the procedure of the uncertainty evaluation method in JIS R1611.

Also, I expect that this movement will spread to the standardization of measurement methods of physical properties other than thermal diffusivity as well as to standardization of general test and evaluation method. What would be the point in enabling this spread in the future?

Answer (Megumi Akoshima)

The explanation of the evaluation procedure for uncertainty was added in the 2010 revision of JIS R1611 “Measurement methods of thermal diffusivity, specific heat capacity, and thermal conductivity for fine ceramics by flash method.”

Specifically, the following appendices were added in this revision.

Appendix E: Reference data and reference materials of thermal diffusivity

Appendix JB: Calibration/validation using reference specimen and correction of the measurement result

Appendix JD: Uncertainty evaluation of thermal diffusivity

Appendix JC: Evaluation method for the effect of coating for thermal diffusivity measurement by flash method

With the measurement of thermal diffusivity by the flash method, there are cases in which calibration or correction is done using the reference specimens shown in Appendix JB, and cases in which absolute measurement is taken for heat diffusion time without the reference specimen, and the method for evaluating the uncertainty in both cases are shown in Appendix JD.

When the specimen is transparent, or in a case where the absorption of flash heating light of the specimen or the radiation at the radiation thermometry wavelength are not sufficiently high, black, opaque thin film is applied on both surfaces of the specimen. The presence of this thin film increases the heat diffusion time more than an untreated specimen. The method for evaluating and correcting the increase is described in Appendix JC.

As an assumption for determining the uncertainty evaluation method for the measurement data and the calibration/validation method in the measurement of physical quantities, I think the physical quantities must be intrinsic quantities of the materials that do not depend on the measurement method.

In a case of quantity dependence on the measurement method (procedural quantity), even if the same material is measured, the measured value may differ depending on the measurement method.

In JIS R1611, the uncertainty evaluation method is much more effective because it explains in the standard the measurement method including the method for verifying whether the measured thermal diffusivity is an intrinsic quantity.

To spread the calibration/validation method and the uncertainty evaluation method of the measured data, I think the points are to systematically work on the development of precise measurement technology indicated in this paper, the organization of metrological standards and reference materials, and the standardization of the measurement methods.

5 Importance of the triangle set of efforts

Question & Comment (Akira Ono)

In subchapter 5.4 and chapter 6, the authors state that the root of the social system for production and utilization of the thermophysical property data is the “organization of the metrological standards and reference materials,” “standardization of the measurement method,” and “development and popularization of practical measurement apparatus.” I expect that such efforts will occur in fields other than thermophysical quantities as well in the future. What were the factors that led to such efforts in the field of thermophysical properties? I think it will be useful for people of other fields if the authors, who were directly involved, provide comments.

Answer (Tetsuya Baba)

In the basic quantities such as length or temperature, there are several digits of differences in precision between the metrological standards and the practical measurements, and the technologies used for metrological standards are usually quite different from the technologies of practical measurements. In this paper, approach of organizing the national standard for thermal diffusivity by the laser flash method was chosen since it was widely used as the practical measurement technology, and therefore, the result of the technological development contributed to both the metrological standard and the practical measurement technology. Moreover, the developed elemental technologies, the achievement of high precision of practical measurement technology through their systemization, and the realization of traceability to the national standard through the developed reference materials were reflected in the standardization of the measurement technology.

Such an approach is employed in the technology for applying the ultra high-speed laser flash method to thin films, and a similar triangle set of a social system is being organized.

I expect similar approaches can be applied to the social system not only for thermophysical quantities, but also to the production and utilization of data for mechanical electrical, magnetic, and optical quantities.

6 Expectations for the stakeholders in the development of a social system

Question & Comment (Akira Ono)

Figure 1 shows the research scenario for developing a social system where the thermophysical property data are efficiently produced and effectively used. Currently, the authors have the skeleton of this social system, and I think the content will be filled in gradually along with the strengthening of the skeleton.

For this system to become established in society, I think there are many roles expected from the various stakeholders of thermophysical properties (such as universities, public research institutions, material manufacturers, material users, and standardization organizations). What activities do the authors expect from the stakeholders?

Answer (Tetsuya Baba)

For the manufacturers of measurement apparatus for thermophysical quantities, I expect them to widely provide to society the thermophysical quantity measurement apparatus for which the uncertainty of measurement has been objectively guaranteed and is traceable through the reference materials and standards.

For the universities and academic societies, I hope they will discover new approaches for evaluating the thermophysical quantity data that continue to increase with the advances in measurement technology, utilizing the advancements in traceability, uncertainty evaluation, and information technologies.

For public research institutions, I expect them to collect and evaluate the thermophysical quantity data of their field of specialty, in a systematic and continuous manner. For example, I expect that AIST will be involved in the thermophysical quantity data for energy or electronics related materials. Likewise I expect that the Japan Atomic Energy Agency will be handling the thermophysical quantity data needed for nuclear industry and research.

The material manufacturers can make measurements of thermophysical quantity values using the measurement technology traceable to the metrological standards, and provide these data along with the uncertainty of measurement. Also, indication of the variation of thermophysical quantity values for all material products that they supply would be useful.

The material users can discover materials with overall properties (including thermophysical quantities) needed to advance products or innovation through our social system. If the existing materials cannot fulfill the overall properties, AIST shall serve as an information hub for passing on the demand for a material that may fulfill the necessary overall properties to the material manufacturers.