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Development of Network Database System for Thermophysical Property Data of Thin Films

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The database system for thermophysical property data, which has been developed by the National Metrology Institute of Japan (NMIJ), is evolved to store comprehensive information on thin films. Since a thin film is identified by not only its constituent elements, phases, or compositions but also its method of synthesis, a strategy for storing all thin-film specimens is adopted when the data are measured by NMIJ. In addition, new criteria of material classification in our database are introduced to systematically manage material information on multilayer thin-film specimens. For example, when a multi layered film is measured by an ultrafast laser flash method, the database stores not only analyzed results but also data at various measurement stages, which can follow analysis steps from an observed signal data to derived results: thermal diffusivity and boundary thermal resistance of the film. In order to store new data items, the database system has been updated. The updated database system is demonstrated in terms of its storage of record items and its user interface using a set of thermophysical property data of a "TiN single-layer thin film on a synthesized quartz substrate", "Al_2O_3 coated with Mo three-layer thin films on a fused silica substrate", and "ITO coated with Mo three-layer thin films on a fused silica substrate". This database system is available at http://riodb.ibase.aist.go.jp/TPDB/DBGVsupport/index_en.html.

1. Introduction

A thin film is an indispensable material for recent informative devices such as central processing units, phase-change random access memories, optical disks, light-emitting diodes, and flat panel displays, which are composed of thin films[1–9] with thickness of several nanometers to several 100 nanometers. The thermophysical properties of thin films are generally different from those of bulk materials of same name/same composition, because material characters such as crystal grain size differ depending on the type of synthesis method. For example, it was reported that the thermal diffusivity of a Mo thin film deposited on a Pyrex 7740 glass substrate by rf-sputtering at 11 Pa of Ar pressure is less than one-tenth of that of bulk Mo.[10,11] Consequently, it is desirable for thermophysical properties of a thin film synthesized to the same conditions, of which films in devices are made, in terms of film thickness and the type of deposition method. Regarding multilayered structures seen in recently developed devices, it is also necessary to know the boundary thermal resistance at an interface to understand internal heat transfer in devices.[4–6,8,9,12)]

Although several data books[13–16] have been published and several databases[17–22] have been developed concerning thermophysical properties, the majority of the data are for pure bulk materials except those in the "Interfacial thermal conductance database" developed by the National Institute of Materials Science,[23] Japan, which stores the data of boundary thermal conductance calculated using the diffusion mismatch model.[24,25]

The National Research Laboratory of Metrology (NRLM), from which the National Metrology Institute of Japan (NMIJ) was reorganized in 2001, started the development of a network database system for thermophysical properties in 1995.[26–29] NRLM organized a national project entitled "Research on Measurement Technology and Reference Materials for Thermophysical Properties of Solids" started in 1997 with a five-year term having more than ten participating laboratories and universities supported by the Promotion System for Intellectual Infrastructure of Research and Development, promoted by the Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT), where a prototype network database system for thermophysical properties was developed.[26] After NMIJ was founded, the Nanotechnology Material Metrology Project, which aims to develop measurement methods and related reference materials for nanomaterials, was started in 2001 and completed in 2007 under the support by the Japanese Ministry of Economy, Trade and Industry and the New Energy and Industrial Technology Development Organization (NEDO).[30] Under the project, the improvement in the thermophysical property database has been started to store multifunctional information on nanomaterials such as thin films.[31,32]

The details of the network database system for thermophysical property data are described in §2. The ultra fast laser flash method, which is designed to measure the thermal diffusivity of thin films, is explained in §3. The extension of the database system for storing the thermophysical property data of thin films is described in §4. In §5, examples of stored thermophysical property data of thin film in the database are shown.

2. Network Database System for Thermophysical Property Data

Thermophysical property database is one of material databases and has difficulty in characterization of materials and systematic storage of material data, as well as other material databases. The origin of these difficulties and solutions in our database are described in following subsections.

2.1 Identification of material

When databases or data books of thermophysical properties are used, we face the difficulty in determining how the material of interest can be identified to the material listed in the databases or databooks.

In the case of gases and liquids including melts of solid materials, they are sufficiently identified if the composition

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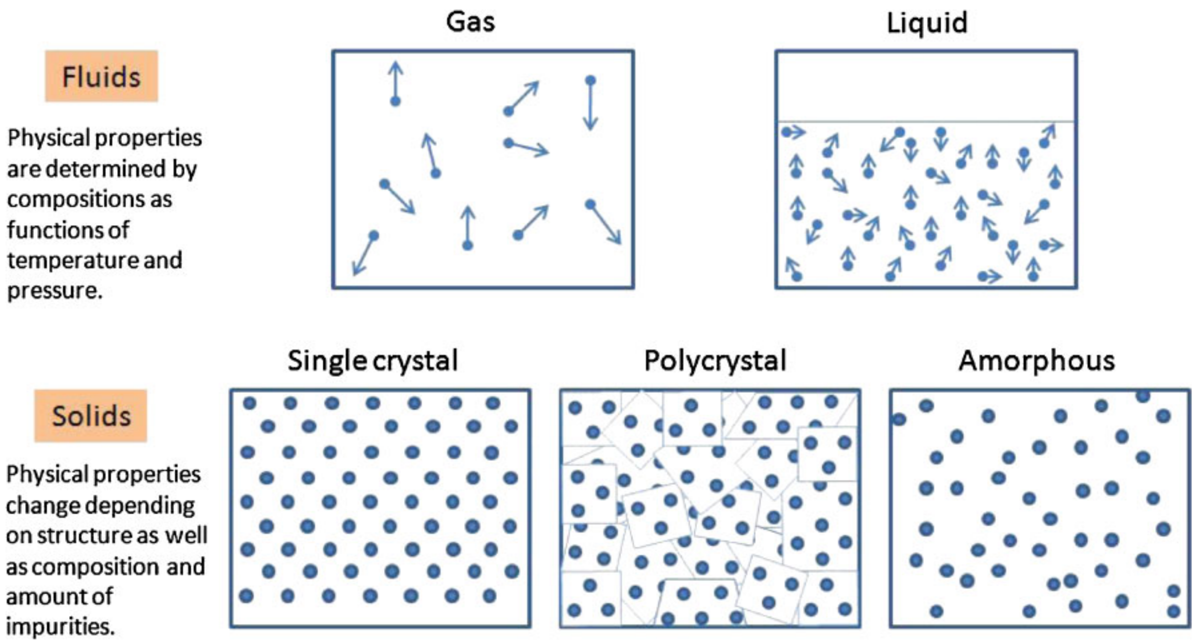

Fig. 1. (Color online) Schematic view of configurations of atoms of gas, liquids, solids, and complex structures of solids.

of main components and the concentration of impurities are known. However, the identification of solid materials is much more difficult than that of gases and liquids since their properties change depending on their structure. For example, a specimen might be single-crystalline, polycrystalline, or amorphous, as shown in Fig. 1. In the case of the single-crystalline specimen, it might take different crystal systems such as cubic and hexagonal systems. In the case of the polycrystalline specimen, they have large variations in the size, shape, and direction of the constituent single crystals. Information about grains and grain boundaries is also necessary. The specimen might be porous. Then, the porosity, size, shape, distribution, and orientation of pores are essential information.

In particular, transport properties such as thermal conductivity and thermal diffusivity are strongly dependent on the structure of materials. In contrast, thermodynamic properties such as specific heat capacity and enthalpy are not sensitively dependent on the structure. Figure 2 shows a schematic view of the difference in the temperature dependence of thermal conductivity among different structures of a single crystal bulk material, a polycrystalline bulk material, a polycrystalline thin film, and an amorphous bulk material, for a typical oxide of the same atomic composition.

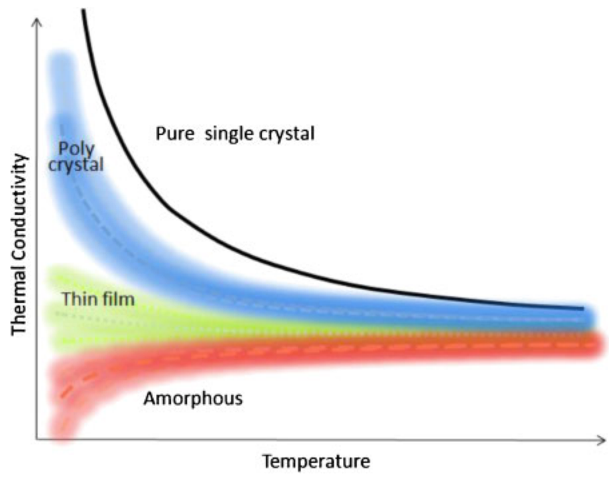

Fig. 2. (Color online) Schematic view of difference in temperature dependence of thermal conductivity among different structures, i.e., single crystal bulk material, polycrystalline bulk material, polycrystalline thin film, and amorphous bulk material, for typical oxide of same atomic composition.

2.2 Identification and classification of materials for database

2.2.1 Specimen and lot

In many cases, bulk materials are produced in a batch sintered at the same time in a furnace or as an ingot grown by the Czochralski method. A batch of thin films is synthesized, for example, by a single sputtering process in a chamber. In the thermophysical property database of NMIJ, "Lot" is assigned to this batch of materials produced or synthesized at the same time.

Each thermophysical property is measured for each specimen. If a series of measurements are completed for a set of specimens sampled from a specified lot, a set of statistical information is obtained about the thermophysical property of the lot including the mean and standard deviation of the measured thermophysical property values attributed to the nonuniformity of the lot. Note that the scattering of the measured thermophysical property values can be attributed to both the nonuniformity in the lot and the uncertainty of the measurements caused.

2.2.2 Grade

This level stores information on a set of materials produced under a well-defined procedure or using the specification prescribed in documents. The documents can contain international standards such as ISO or regional standards (e.g., CEN) or national standards (e.g., JIS). Reference materials can be assigned to a grade, and "Data of pure

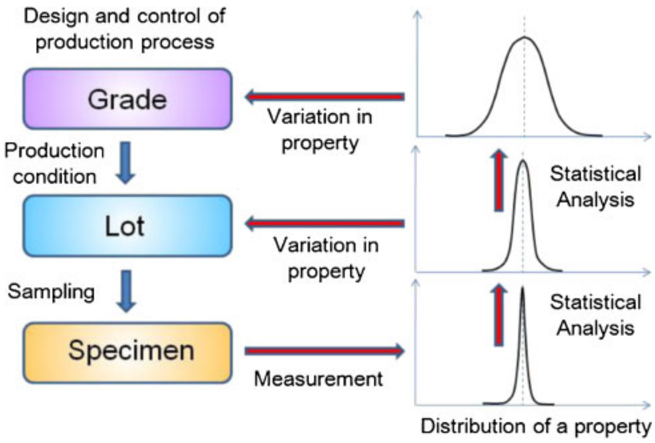

Fig. 3. (Color online) Evaluation process of material property from measured data for specimens to lot and grade.

single crystals" can also be stored at this level if they can be defined quantitatively by the maximum density of impurities, that of defects and dislocations.

Examples of commercial grade names are Corning Pyrex® 7740 or Toray TORACA® T300 carbon fiber. An example of document standard names is SUS310 stainless steel. Examples of reference materials are NMIJ RM1103-a for thermal expansion coefficient, NMIJ RM1201-a for thermal diffusivity, and NMIJ RM1301-a for the thermal diffusivity of thin films.

Material producers specify and control the process for a grade of material and supply the material to users with the grade name. Generally, they release datasheets corresponding to the grade for their products.

It is difficult to obtain complete information on the scattering of material properties under a specified process corresponding to the grade name since complete sampling cannot be made from the entire production by a specified process that has been and will be operated for a long time in the future. It is important to point out that the mean is not sufficient for the determination of material properties when the property varies in the lot because of nonuniformity or under a specific production process that varies or fluctuates over a long time of operation, as shown in Fig. 3.[33]

2.3 Complementary role of measurements and data for thermophysical properties

Since the physical properties of solid materials are generally dependent on the material structure which shows diversity and changes depending on the synthesis method and process, it is not easy to find the material of interest from a material name presumed in databases and handbooks. It is often more difficult, expensive, or time-consuming to characterize a specimen quantitatively to specify property values from databases or handbooks than from real measurement of the specimen, especially in the case of transport properties.

For example, the transport properties of graphite, such as electrical resistivity, thermal conductivity, and thermal diffusivity, are dependent on the microscopic structure of graphite over a wide temperature range. These transport properties are more dependent on the structure, which is changed by heat treatment, than on the chemical composition, i.e., amount of impurity. Although the structure of these graphite materials can be characterized by X-ray diffraction (XRD) analysis, thermal diffusivity is more dependent than the XRD spectrum on heat treatment temperature.[34] Thus, it is difficult to predict thermal diffusivity from the XRD spectrum. A similar example is reported for the thermal diffusivity of molybdenum thin films synthesized by sputtering.[10,11]

On the other hand, thermodynamic properties, such as specific heat capacity and enthalpy, are not as sensitive to structures as transport properties. Characterizations for the prediction of thermodynamic properties are much easier than those for the prediction of transport properties.

2.4 Network database system for thermophysical property data developed by NMIJ/AIST

2.4.1 Thermophysical property database developed by NMIJ/AIST

A new type of database, which is optimized to store and evaluate traceable thermophysical property data with uncertainty and material identification based on hierarchical classification, has been developed by NMIJ. The criteria of material classification and material information stored at each hierarchy are shown in Table I. Grade, lot, and specimen in a hierarchical classification are defined according to §2.2. In Fig. 4, fused silica is assigned to Material Class 3 under SiO_2, i.e., "silicon dioxide" under oxide materials in Material Class 1 of the Ceramics and Glasses group.

This classification approach can be harmonized to a future universal guideline for material classification, which is expected to be established under the consensus of the material community.

2.4.2 Network database system

In this paper, "Network database system" means a system whose function provides the simultaneous connection of databases using both exclusive browsing software and the Internet.

Database servers with a common database system have been constructed at both AIST and the Japan Society of Thermophysical Properties (JSTP). These databases can be accessed using the browsing software called TPDS-web and InetDBGV via the Internet. Since these databases have common database system, the function of cross search among databases is also available on the InetDBGV and TPDS-web. A powerful, interactive, and graphical user interface is available by installing InetDBGV to the user's PC. Figure 5 shows a screenshot of the temperature dependence curves for the thermal diffusivity of metals displayed on InetDBGV. Since the TPDS-web runs on general web browsers, the database can be accessed easily and quickly using the TPDS-web.

3. Ultrafast Laser Flash Method for Measuring Thermophysical Properties of Thin Films

NMIJ/AIST developed an "ultrafast laser flash method" that can measure the thermal diffusivity of metallic thin films from several 10 nm to several micrometers thick on a transparent substrate in the thickness direction by picosecond/nanosecond light pulse heating.[4,7–9,36] This has

Table I. Material classification for homogeneous material.

Hierarchy level	Material information used in classification	Examples of material folder name
Domain	Phase at room temperature for majority of materials in group	Fluid, Solid and Melts
Group	Group of periodic table to which element of simple substance belongs. Combination of groups to which elements of chemical compound belong[35]	Metal, Ceramics and glasses, Semiconductor
Material Class 1	Name of material group having similar material character and similar behavior of properties	Noble metal, Nitride, Oxide
Material Class 2	Substance name, Chemical formula, CAS registry number, IUPAC Name	Aluminum, Gold, Silicon oxide
Material Class 3	Material name, crystal structure, phase, application field, form	Silicon single crystal, Silicon poly -crystal, fused silica
Material Class 4 (Grade)	Grade of commercial material, Material standard, chemical composition, RM/CRM code, main material manufacturing process such as equipment	JIS SUS 310, Pyrex 7740, fused silica_Grade1
Material Class 5 (Lot)	Lot name, information related to fine material manufacturing process such as pre- and post -processes	Fused silica_G1, lot1
Material Class 6 (Specimen)	Specimen name, specimen shape and size	Fused silica_G1, lt1, specimen1

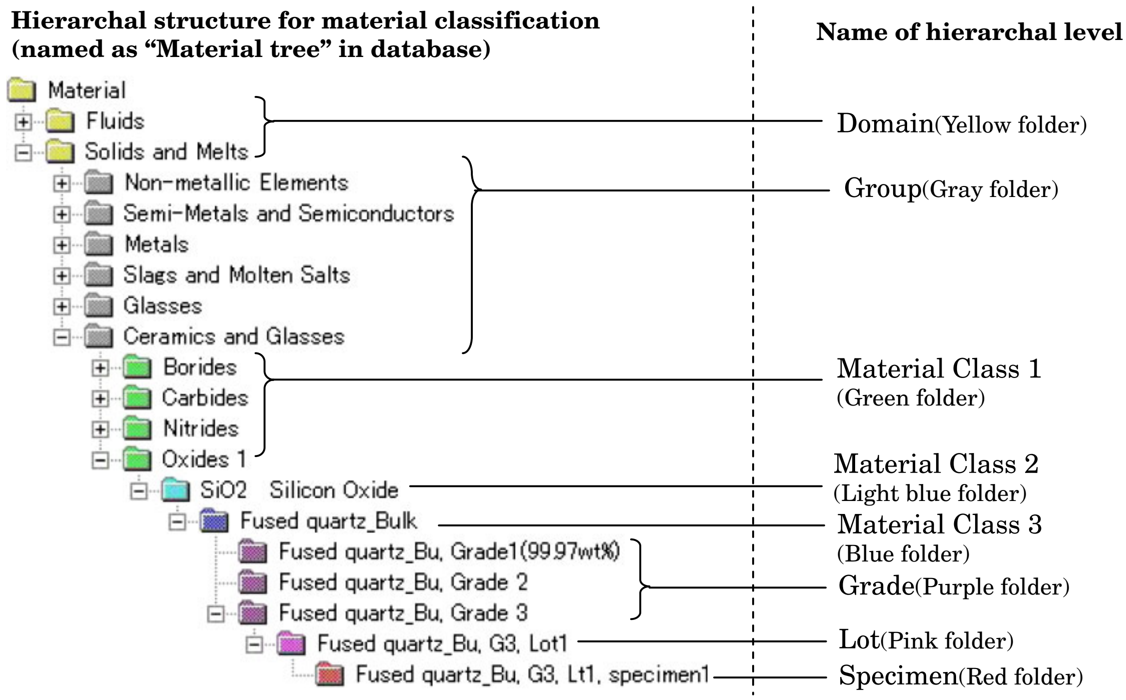

Fig. 4. (Color online) Example of material tree for homogeneous material.

the same configuration as the conventional laser flash method. The configuration is also called the rear-face heating/front-face detection (RF)-type pulsed light heating thermoreflectance method. NMIJ also improved the front-face heating/front-face detection (FF)-type pulsed light heating thermoreflectance method.[6,37–41]

In the case of a transparent oxide thin film, the film is coated with metallic thin films synthesized on a transparent substrate, as shown in Fig. 6.

The analysis of heat diffusion across a three-layer thin film on a substrate must be considered in the calculation of the thermal diffusivity of the thin film coated with metal thin films. Since the heating area is sufficiently large compared with the film thickness, heat diffuses from the rear surface to the front surface one-dimensionally. The one-dimensional heat diffusion of multilayer films can be calculated by the impulse response function method, which is a general technique for analyzing heat diffusion.[8,9,42,43)]

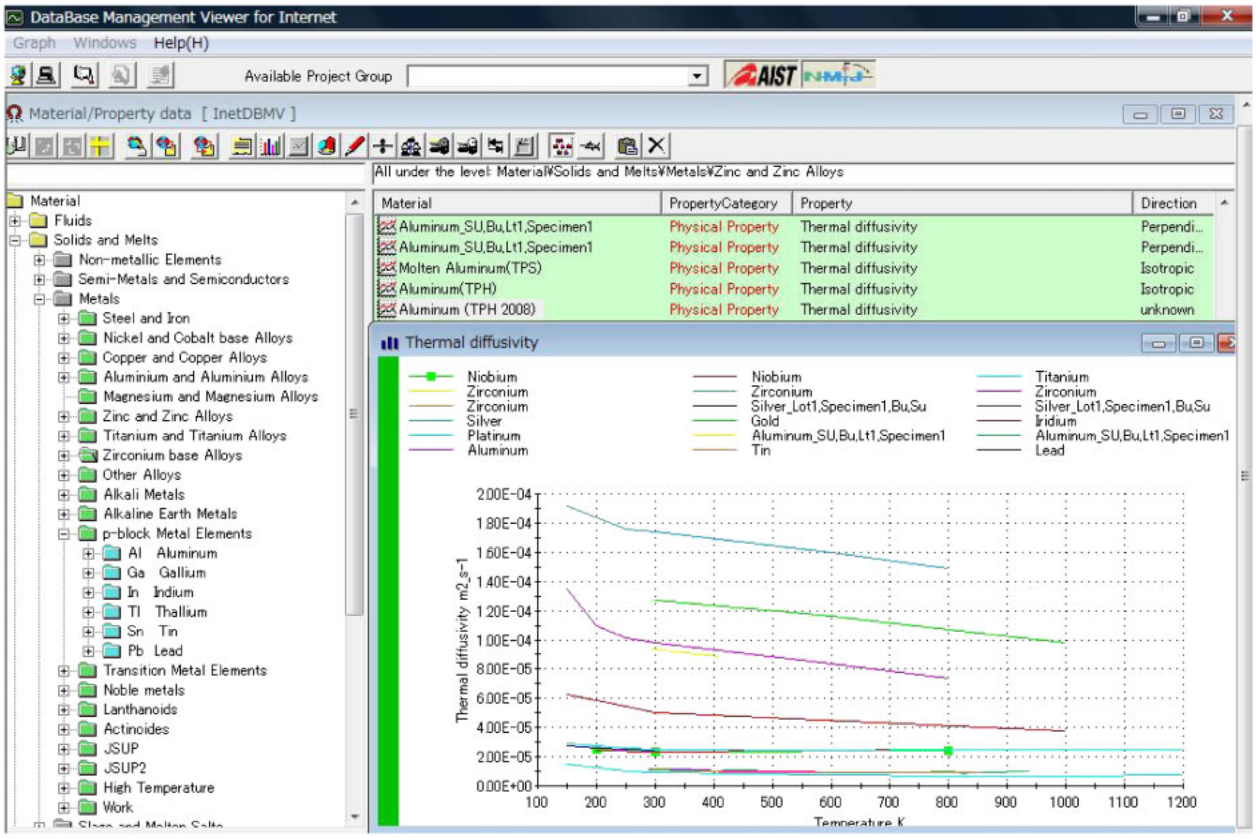

Fig. 5. (Color online) Screenshot of temperature dependence curves for thermal diffusivity of metals displayed by graphical user interface of network database system for thermophysical property data.

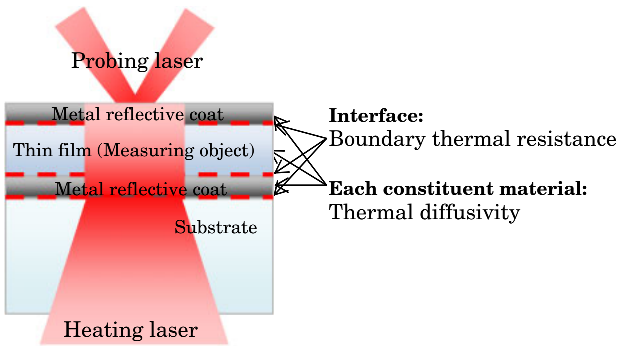

Fig. 6. (Color online) Schematic diagram of ultrafast laser flash method (RF-type thermoreflectance measurement).

4. Extension of Network Database System for Thermophysical Property Data to Store Data of Thin Films

4.1 New material classification rule and material information storage system

As stated in the previous section, thin-film specimens used in the ultrafast laser flash method usually consist of multi-layers. Since the material classification of the current thermophysical property database is designed to store information on homogeneous materials, the database must be highly advanced to store composite materials such as laminar, dispersed, and fabric materials, as shown in Table II. Therefore, a new domain "composite" has been created under which materials are classified on the basis of morphology. At the second hierarchy level of "Group", composites are classified into "laminar", "dispersed", "fabric" and others. In the case of a laminar composite, the combination of constituent layers and their compositions and dimensions are stored in folders of lower classes.

4.2 Preservation strategy of specimens

As stated in §2.1, the properties of solid materials are determined not only by their composition but also by their structure on the nanoscale, microscale, and macroscale. The structures and properties of thin films are different not only from those of the bulk materials of the same condition but also from each other since thin films of the same composition can sometimes be synthesized by processes, such as physical vapor deposition methods, chemical vapor deposition methods, dip-coating, and spin-coating, completely different from those for bulk materials. Therefore, the dynamic ranges of the thermophysical properties of thin films are larger than those of the bulk materials, and it is very difficult to sufficiently characterize them to correlate with their properties, particularly in the case of transport thermophysical properties, such as thermal conductivity and thermal diffusivity.

A method of identifying and expressing thin film structures and characters has not been sufficiently established to enable the storage of thermophysical property data of thin films into the database as universal information. For example, there has been no systematic investigation that quantitatively correlated thermal transport properties with TEM images or X-ray diffraction data.

Table II. Material classification for composite materials.

Hierarchy level	Material information used in classification	Example of material folder name
Domain	Classified by fundamental attribute: If material flows (liquids) or keeps its shape constant. If material is homogeneous or inhomogeneous.	Composite
Group	Geometry of composite	Dispersed, laminar, fabric
Material Class 1[a]	Detailed information of laminar structure	Multilayer thin films on substrate, Thermal barrier coating on super alloy
Material Class 2[a]	Substance name of each layer corresponding to "solid and melts" domain	Oxides and metals multilayer thin film on glass substrate, oxide thin film on silicon substrate
Material Class 3[a]	Material name of each layer corresponding to "solid and melts" domain	Mo/Al_2O_3/Mo thin film on fused silica substrate, TiN thin film on synthesized quartz substrate
Material Class 4[a] (Grade)	Explained in §2.2.	
Material Class 5[a] (Lot)		
Material Class 6[a] (Specimen)		

a) Classification under material class1 is explained under the assumption that the structure of the composite is laminar.

Table III. Record items for thermophysical property and material character of thin film specimen.

Category of data	Kind of specimen	Recorded items
Thermophysical property data	Single-layer thin film	Raw signal data, curve fitting profile, heat diffusion time, unique code
	Multilayer thin film	Raw signal data, areal heat diffusion time, boundary thermal resistance, volumetric heat capacity of constituent materials, thickness of each layer, unique code
Optical property data	Single-layer thin film and multilayer thin film	Reflectivity, transmissivity, refractive index, absorption coefficient, thermoreflectance coefficient
Material data	Single-layer thin film and multilayer thin film	Film thickness, XPS data, XRD data, RBS data, SEM image and TEM image, crystal grain size, concentration of impurities

To solve the fundamental problem of the thermophysical property database for thin films, the current approach of NMIJ is to store all specimens whose thermophysical properties are measured and stored into the database. Then, further characterization of the specimens is possible as needed.

In the database, the identification code including the substance name is given for the systematic management of both data stored in the database and real specimens in storage. For example, the code of a titanium nitride thin film stored in AIST specimen storage is "TiN_TF-AIST-No. 1".

4.3 Record items for thermophysical property data of thin-film specimens

The thin film database is originally designed to store data observed by the ultrafast laser flash method and information for identifying and characterizing the specimen for the method.[7–9,36] The database can also store thermophysical property data measured by any measurement method such as 3-omega,[44] 2-omega,[45,46] and a variety of other photothermal methods.

In the case of the ultrafast laser flash method, the properties listed in Table III are expected to be stored. Optical properties at the wavelength of the lasers for heating and thermoreflectance thermometry are required for the data analysis of the ultrafast laser flash method. Recorded items related to material characterization are useful to analyze the relationship between thermophysical properties and material characters.

Examples of how to obtain and store the thermophysical property data for a thin film and how to use the new classification rule for a thin-film specimen are shown in the following section.

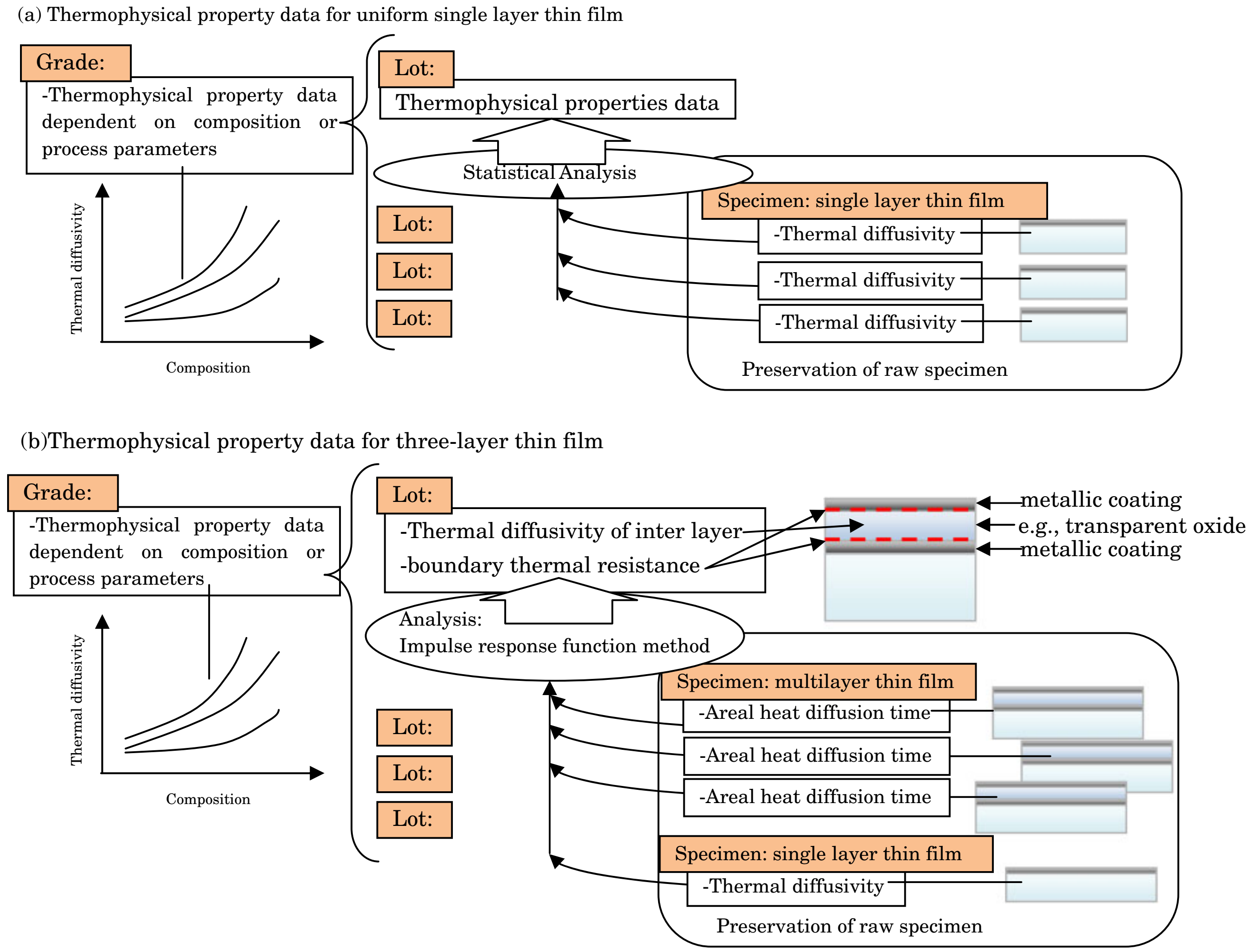

Fig. 7. (Color online) Flow of data analysis from measurements for set of specimens to lot, and grade.

5. Thermophysical Property Data of Thin Films

Since multilayer thin films are a combination of layers, there may be various approaches to determining the thermophysical properties of thin films. Here, three of them in which thermophysical properties are measured using the ultrafast laser flash method[7–9,36] are introduced where the configurations of the specimen are shown in Fig. 7.

5.1 Thermophysical property measurement of single-layer thin film

One example is thermophysical property measurement for a single-layer thin film on a substrate, as shown in Fig. 7(a). The thermal diffusivity of the thin film can be determined by fitting the following analytical solution derived from the response function method[8,9,42] and mirror image method[42] to the observed temperature response curve:

$$T(t) = \Delta T \cdot \left[1 + 2\sum_{n=1}^{\infty}(-1)^n \exp\left(-(n\pi)^2 \frac{t}{\tau}\right)\right] = \frac{2}{b\sqrt{\pi t}}\sum_{n=0}^{\infty}\exp\left(-\frac{(2n+1)^2\tau}{4t}\right). \quad (1)$$

Here, T is the temperature, b is the thermal effusivity, t is the time, $\Delta T = Q/C$, Q is the total energy absorbed by the thin film per unit area and C is the heat capacity of the thin film per unit area.

For sample preparation, a titanium nitride (TiN) thin film of 681 nm thickness was deposited on a synthesized quartz substrate by reactive dc magnetron sputtering under an one process parameter set. Figure 8 shows an example of the material folders created in order to classify a specimen in the database. At the grade level, "TiN on synthesized quartz substrate, Grade1" is created under the material class 3 folder, i.e., "TiN thin film on glass substrate" in the "composite" domain. As well as the creation of a material folder at the grade level, the material folder at the lot level, i.e., "TiN on synthesized quartz substrate, G1, lot1", is created under the material folder of grade. Finally, the material folder assigned to this specimen is named "TiN on synthesized quartz substrate, G1, lt1, specimen1" and created under the material folder of lot. Information indicating that the specimen consists of a TiN thin film of 681 nm thickness and synthesized quartz is stored in "composite material structure information" as material information of the specimen folder.

The thermal diffusivity of this specimen was measured using a nanosecond thermoreflectance system of the ultrafast laser flash method.[41,47] After the measurement, the specimen was stored in a tray so that the specimens can be

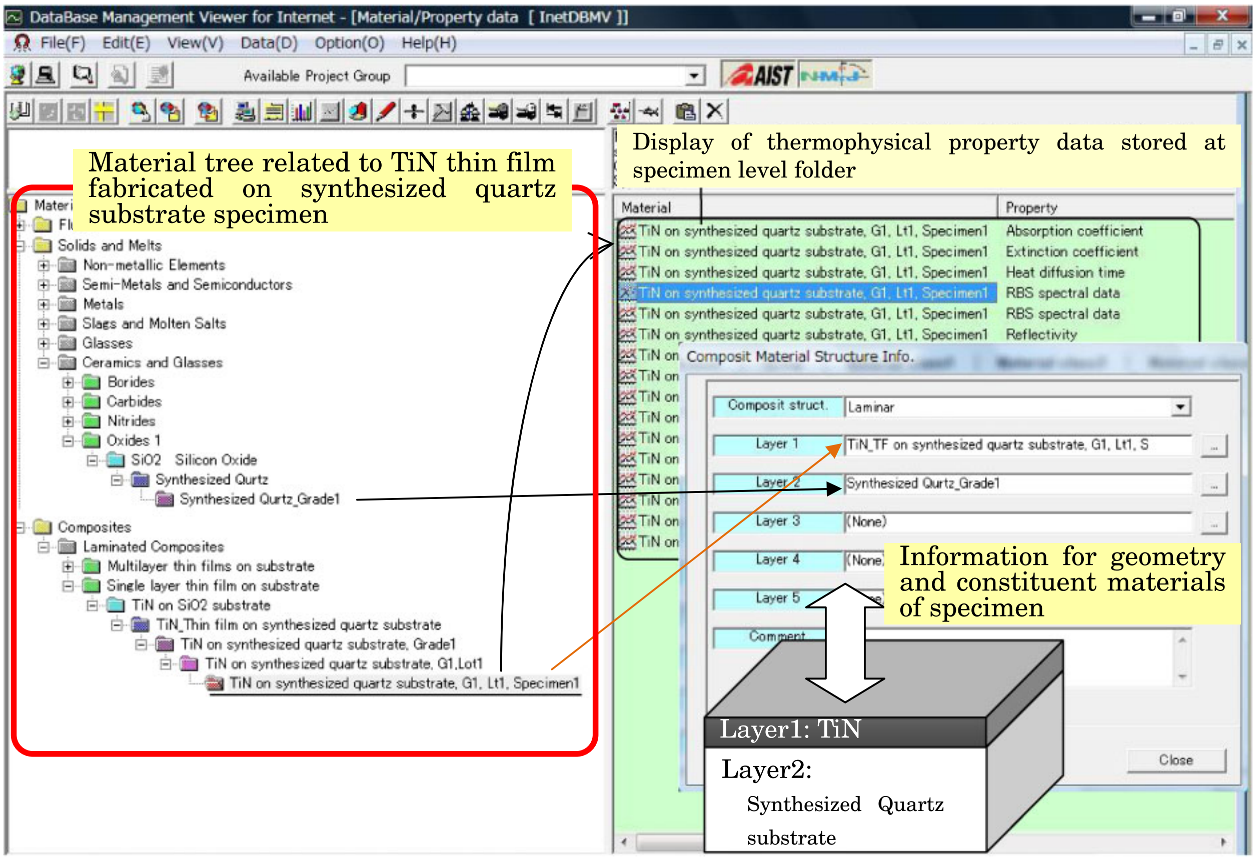

Fig. 8. (Color online) Example of material tree and material information related to TiN on synthesized quartz substrate.

characterized at any time using an analysis technique according to the requirement. The observed thermoreflectance signal stored in a database and a theoretical curve (1) fitted to the thermoreflectance signal are shown in Fig. 9. The calculated heat diffusion time was 141 ns from curve fitting. Thermal diffusivity was calculated as 3.3×10^{-6} $m^2 \cdot s^{-1}$ by dividing the square of the film thickness (681 nm) by the heat diffusion time. The data of the thermal diffusivity of this specimen is stored in the material folder “TiN on synthesized quartz substrate, G1, lt1, specimen1” at the specimen level. Usually, after repeating thermophysical property measurement for a set of specimens, the thermophysical property data of the lot is calculated using statistical analysis to a set of thermophysical property data from TiN thin film specimens of the same lot as shown in Fig. 7(a).

5.2 Thermophysical property measurements for three-layer thin films with boundary thermal resistance

The second approach is to determine both thermal diffusivity and boundary thermal resistance simultaneously from a set of specimens by the areal heat diffusion time method, as shown in Fig. 7(b). This approach can be applied to the measurement of thin films that cannot be measured as a single layer because of their transparency or low sensitivity to thermoreflectance temperature detection.

5.2.1 Areal heat diffusion time

Areal heat diffusion time is defined as the area surrounded by the horizontal line at the height of the maximum temperature rise and the transient temperature response

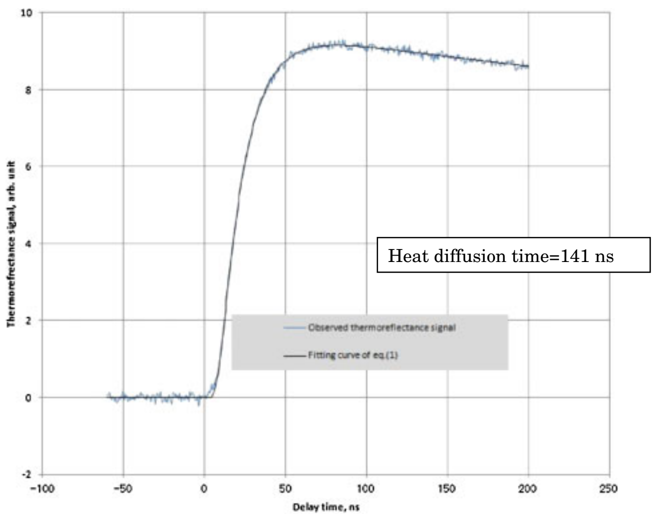

Fig. 9. (Color online) Theoretical curve and thermoreflectance signal of TiN thin film on synthesized quartz substrate.

curve at the rear face after pulse heating, as shown in Fig. 10.[8,9,48,49] On the basis of the response function method, areal heat diffusion time is calculated using eq. (2) when temperature is normalized by the maximum temperature rise $T_{max} = 1/(b\sqrt{\tau})$.

Areal heat diffusion time[8] is defined as

$$A = \int_0^\infty [1 - b\sqrt{\tau} \cdot T_r(t)]\,dt = \lim_{\xi \to 0}\left[\frac{1}{\xi} - b\sqrt{\tau} \cdot \tilde{T}_r(\xi)\right], \quad (2)$$

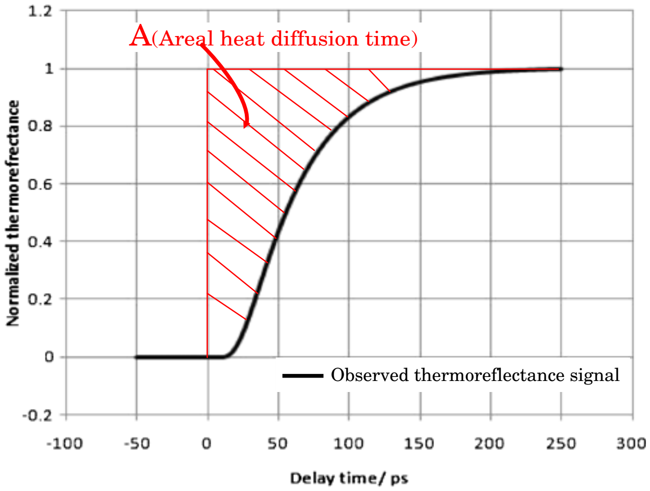

Fig. 10. (Color online) Area surrounded by maximum temperature rise line and temperature response at specimen rear face after pulse heating.

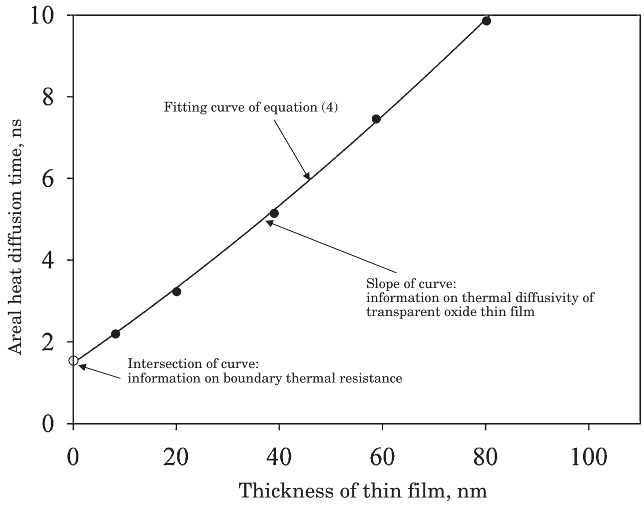

Fig. 11. Schematic diagram of curve fitting of eq. (4) and information on thermophysical properties derived from fitting curve.

where A is the areal heat diffusion time, ξ is the Laplace parameter, and $\tilde{T}_r(\xi)$ is the Laplace transform of $T_r(t)$ defined by

$$\tilde{T}_r(\xi) = \int_0^\infty T_r(t)\exp(-\xi t)\,dt. \tag{3}$$

Here, t: time, ξ: Laplace parameter, d: thickness, α: thermal diffusivity, b: thermal effusivity, $T(t)$: temperature, $\tilde{T}(\xi)$: Laplace transform of temperature, $q(t)$: heat flux density, $\tilde{q}(\xi)$: Laplace transform of heat flux density, and $\tau = d^2/\alpha$: heat diffusion time across film or layer.

5.2.2 Areal heat diffusion time for three-layer thin film

In the case that the three-layer thin film consists of a transparent oxide thin film and coating layers that are deposited at the same film thickness, the same material character, and the same thermophysical property between coating layers, the areal heat diffusion time of a three-layer thin film[8] is

$$A = \frac{b_m\sqrt{\tau_m}\left(\frac{4}{3}\tau_m + \tau_f\right) + b_f\sqrt{\tau_f}\left(\tau_m + \frac{1}{6}\tau_f\right) + 2b_m b_f R_b \tau_m^{1/2}\tau_f^{1/2} + 2R_b b_m^2 \tau_m}{2b_m\sqrt{\tau_m} + b_f\sqrt{\tau_f}}$$
$$= \frac{\left(c_m d_m + \frac{4}{3}c_f d_f\right)\frac{d_m^2}{\alpha_m} + \left(\frac{c_m^2 d_m^2}{c_f d_f} + c_m d_m + \frac{1}{6}c_f d_f\right)\frac{d_f^2}{\alpha_f}}{2c_m d_m + c_f d_f} + \frac{2R_b c_m d_m(c_f d_f + c_m d_m)}{2c_m d_m + c_f d_f}, \tag{4}$$

where R_b is the boundary thermal resistance between the metallic coating layer and the transparent oxide thin film, c is the specific heat capacity per unit volume, and the subscripts m and f denote the metallic coating layer and the transparent oxide thin film, respectively.

If the specific heat capacities, c_m and c_f, the film thicknesses d_m and d_f, and the thermal diffusivity of the coating layer, α_m, are known in eq. (4), the unknown parameters are only the thermal diffusivity of a transparent oxide thin film, α_f, and the boundary thermal resistance R_b. Therefore, by fitting eq. (4) to the areal heat diffusion time of a set of specimens belonging to the same lot but having different thicknesses of the transparent oxide thin film, as shown in Fig. 11, the boundary thermal resistance and thermal diffusivity of the transparent oxide thin film are derived from the intersection of the zero thickness and the slope of the fitting curve, respectively.

5.2.3 Example of thermophysical property measurement of three-layer thin films

This approach was applied to the study of amorphous Al_2O_3.[50] For sample preparation, a series of three-layer thin films in which the thickness of Al_2O_3 varies from 0.5 to 100 nm on a fused silica substrate and a Mo single layer thin film of 70 nm thickness on a fused silica substrate were synthesized in the same lot. In the database, material folders for a set of specimens are made under the material folder "Al_2O_3 coated with Mo on fused silica substrate, G1, Lot1" in a "composite" domain as shown in Fig. 12.

Before data analysis, the thermal diffusivity of a Mo single-layer thin-film specimen was measured using the ultrafast laser flash method by picosecond pulsed light heating[40] and found to be $2.1 \times 10^{-5}\,m^2{\cdot}s^{-1}$. The data is stored in the material folder "Mo on fused silica substrate, Specimen1" shown in Fig. 12. Literature values for the bulk materials are used for the specific heat capacities of Mo and Al_2O_3. The total thickness of the three-layer thin film is measured using a surface profiler, and the thickness of each layer is estimated from the deposition rate.

The areal heat diffusion time of each sample is measured by the ultrafast laser flash method[41,47] and the data are stored in material folders from "Al_2O_3 coated with Mo on fused silica substrate, G1, Lt1, Specimen1" to "Al_2O_3 coated with Mo on fused silica substrate, G1, Lt1,

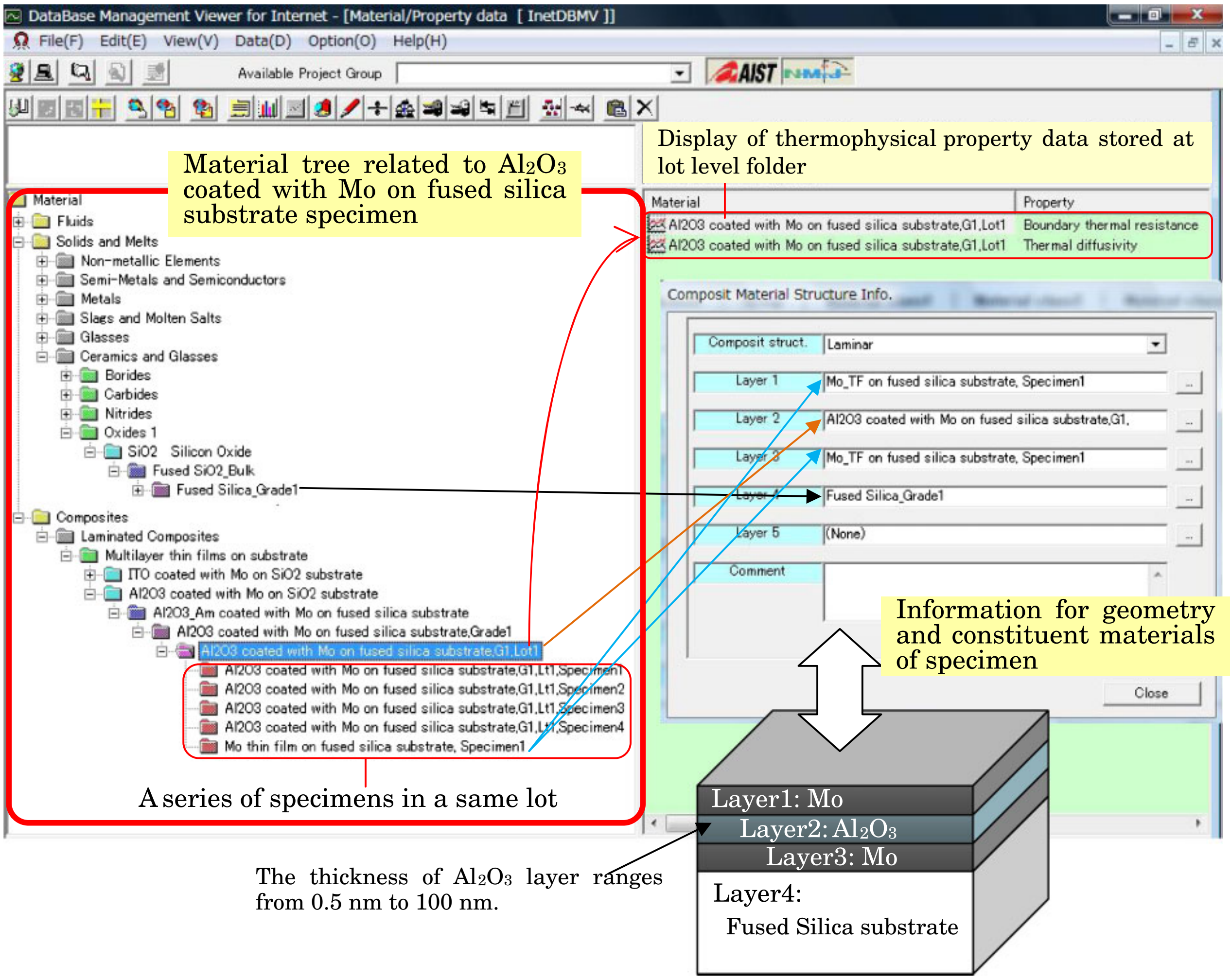

Fig. 12. (Color online) Example of material tree and material information related to Al_2O_3 coated with Mo on fused silica substrate.

Specimen4". The thermal diffusivity of Al_2O_3 and the boundary thermal resistance between a Mo thin film and an Al_2O_3 thin film are calculated by fitting the curve of eq. (4) to a set of areal heat diffusion times from a set of specimens in the same lot, as shown in Fig. 11.

Consequently, the thermal diffusivity of a set of amorphous Al_2O_3 thin films and the boundary thermal resistance between an amorphous Al_2O_3 thin film and a Mo thin film are $9.5 \times 10^{-7}\,m^2{\cdot}s^{-1}$ and $1.5 \times 10^{-9}\,m^2{\cdot}K{\cdot}W^{-1}$, respectively. The calculated data are at the lot level and stored in the "Al_2O_3 coated with Mo on fused silica substrate, G1, Lot1" folder at the lot level of the database shown in Fig. 12.

5.3 Dependence of thermal diffusivity of ITO thin films on oxygen flow ratio at sputtering process

This database can systematically store the dependences of the thermophysical properties of thin films on process parameters.

5.3.1 Sample, lot and grade

For sample preparation, three-layer thin films were fabricated on a fused silica glass substrate by the dc magnetron sputtering method.[51] The nominal thickness of the ITO layer was 200 nm, and the ITO film was transparent to the laser beam employed in the thermoreflectance measurement system so that the Mo layers whose thickness was 70 nm were deposited on both sides of the ITO film as reflective coating layer. An ITO layer was deposited with varying oxygen flow ratio between 0.0 and 5.0%. Here, oxygen flow ratio means the ratio of oxygen flow rate to the sum of oxygen flow rate and argon flow rate. In the fabrication process, oxygen flow ratio was controlled to adjust the material characters, such as carrier density, of ITO films.

5.3.2 Creation of material folders

A material folder at the grade level, "ITO coated with Mo on fused silica glass substrate, Grade1" is created under the material folder at the material class 3 level, "ITO_Pc coated with Mo on fused silica glass substrate", in the database, as shown in Fig. 13. A grade consists of 6 lots and a set of materials of different process parameters were assigned to different material folders at the lot level. A series of fabricated specimens of the same lot are assigned to material folders at the specimen level. One lot consists of three specimens. Two of them are an "ITO coated with Mo on fused silica glass substrate" specimen and a "Mo on fused silica glass substrate" specimen for the measurement of thermophysical properties. Another is an "ITO on fused silica glass substrate" specimen for the measurement of electrical resistivity.

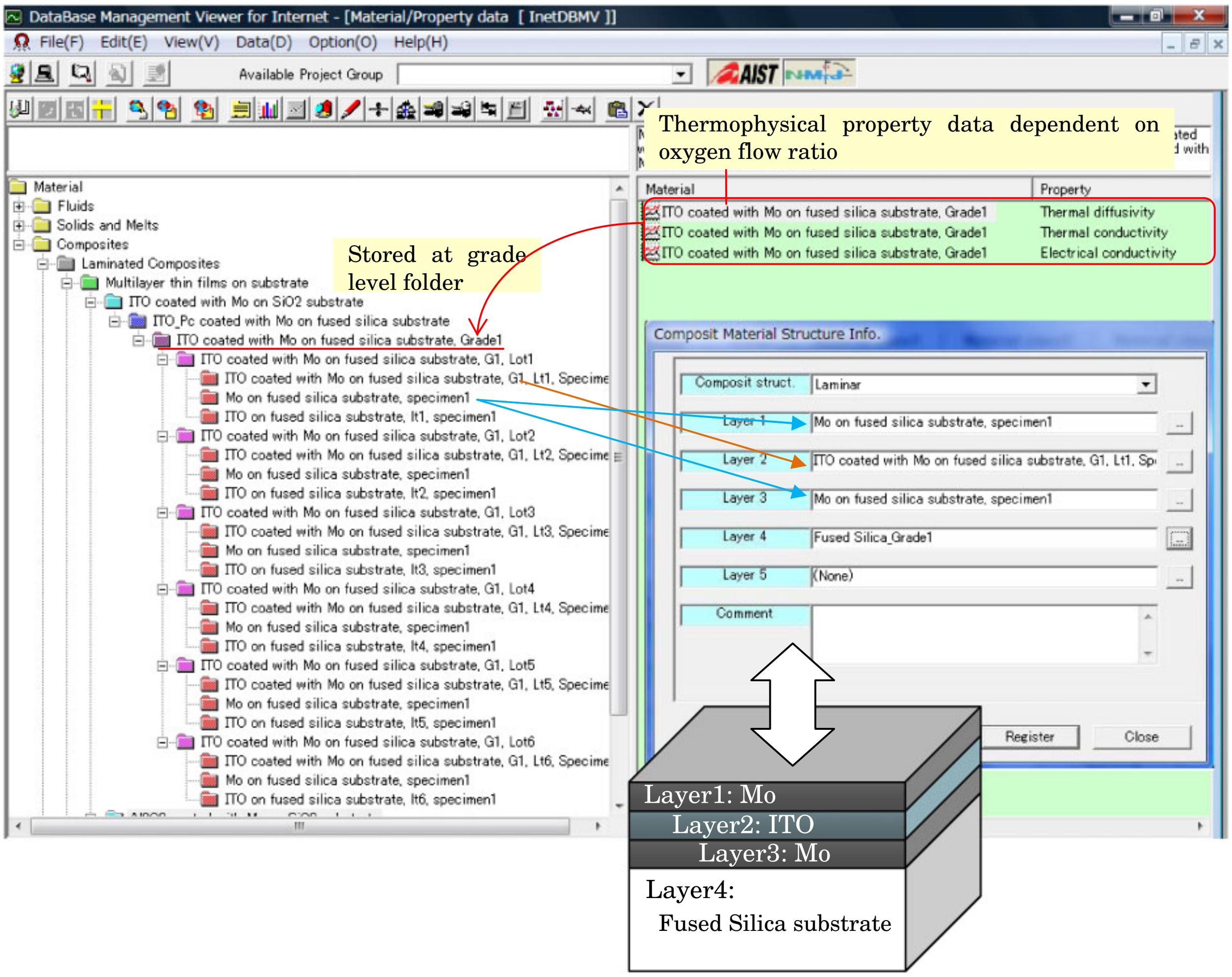

Fig. 13. (Color online) Example of material tree and material information related to ITO coated with Mo on fused silica substrate.

5.3.3 Data analysis

In the case of an ITO film of thickness 200 nm, the boundary thermal resistances between layers are negligible compared with the total thermal resistance across the three-layer thin film. R_b in eq. (4) can be assumed to be zero. The thermal diffusivity of the ITO thin film is calculated from a couple of three-layer thin film specimens and Mo single layer specimens of the same lot because the unknown parameter is only the thermal diffusivity of the ITO thin film under this assumption. The data of the thermal diffusivity of ITO thin films are systematically stored in each lot-level folder depending on the oxygen flow ratio.

5.3.4 Example of thermophysical property measurement

The dependence of the thermal diffusivity of ITO films on oxygen flow ratio is shown in Fig. 14 and stored in the material folder "ITO coated with Mo on fused silica substrate, Grade1" at the grade level of the database shown in Fig. 13. The dependence of thermophysical property data on process parameters at the grade level are synthesized from a set of thermophysical property data at the lot level under a specified grade. A maximum thermal diffusivity of $2.27 \times 10^{-6}\ \mathrm{m^2 \cdot s^{-1}}$ was observed for the ITO coated with a Mo three-layer thin film specimen at 0.5% oxygen flow ratio, it decreases with an increase in oxygen flow ratio in the range from 1.0 to 5.0%.

Figure 15 shows the correlation between thermal conductivity and electrical conductivity.[51] Here, electrical conductivity was calculated from the electrical resistivity measured by the four-probe method. For electrical resistivity measurement, a series of ITO monolayers deposited on a fused silica substrate specimen were synthesized. Corresponding material folders, e.g., "ITO on fused silica substrate, lt1, specimen1", are created at the specimen level, as shown in Fig. 13 and the electrical resistivity data of each specimen are stored in these folders. Thermal conductivity was calculated by multiplying thermal diffusivity by specific heat capacity per unit volume.

In Fig. 15, a set of plots can be fitted by a linear line according to the Wiedemann-Franz law. This result demonstrates that the change in thermal conductivity can be attributed to the change in thermal conductivity carried by free electrons.[51]

These data correlating thermal conductivity and electrical conductivity with oxygen flow ratio shown in Fig. 15 are stored in material folders, e.g., "ITO coated with Mo on fused silica substrate, Grade1", at the grade level, similarly to thermal diffusivity.

6. Conclusions

The thermophysical property database of NMIJ was evolved to store comprehensive information on thin films. To solve

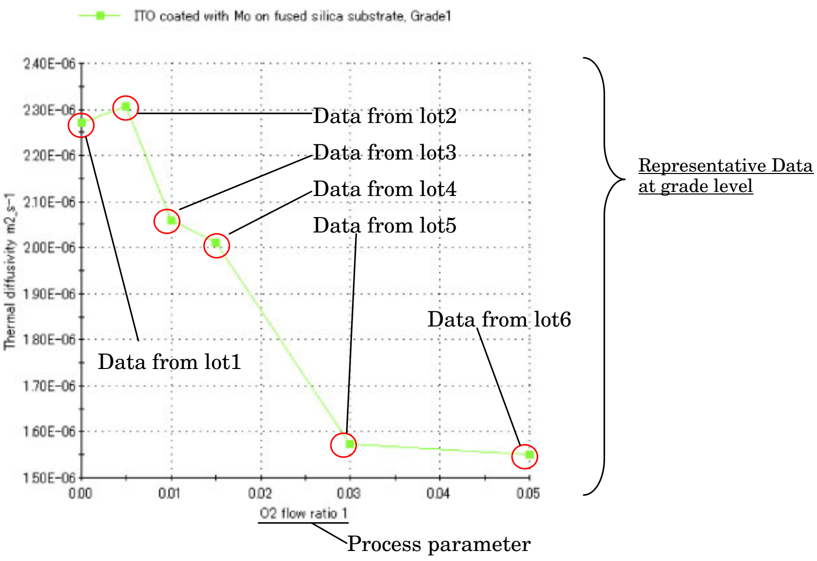

Fig. 14. (Color online) Dependence of thermal diffusivity on O_2 flow ratio for ITO coated with Mo on fused silica substrate.

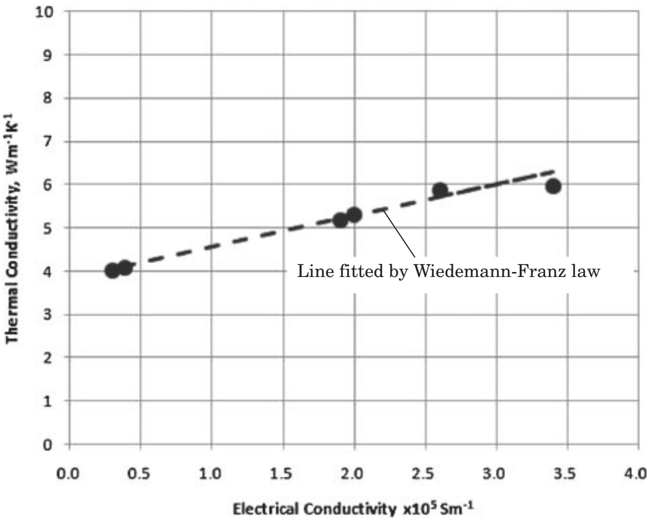

Fig. 15. Correlation between thermal conductivity and electrical conductivity of ITO film.

the difficulty in the identification and characterization of thin films, a strategy was introduced to preserve the specimens, whose thermophysical properties were measured and synchronized with the records stored in the database.

The design and structure of the database were improved for the systematic management of the property data of and material information on multilayer thin-film specimens. In the case of thermophysical property measurement using the ultrafast laser flash method, record items including boundary thermal resistance, observed thermoreflectance signal, and areal heat diffusion time, are listed.

The storage of thin-film data obtained using the ultrafast laser flash method and user interface is demonstrated for a set of thermophysical property data of a "TiN single layer thin film on a synthesized quartz substrate", "Al_2O_3 coated with Mo three-layer thin films on a fused silica substrate" and "ITO coated with Mo three-layer thin films on a fused silica substrate".

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