

## Japanese Research on Thermophysical Properties of Solids

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The research activities on thermophysical properties in many engineering fields in Japan have been promoted for the past 10 years. In this paper, the outlook is reviewed on recent research work on thermophysical properties, mainly on thermal conductivity and thermal diffusivity of solids, in Japan. The research activities are described in such areas as measuring methods, metals, nuclear fuels and ceramics, building materials and insulators, soils and rocks, energy storage materials and compound materials, and dispersed materials.

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**KEY WORDS:** solids; thermal conductivity; thermal diffusivity; thermophysical properties.

### 1. INTRODUCTION

In the past, research on thermophysical properties of solids in Japan was sporadically carried out in many engineering fields where needed for specific problems. However, because of recent advances in applications of heat transfer engineering and of new materials, the data on thermophysical properties have become insufficient. Thus research activities on thermophysical properties in many engineering fields have been promoted for the past ten years. In particular, since the Japan Society of Thermophysical Properties was established in 1980 and began to hold annual symposia, which have been supported by more than 20 domestic academic societies, an opportunity to make a clear and close communication among various research workers has been offered; as a result, the interest of related research has been stimulated. In the following section, the outlook on the recent work in thermophysical properties of solids in Japan will be discussed, mainly on thermal conductivity and thermal diffusivity.

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## 2. CURRENT RESEARCH ACTIVITY

### 2.1. Measuring Methods

Research and development on the methods and apparatus to measure thermal conductivity and thermal diffusivity have been carried out by various workers, although the conventional methods by steady heat flow and by laser pulse are still used in many cases. Katayama et al. [1] studied a method to measure thermal diffusivity and heat carrying capacity of anisotropic materials simultaneously with temperature response of a specimen by stepwise heating. Kumada et al. [2] theoretically investigated the error in thermal diffusivity measurement due to nonuniform distribution of the energy intensity in a laser beam in a pulse heating method. Takahashi et al. [3] used a glassy carbon plate 0.2 mm thick as an absorbing disk bonded by grease to the specimen in order to obtain the always constant energy absorption in measuring heat capacity by a pulse heating method. Kobayasi et al. [4, 5] investigated a method to measure thermal diffusivity and specific heat simultaneously by radiative heating with a single rectangular pulse on a specimen with radiation heat loss from the specimen at high temperatures. This is an extended method of stepwise heating which they had developed previously [6, 7].

Araki et al. [8] analyzed the error due to the time lag of temperature response in measuring thermal diffusivity by a laser pulse method and discussed the permissible limits of the time lag. Iida et al. [9, 10] developed a method for the simultaneous determination of thermal diffusivity and thermal conductivity of solids under arbitrary heating and radiating conditions. The method utilizes the principle of the Laplace transform method. Takegoshi et al. [11] analyzed a hot ribbon probe method for thermal conductivity measurements which was extended from a hot wire method. Further, they studied theoretically and experimentally the principle to determine thermal conductivities of three principal axes in orthogonal anisotropic materials [12]. Matsuda et al. [13] proposed an effective method of measuring thermal conductivity with reaction, applying the principle of differential thermal analysis. As a result, it was found that the thermal conductivity of solid materials in the course of reaction was smaller than that in pre- and postreaction states, and that the thermal conductivity of some organic materials in the course of melting and/or solidification yielded almost a constant value approximately equal to that in liquid state.

### 2.2. Metals

Takahashi et al. [14] measured thermal diffusivities of tantalum and nickel by isoperibol, laser pulse apparatus within the range from room temperature to 1300 K. Suhara et al. [15] measured thermal diffusivity, specific heat, and thermal conductivity of three kinds of Ni-base and three

kinds of Co-base superalloys for an advanced high efficiency gas turbine, by the stepwise heating method proposed by Kobayasi et al. [6]. The temperature was in the range from room temperature to roughly 1000 K. Sugawara [16] also measured thermal conductivity of Ni-base alloys which were used for gas turbine blades. Azumi et al. [17] measured the thermal diffusivity of metallic thorium in the range from 100 to 500 K by the laser pulse method. Kobayasi et al. studied the temperature dependencies of thermal diffusivity, specific heat, thermal conductivity, and electric conductivity of ferrous binary alloys which were carefully prepared to exclude impurities. They also presented the effect of concentration of alloying compositions on the thermophysical properties. These values were measured by the stepwise heating method within the range from room temperature to 1473 K, and the specimens were Fe, Fe–Cr, Fe–Mo [18], Fe–Ni, Fe–Si [19], and Fe–Ti, Fe–V, Fe–W [20].

### 2.3. Nuclear Fuels and Ceramics

T. Kikuchi and his colleagues studied the thermal conductivities of ceramic nuclear fuels such as uranium oxide, carbide, and related materials as well as lithium oxide, at the Japan Atomic Energy Research Institute. The value of the thermal conductivity of a sintered specimen was obtained from the measurement of thermal diffusivity by the laser pulse method. They investigated the thermal conductivity of UN [21] with density of 70–95% the theoretical density (TD) in the temperature range from 570 to 1670 K and of  $\text{Li}_2\text{O}$  with a density of 70–93% TD in the temperature range from 470 to 1170 K [22].

Fukushima et al. [23] studied the temperature dependence of thermal conductivity of  $\text{UO}_2\text{--YO}_{1.5}$  with various compositions of  $\text{YO}_{1.5}$  from 773 to 1973 K and obtained results that obeyed the following phonon conduction equation:

$$\lambda = (A + BT)^{-1} \quad (1)$$

where  $\lambda$ ,  $A$ ,  $B$ , and  $T$  are thermal conductivity, numerical constants, and absolute temperature, respectively. Fukushima and Abe [24] studied the temperature dependence of thermal conductivity of  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$  from 723 to 1873 K and obtained a result that was roughly fitted to the equation for both electron and phonon conduction:

$$\lambda = (LT/\rho) + (BT)^{-1} \quad (2)$$

where  $L$  and  $\rho$  are the Lorenz number and electric resistivity, respectively. Nasu et al. [25] investigated the thermal conductivity of a  $\text{U}(\text{C}_x\text{N}_{1-x})$  solid solution in the temperature range from room temperature to 1073 K, and they also measured the thermal conductivity of US from 633 to 1623 K and

discussed the electron contribution [26]. Further, they measured the thermal diffusivity of UP in the range from 573 to 1673 K [27].

Takahashi et al. [28] investigated the thermal conductivity of UC in the range from 473 to 1873 K and discussed the effect of oxygen concentration on the thermal conductivity. This group [29] measured the thermal conductivity of several nuclear related ceramics and their solid solutions of  $\text{UO}_2$ ,  $\text{ThO}_2$ ,  $\text{UO}_2\text{-ThO}_2$ ,  $\text{BeO}$ ,  $\text{CeO}_2$ , UC, UN,  $\text{ThO}_2\text{-BeO}$ , UC-UN, UP, US, and UP-US. Further, the thermal conductivities of  $\text{Y}_2\text{O}_3$  [30], AlN [31],  $\text{Si}_3\text{N}_4$  [32], and Sialon [33] were measured. These are interesting materials as high strength ceramics.

Hayashi et al. [34] improved a hot wire method for the measurements of thermal conductivity and measured an insulator consisting of ceramic fibers and powder. Kobayasi et al. [35] measured the temperature dependence of thermal diffusivities of several kinds of graphites from room temperature to 1273 K by a stepwise heating method. Eight specimens, i.e., an extruded natural graphite, a glassy carbon, vitrocarbon, two reactor grade graphites, porous carbon, high density graphite, and pyrolytic graphite were measured. In these specimens, two kinds of reactor grade graphites, the high density graphite and the pyrolytic graphite, were irradiated in a research reactor and measured to determine the effect of neutron irradiation on the thermal diffusivities.

Soga et al. [36] studied the thermal conductivity and mechanical properties of porous glass and discussed the effect of the porosity. Measurements of the thermal conductivity were carried out by a steady state method. Hayashi et al. [37] measured the thermal conductivity of refractory and insulating bricks up to 1473 K by a hot wire method. Kobayasi [38, 39] measured the temperature dependence of thermal diffusivity, specific heat, and thermal conductivity in the refractory materials, carbon brick, zirconia brick, blast furnace bottom brick, chamotte brick, roseki brick, high alumina brick, magnesia brick, and calcined magnesia brick in the range from room temperature to 1273 K. The thermal diffusivity and specific heat were simultaneously measured by a transient method based on a single rectangular pulse heating of the specimen.

#### 2.4. Building Materials and Insulators

Hokoi et al. [40] measured thermal conductivity of moist wood fiber board and discussed the effect of moisture content and the results both by the steady state and the periodic methods. Miyano et al. [41] presented the specific heat of building materials measured by a method of drop-calorimeter-mixtures and also showed the relation between specific heat and moisture content. The building materials were some pearlite mortar boards, glass and rock wools, several boards, plywoods, some gypsum boards, and several cellular plastics. Takegoshi et al. [42] studied the

effective thermal conductivities of hydrous porous insulators and the influences of vapor diffusion. As experimental samples, glass wool, rock wool, and plaster board were used, and the effective thermal conductivities were determined by a steady state comparative method.

Sakatsume et al. [43] investigated the thermophysical properties of cellular insulating materials. Thermal conductivity, heat capacity, and thermal diffusivity of polystyrene and polyurethane materials were determined. The technique for evaluation of the above three thermophysical properties was that developed by Krischer [44]. Tonokawa et al. [45] studied a new extruded polystyrene foam suitable for cryogenic use. They considered the relation between thermophysical properties and temperature in the range 296 to 103 K, and compared their material extruded polystyrene and polyurethane foams as well as phenolic foam in current use.

## 2.5. Soils and Rocks

Ohnaka et al. [46] measured the thermal conductivity of dry sand mold and vacuum process mold in the range from room temperature to 1273 K by a pouring method. Gray iron was poured into a mold 100 mm in diameter and 70 mm high. The metal-mold interface temperature and mold temperature at 4, 8, 12, 16, 20, and 24 mm depth from the interface were measured and values found by use of a computer. The effect of grain size was discussed. Hasatani et al. [47] studied the effective thermal conductivity of wet soils by a hot wire method. The measured data for several soils were correlated with the moisture content and the temperature. After these confirmations, actual measurements were carried out for a natural wet soil in a vegetable field, and not only the effect of moisture content but also the effects of other parameters, particularly of organic compounds content, were discussed.

Sawada [48] studied the thermophysical properties of frozen soil for application to the construction of roads, buildings, and airfields in cold regions. Seki et al. [49] also studied the thermal conductivity of frozen and unfrozen soils as basic research for the study of the phenomenon of frost action. Thermal diffusivity was also calculated from the thermal conductivity and associated equivalent heat capacity. The measurements were made by a nonstationary absolute method using a thermal conductivity probe. The temperature range of these measurements was from about 298 to 253 K. The soil samples were prepared from four kinds of soils, such as clay, silt, sand, and gravel. The moisture content of each sample was arranged by adding an appropriate amount of water to oven-dried soil. Watanabe [50] measured the thermal conductivity of frozen and unfrozen soils. This experiment was mainly conducted by a thermal probe method. A steady state method was also performed to measure the thermal conductivity of clay. Inaba [51] dealt with the measurements of thermophysical properties

and unfrozen water content for several fine and coarse grain size soils over a range above or below freezing temperature under a small freezing rate. The effects of temperature, soil type, and initial moisture content on the thermophysical properties and unfrozen water content were examined.

Kiyohashi et al. [52] reported on the technique of a transient hot wire method for the measurements of thermal conductivity of several igneous rocks as a function of temperature. Inada granite, KG-1 granite from ROK, Andes black granite from Brazil, Emochi andesite, and Kofu andesite were studied. The measurements were carried out in the temperature range from 293 to 773 K. Kiyohashi et al. [53] also measured effective thermal conductivities of dry rock sample taken from cores of U-1 well bore (about 1100 m in depth) at the Onikobe geothermal area of Japan, a hot water geothermal area, in the temperature range from room temperature to 773 K and estimated effective thermal conductivities in situ of the wet reservoir rocks using their data and Maxwell's equation. The measurements were performed by a needle probe type transient hot wire method at atmospheric pressure in this case.

## 2.6. Energy Storage Materials

Kamimoto [54] measured enthalpy change for three promising thermal energy storage materials, i.e.,  $\text{NaNO}_2$ ,  $\text{LiNO}_3$ , and  $\text{NaNO}_2\text{-KNO}_3$  (50-50 mol%). The measurements were done by a twin high temperature calorimeter. Ozawa [55] evaluated the heat of fusion or transition, melting and crystallization behavior, specific heat, and thermal stability by differential scanning calorimetry. Urea, polyethylene, pentaerythritol, and alkaline hydroxides were examined. Kobayasi et al. [56] measured thermal diffusivity, specific heat, thermal conductivity, and latent heat of fusion of several salts in both phases of solid and liquid by four kinds of measuring methods, i.e., a single rectangular pulse heating method, an arbitrary heating method for solid phase, a stepwise heating method for liquid phase, and an improved differential thermal analysis for specific heats of solid and liquid phases and latent heat of fusion. The results were presented for  $\text{NaNO}_3$ ,  $\text{NaNO}_2$ ,  $\text{KNO}_3$ ,  $\text{LiNO}_3$ , and HTS (Heat Transfer Salt). This group also measured the same thermophysical properties of  $\text{NaOH}$ ,  $\text{KSCN}$ ,  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{C}_{12}\text{H}_{10}$  (diphenyl), and polyethylene glycols Nos. 2000, 4000, and 6000 from the solid phase at room temperature to the liquid phase beyond melting temperature [57].

## 2.7. Compound and Dispersed Materials

On the problem of analytical prediction of effective thermal conductivity, Oka et al. [58] proposed using a cylindrical model in order to estimate the effective thermal conductivity of plastic foam material. On the other

hand, Nagatani [59] presented his estimation of the effective thermal conductivity based on statistical theory. Most of the papers cited have dealt with the effective thermal conductivity of a porous media with a single pore diameter, but Fujitsu et al. [60] proposed a way to estimate the effective thermal conductivity of insulating-refractory materials whose macropores were uniformly dispersed in a continuous solid of distributed micropores. Sugawara et al. [61–64] proposed an estimation based on their experiments using rocks, glass beads, and rubber plates with holes. Imura et al. [65] analyzed a model with three phase mixtures by considering the experimental results from particle-bound grinding stone. Yamakawa et al. [66] also clarified experimentally the behavior of effective thermal conductivity.

On the problem of thermal conductivity including the effect of radiation, Saegusa et al. [67] proposed a modified estimation of the equation derived by Eucken [68], with consideration of the radiation effect in the pore. Saegusa et al. [69] also investigated the effective thermal conductivity of porous media of cubic particles having cubic arrangement. Kumada [70] discussed the agreement of the previously proposed estimations with his experiments. Yamada et al. [71] proposed a predicting formula based on the measured data for dispersed materials including parallelepiped particles and numerical results. Further, he and his colleagues [72] made a numerical analysis on the effective thermal diffusivity of dispersed materials including square particles as the discontinuous phase. The square particles were randomly dispersed with an uniformly random number. They also analyzed the effective thermal diffusivity of a semiinfinite media in which the rectangular particles were randomly dispersed [73].

### 3. SUMMARY REMARKS

In this review, an attempt has been made to keep engineering use in mind. We surmise that some research results in other fields might have been omitted, but we hope that our outline of the major part of Japanese research, principally on thermal conductivity and thermal diffusivity of solids, is comprehensive.

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