Thermal Conductivity Measurement of Molten Silicon by a Hot-Disk Method in Short-Duration Microgravity Environments

Jpn. J. Appl. Phys. Vol. 39 (2000) pp. 1405–1408
Part 1, No. 3A, March 2000

Thermal Conductivity Measurement of Molten Silicon by a Hot-Disk Method in Short-Duration Microgravity Environments

Hideaki NAGAI[1], Yoshinori NAKATA[1], Takashi TSURUE[1], Hideki MINAGAWA[1], Keiji KAMADA[2], Silas E. GUSTAFSSON[3] and Takeshi OKUTANI[1]

[1]*Hokkaido National Industrial Research Institute (HNIRI), 2-17-2-1, Tsukisamu-higashi, Toyohira-ku, Sapporo 062-8517, Japan*
[2]*Japan Space Utilization Promotion Center (JSUP), 3-30-16, Nishiwaseda, Shinjyuku-ku, Tokyo 169-8624, Japan*
[3]*Department of Physics, Chalmers Institute of Technology, SE-412 96 Gothenburg, Sweden*

(Received October 27, 1999; accepted for publication January 6, 2000)

The thermal conductivity of molten silicon was measured by a hot-disk method in short-duration microgravity environments. The hot-disk sensor was made of molybdenum foil cut in a conducting pattern and covered with an aluminum nitride plate. Aluminum nitride has good resistivity against corrosion from silicon melt and the molybdenum foil was protected from the molten silicon. The thermal conductivity of molten silicon measured on the ground was estimated to be 45.6 $W \cdot m^{-1} \cdot K^{-1}$ at the melting point (1687 K). The thermal conductivity of molten silicon measured in microgravity was about 5% lower than that measured on the ground.

KEYWORDS: thermal conductivity, molten silicon, aluminum nitride, hot-disk method, microgravity

1. Introduction

Thermal conductivity is an important property in many heat-transfer simulations. This value can be measured by the thermal response when heat or energy is applied to a specimen. However, it is difficult to precisely measure the thermal conductivities of liquid materials (water, organic solvents, molten metals, etc.) because heat is transferred by convection as well as conduction. It is possible to precisely measure the thermal conductivities of liquid materials in microgravity because thermal convection is suppressed.

The thermal conductivities of liquid materials can be measured by (1) a steady heat flow method,[1)] (2) a stepwise heating method,[2)] (3) a transient hot-wire method[3)] or (4) a laser flash method.[4)] The hot-wire method is more suitable in microgravity experiments because of its short measurement time. Nakamura *et al*. reported on the thermal conductivity of molten indium antimonide measured by the hot-wire method in microgravity [using sounding rockets (6 min microgravity) and drop shafts (10 s microgravity)].[5,6)] However, there are no reports of the thermal conductivity of a high-temperature melt, such as molten silicon, in microgravity.

Gustafsson recently developed the hot-disk method for measuring thermal conductivity.[7)] This method uses a transient plane source (TPS) element as both the heat source and temperature sensor, in the same way that a thin wire is used in the hot-wire method. The TPS element is made of thin metal foil, and its conducting pattern is a double spiral, which with some approximation resembles a hot disk. A thin bare wire was utilized for the hot-wire method, and only the thermal conductivity of the insulating materials could be measured. In contrast, the thermal conductivity of both the insulating materials and the electrical conducting materials could be measured by the hot-disk method because both sides of the TPS element were covered with a thin insulating layer. However, it was impossible to measure the thermal conductivity of a high-temperature melt like molten silicon because the insulator of the commercial hot-disk sensor was made of kapton or mica. Aluminum nitride (AlN) is a good insulator, and its reactivity against molten metal is poor. It is therefore possible to make a hot-disk sensor for a high-temperature melt by using AlN as the insulator. In this study, we developed a hot-disk sensor for molten silicon and measured the thermal conductivity of molten silicon by the hot-disk method in microgravity using a drop tower.

2. Experimental

2.1 Principle of the hot-disk method

The normal procedure in hot-disk method experiments is to pass a constant current through a hot-disk sensor and simultaneously record its voltage changes. It is convenient to express the time-dependent resistance $[R(t)]$ by the following equation when analyzing the behavior of a hot-disk sensor during a transient recording:

$$R(t) = R_0[1 + \alpha \Delta T(\tau)] \tag{1}$$

where R_0 is the resistance of the hot-disk sensor before the transient recording, α is the temperature coefficient of resistance (TCR), and $\Delta T(\tau)$ is the time-dependent temperature increase of the hot-disk sensor.[7)] The temperature increase is expressed in this equation as a function of only one variable, τ, which is defined as:

$$\tau = (t/\theta)^{1/2},\ \theta = d^2/\kappa \tag{2}$$

where t is the time elapsed from the start of the transient heating, θ is the characteristic time, d is the radius of the hot disk, and κ is the thermal diffusivity of the sample material.

The evaluation of $\Delta T(\tau)$ in the heater depends on the power output in the TPS element, the design parameters of the sensor, and the thermal transport properties of the surrounding sample. $\Delta T(\tau)$ is given by the following equation for a disk-shaped sensor, from which the thermal conductivity and diffusivity can be obtained:

$$\Delta T(\tau) = P_0(\pi^{3/2} d\lambda)^{-1} D(\tau) \tag{3}$$

where

$$D(\tau) = [m(m+1)]^{-2} \int_0^{\tau} \mathrm{d}\sigma\ \sigma^{-2} \left[\sum_{l=1}^{m} l \sum_{k=1}^{m} k \exp\left(\frac{-(l^2+k^2)}{4m^2\sigma^2} \right) I_0\left(\frac{lk}{2m^2\sigma^2} \right) \right] \tag{4}$$

P_0 is the total output power, λ is the thermal conductivity of the sample material. $D(\tau)$ is the theoretical expression of the time-dependent temperature increase, which describes the conducting pattern of the disk-shaped sensor, assuming that the disk consists of a number m of concentric ring sources.[7)]

We substitute eq. (3) for eq. (1), and then obtain eq. (5).

$$R(t) = R_0[1 + \alpha P_0(\pi^{3/2}d\lambda)^{-1}D(\tau)] \quad (5)$$

When the characteristic time (θ) has a reasonable value, a plot of the measured resistivities $R(t)$ vs $D(\tau)$ will, according to eq. (5), yield a straight line. In actuality, we calculate $D(\tau)$ on each data recording time for any value of θ and then calculate the correlation coefficients of the straight line fit of $R(t)$ and $D(\tau)$ for any value of θ. The θ value for the best straight line fit [eq. (5)] is used to obtain the thermal diffusivity of the sample material [eq. (2)]. The thermal conductivity is calculated from the slope of the best straight line fit [eq. (5)]. Thermal conductivity and thermal diffusivity of the sample material are related as follows:

$$\lambda = \rho C_p \kappa \quad (6)$$

where ρ is the density of the sample and C_p is the specific heat of the sample. The specific heat per unit volume (ρC_p) can be calculated by using the values of the thermal diffusivity and thermal conductivity of the sample material. When the measurement time from the start of the transient heating differs significantly from the characteristic time (ideally between 0.5 to 1.0 times as long as the characteristic time), it becomes necessary to know the specific heat of the sample to obtain reliable results.

2.2 Development of a hot-disk sensor for molten silicon

Figure 1 shows the hot-disk sensor for molten silicon. Molybdenum foil (Nilaco; 10 μm thick) was cut in a conducting pattern using a plasma discharge cutting technique, as shown in Fig. 1(a). The radius of its double spiral part was 2.75 mm. After this foil was annealed at 1273 K in hydrogen gas, its double spiral part was covered with thin AlN plates (SH-15, Tokuyama; 0.1 mm thick, polished by the author) and other parts were covered with thick AlN plates (0.63 mm thick). The molybdenum foil and aluminum nitride plate were glued together using alumina cement (Sumiceram S-208B, Asahi Chemical Industry) and heated to 1273 K in hydrogen gas.

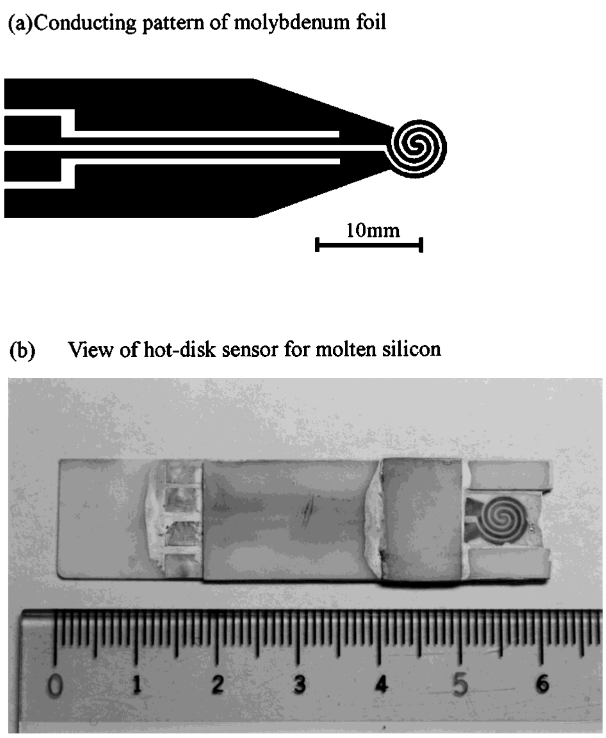

Fig. 1. Structure of hot-disk sensor for molten silicon.

2.3 Equipment setup and experimental procedure

The thermal conductivity of molten silicon was measured by a hot-disk thermal constant analyser (Hot Disk Inc., Sweden). A schematic diagram of the thermal conductivity measurement setup is shown in Fig. 2. The hot-disk method utilizes a hot-disk sensor as a heat source and a temperature sensor to measure the thermal conductivity. A constant electric power was supplied to the hot-disk sensor by the source meter to measure the sensor resistance. We used a computer to control the source meter, record the data, and analyze the thermal response during measurements. The hot-disk sensor was set up vertically in a sample container made of alumina (Nikkato; inner diameter, 17 mm; height, 60 mm). The sample was a non-doped silicon single crystal (Shin-Etsu Handotai; 99.999999999% purity). The sample was cut and polished into the proper shape and put into a sample container after the oxidized surface layer of the sample had been chemically etched by a mixture of nitric acid and hydrofluoric acid (HNO_3 : HF = 90 : 10 vol%) for 5 min. The hot-disk sensor and sample container were set into a measurement chamber made of stainless steel; argon gas (purity 99.9995%, oxygen content less than 0.2 ppm) was then introduced into the chamber up to a pressure of 1.0×10^5 Pa after evacuation. The sample was heated by a tungsten heater. A small temperature gradient was applied in the heated zone to avoid thermal convection before dropping, a procedure similar to that in a report by Nakamura *et al.*[5)] The temperature of the top part was about 15 K higher than that of the bottom part. The thermal conductivity was measured when the sample reached the

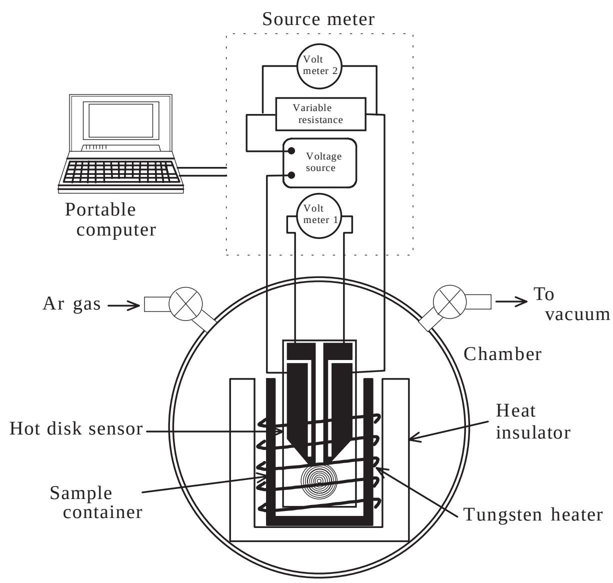

Fig. 2. Schematic diagram of experimental apparatus for thermal conductivity.

measurement temperature.

The microgravity experiments were performed using the 10 m drop tower at HNIRI.[8] The microgravity quality and time were 10^{-3} g and 1.2 s.

3. Results and Discussion

The thermal conductivities of standard materials were measured at room temperature to confirm the reliability of the thermal conductivity measured by our developed hot-disk sensor. Table I shows the experimental results and reference data for the thermal conductivities of some materials.[9] Data recorded for the first period, for example 0.2 s on brass and 0.8 s on silica glass, were not used to calculate the thermal conductivity because these data were strongly affected by the insulating layer of the hot-disk sensor, and the linearity of eq. (5) was worse. The thermal conductivities of silica glass, mercury, and stainless steel were greater than the reference ones; those of lead and brass were similar to the reference ones. In the hot-disk method experiments, heat generated from the double spiral conducting pattern was conducted through the insulator. When the thermal conductivity of the insulator is low, heat is not diffused in the radial direction of the insulator, and the same size heat pattern forms on the surface of the insulator. When the thermal conductivity of the insulator is high, heat is easily diffused in the radial direction of the insulator, and the radius of the heat pattern formed on the surface of insulator is bigger than that of the double spiral conducting pattern. This indicates that the apparent radius of the hot-disk sensor increases, shifting the calculated result. This effect is enhanced as the differences in thermal conductivities between the insulator and the sample increase. The result in Table I demonstrates that a sample with a thermal conductivity exceeding 30 $W \cdot m^{-1} \cdot K^{-1}$ can be measured correctly by our hot-disk sensor. The thermal conductivity of molten silicon was reported to be 56 $W \cdot m^{-1} \cdot K^{-1}$.[10] Therefore, our hot-disk sensor can be used to measure the thermal conductivity of molten silicon.

Figure 3 shows a cross section of the hot-disk sensor for molten silicon after contact with the molten silicon. A part of the AlN was corroded by the molten silicon, but the molybdenum foil was completely protected from the molten silicon. We could not find any bubbles at the interface between the molten silicon and the hot-disk sensor. Wetting between the molten silicon and the hot-disk sensor was evidently good, because the surface of the AlN was corroded by the molten silicon. Therefore, the thermal contact between the molten silicon and the hot-disk sensor was satisfactory.

Figure 4 shows a typical apparent thermal conductivity of molten silicon. Each thermal conductivity was calculated by recording data for 0.26 s on the assumptions that the specific heat of the molten silicon was 1020 $J \cdot kg^{-1} \cdot K^{-1}$[11] and that its density was 2.53×10^3 $kg \cdot m^{-3}$;[12] the measurement time from the start of the transient heating to the time corresponding to the pure heat response of the molten silicon (between 0.8 s and 1.2 s in Fig. 4) was much bigger than the characteristic time [about 0.4 s when the thermal conductivity of molten silicon is 45 $W \cdot m^{-1} \cdot K^{-1}$ (based on our experimental result cited bellow)]. The apparent thermal conductivities were classified into three types by the behavior of the heat conduction, as shown in Fig. 4. The thermal conductivity of region (a) (high value but quickly decreased) was strongly affected by the high thermal conductivity of AlN. We did not use the data of this region to calculate the thermal conductivity because the linearity of eq. (5) was worse. The thermal conductivity of region (b) (constant value) was rarely affected by the AlN or the alumina container. This region exhibited pure thermal conductivity of molten silicon. The thermal conductivity of region (c) (value gradually decreased) was affected by the alumina container; the amount of molten silicon was limited by the size of the experimental apparatus.

Figure 5 shows the temperature dependence of the thermal conductivity of molten silicon in region (b) on the ground and in microgravity. The thermal conductivity of molten silicon at the melting point, extrapolated from our result measured on the ground, was 45.6 $W \cdot m^{-1} \cdot K^{-1}$. This value was 20% lower than the result of Yamamoto *et al*.[10] The Wiedemann-Franz law has been widely accepted for the thermal conductivity of

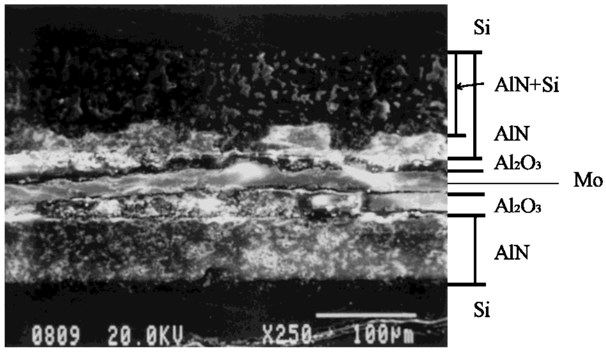

Fig. 3. Photograph of hot-disk sensor after contact with molten silicon. Condition: 1713 K, 10 min, Argon atmosphere

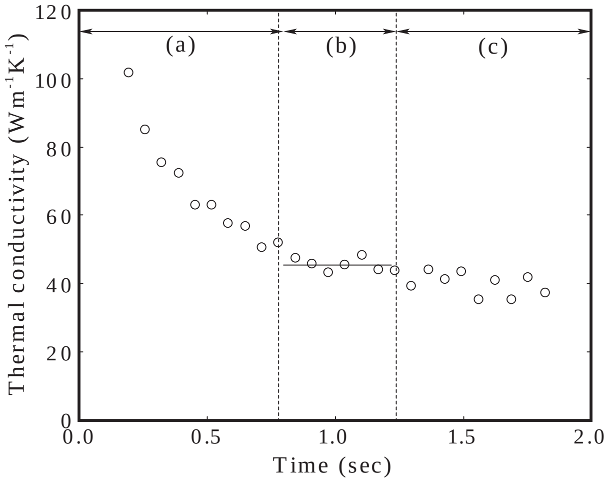

Fig. 4. Time-dependence of apparent thermal conductivity of molten silicon. Conditions: Temperature; 1703 K Each point was calculated by the data collected for 0.26 s (as specific heat ; 1020 $J \cdot kg^{-1} \cdot K^{-1}$ and density; 2.53×10^3 $kg \cdot m^{-3}$).

Table I. Thermal conductivity measurement of standard materials by a hot-disk sensor for high temperature melt at room temperature.

Standard material	Thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$)	
	Experimental	Ref. 9
Silica glass	3.4	1.38
Stainless steel	19.8	15
Lead	32.5	34.8
Brass	112.6	119
Mercury	12.1	8.34

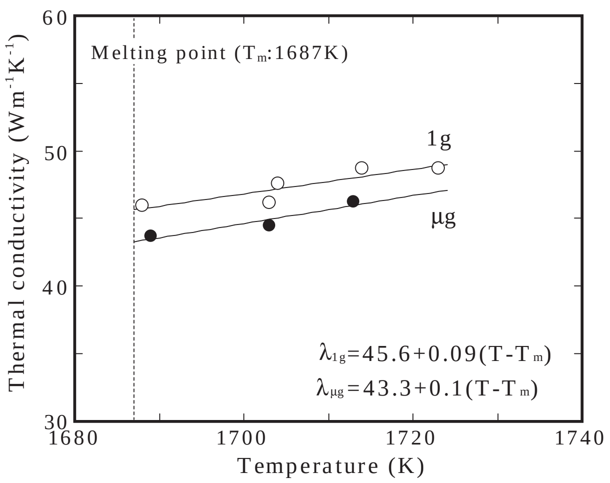

Fig. 5. Thermal conductivity of molten silicon on the ground and in microgravity. Thermal conducitvity of molten silicon was calculated as 1020 J·kg^{-1}·K^{-1} and 2.53 × 10^3 kg·m^{-3}.

liquid metals and is expressed as

$$\lambda = L \cdot T/r \tag{7}$$

where λ is the thermal conductivity, L is the Lorenz number, which is 2.445×10^{-8} (W·Ω·K^{-2}) for a degenerate free-electron gas system, T is the temperature, and r is the resistivity. The thermal conductivities of molten silicon estimated by the Wiedemann-Franz law were 47–57 W·m^{-1}·K^{-1} using the data of electrical conductivities.[12–15] Our result was close to the values obtained from the Wiedemann-Franz law. Its thermal conductivity in microgravity experiments was about 5% lower than that on the ground. It was reported that the accuracy of thermal conductivity measured by the hot-disk method using a commercial hot-disk sensor with a kapton insulator was 2%,[16] however, 2% accuracy could not be achieved with the thermal conductivity of molten silicon in our study because the properties of our developed hot-disk sensor differ from those of commercial sensors. Therefore, we considered the accuracy of the thermal conductivity measurements by the hot-disk method in our study to be as follows, since there are no standard materials for measuring thermal conductivity at high temperatures. In hot-disk method, the thermal conductivity of the sample is analyzed from the temperature increase of the hot-disk sensor that touches the sample. We could estimate the accuracy of the thermal conductivity of molten silicon in our study if the accuracy of the thermal conductivity was directly related to that of the temperature increase. Because the temperature increase of the hot-disk sensor is calculated from its resistance increase and the TCR value of the TPS element, its accuracy is determined by the accuracy of the resistance and TCR value. The typical resistance and TCR value of a commercial hot-disk sensor and our developed hot-disk sensor are shown in Table II. A comparison between the commercial hot-disk sensor and our developed one at room temperature reveals that the resistance and TCR value of our developed hot-disk sensor are 6.2 times and 1.3 times less accurate than those of the commercial sensor. Consequently, the temperature increase of our developed hot-disk sensor is 8.0 times less accurate than that of the commercial sensor, and the accuracy of thermal conductivity in our study could be estimated to be 2% × 8.0 = 16%. Our results of the thermal conductivities of lead and brass shown in Table I had

Table II. Typical resistance and TCR value of the commercial hot-disk sensor and our developed hot-disk sensor.

	Temperature (K)	Resistance (Ω)	TCR (K^{-1})
Commercial hot-disk sensor	293	4	4.7×10^{-3}
Our developed hot-disk sensor	293	0.65	3.6×10^{-3}
	1713	10	1.2×10^{-3}

about a 6% accuracy compared with reference data, our estimate thus seemed consistent with our experimental results. Similarly, the accuracy of thermal conductivity in our study at 1713 K could be estimated to be 2% × 1.6 = 3.2%. The results of the thermal conductivity of molten silicon and the estimated accuracy revealed that the thermal conductivity of molten silicon measured on the ground included the effect of thermal convection introduced by the heat of the hot-disk sensor. We reported that the thermal conductivity of mercury in microgravity was 3% lower than that on the ground when the thermal conductivities were measured using a commercial hot-disk sensor with a kapton insulator.[17] Since molten silicon is a low-viscosity fluid similar to mercury, we considered that the thermal conductivity of molten silicon measured on the ground included effects of thermal convection on the measurement similar to those of mercury.

4. Conclusions

In this study, a hot-disk sensor for molten silicon was developed and the thermal conductivity of molten silicon was measured using the hot-disk method in short-duration microgravity. The thermal conductivity of molten silicon at the melting point was 45.6 W·m^{-1}·K^{-1} and the thermal conductivity of molten silicon in microgravity was about 5% lower than that on the ground.

1) S. Fischer and E. Obermeier: High Temp.-High Press. **17** (1985) 699.
2) Y. Kato, K. Furukawa, N.Arai and K. Kobayashi: High Temp.-High Press. **15** (1983) 191.
3) Y. Nagasaka and A. Nagashima: Rev. Sci. Instrum. **52** (1981) 229.
4) H. Ohta, G. Ogura and Y. Waseda: Rev. Sci. Instrum. **61** (1990) 2645.
5) S. Nakamura, T. Hibiya, F. Yamamoto and T. Yokota: Int. J. Thermophys. **12** (1991) 783.
6) S. Nakamura and T. Hibiya: Microgravity Sci. Technol. **6** (1993) 119.
7) S. E. Gustafsson: Rev. Sci. Instrum. **62** (1991) 797.
8) T. Okutani, H. Minagawa, M. Mori and Y. Sakai: J. Jpn. Soc. Microgravity Appl. **15** (1998) 79 [in Japanese].
9) *Kagaku Binran Kiso-hen* (Chemical Handbook, Basic section), ed. Chemical Society of Japan (Maruzen, Tokyo, 1993) No. 2, p. 69 [in Japanese].
10) K. Yamamoto, T. Abe and S. Takasu: Jpn. J. Appl. Phys. **30** (1991) 2423.
11) K. Yamaguchi: Report of research program of "Technology for production of high quality crystal" (1999) p. 141 [in Japanese].
12) V. M. Glazov, S. N. Chizhevskaya and N. N. Glagoleva: *Liquid Semiconductors*, ed. A. Tybulewicz (Plenum Press, New York, 1969) Chap. 3, p. 60.
13) M. S. Ablova, O. D. Elpat'evskaya and A. R. Regel': Zh. Tekh. Fiz. **26** (1956) 1366; through ref. 12.
14) B. A. Baum, P. V. Gel'd and S. I. Suchil'nikov: Izv. Akademii Nauk SSSR-Met. i Gornoe Delo (1964) No. 2, 149; through ref. 12.
15) H. Sasaki, A. Ikari, K. Terashima and S. Kimura: Jpn. J. Appl. Phys. **34** (1995) 3426.
16) T. Log and S. E.Gustafsson: Fire & Mater. **19** (1995) 43.
17) H. Nagai, F. Rossignol, Y. Nakata, T. Tsurue, M. Suzuki and T.Okutani: to be published in Mater. Sci. & Eng. A.