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## The effects of PSS and Au addition on thermoelectric properties in $\beta$ -SiC/Si composites

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### Abstract

Silicon semiconductor devices such as Power MOSFET, IGBT and CPU are the most important devices to give us modern life. When we use these devices in the electric circuit, the heat removal and the cooling down are one of the most important issues because the silicon device must be the temperature of 423K or lower to keep its function. Recently Yamaguchi *et al.* propose the self-cooling device, which does not need to use the additional power circuits because the Peltier cooling is done by its selfcurrent. This technology carries out the Peltier cooling with the use of flowing current in silicon power device itself by using thermoelectric material. In particular, silicon carbide (SiC) is one of the candidate material since SiC has the higher electrical conductivity, thermal conductivity and Seebeck coefficient different from conventional thermoelectric material. The purpose of this study is to investigate the effects of PSS and Au addition on the thermoelectric properties in *p*-type semiconductor SiC/Si/Au composites in order to apply the self-cooling semiconductor device.

### Introduction

Electronic silicon devices such as power metal oxide semiconductor field effect transistor (MOSFET), insulated gate bipolar transistor (IGBT) and microprocessor (CPU) are used everywhere to keep our modern life in the present time, and at the same time, the cooling down is one of the most important issues because the silicon device must be the temperature of 150°C or lower to keep its function. One of the way to remove the heat from the silicon devices is to use the usual Peltier element, and this is connected with the large fin and fan usually. The usual Peltier element and the fan need the different dc power supplies for cooling, individually. Thus, if we use the usual Peltier element to cool down the silicon devices, the electric power consumption of the system is not negligible. This is one of the reasons why the usual Peltier cooler is not applied for the laptop computers.

Recently Yamaguchi *et al.*[1][2] propose the new method of heat removal from the devices, i.e., the self-cooling device, which does not need to use the additional power supply because the Peltier cooling is done by its self-current. One of the candidate material of the self-cooling device is silicon carbide (SiC) [1] because this applicable material are demanded higher electrical conductivity, thermal conductivity and Seebeck coefficient different from a conventional thermoelectric material. In fact, SiC is a wide band

semiconductor that is considered to be very promising for many applications so that potentially useful material. This material was expected to be a stable thermoelectric material even at high temperatures because of its high chemical stability and mechanical hardness (strength) even at around 1273K.[3] However, a high thermal conductivity of SiC gives a low figure of merit  $Z=S^2\sigma/\kappa$ , where  $S$ ,  $\sigma$  and  $\kappa$  are the Seebeck coefficient, electrical conductivity and thermal conductivity, respectively. A lot of researchers had developed SiC as conventional thermoelectric materials to control transport parameters by the porosity control in various types of sintering methods.[3-5] To the best of the authors' knowledge, no research has yet been carried out to realize an advanced stage for thermoelectric material. SiC has a multitude of polytypes such as 3C, 4H and 6H type[6-8], where the leading number indicates the number of Si-C bilayers in the unit cell with H and C representing Hexagonal and Cubic crystal structures. In general, a hexagonal and a cubic SiC are known as  $\alpha$ -type and  $\beta$ -type SiC, respectively.

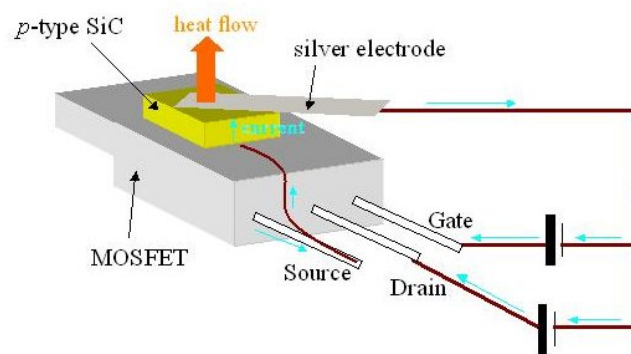


Fig. 1. Self-cooling device which consists of power MOSFET and *p*-type SiC.

Figure 1 shows the self-cooling device which consists of vertical type of power MOSFET with *p*-type SiC. The *p*-type SiC perform the Peltier cooling, and the bottom side of it is connected the Source of MOSFET. Since the electric current is transferred from the Drain to the Source, both electric current and the heat flux flows from the bottom to the upper direction in the *p*-type SiC. Thus, the self-cooling device cools down the power MOSFET in which the temperature of the power device is higher than the room temperature. This is different from the usual Peltier cooling process because the conventional Peltier element use the Peltier heat as the heat pump transferring heat from cool side to hot side, thereby, the

Peltier materials are desired to have low thermal conductivity. As shown in Fig.2, the direction of the Peltier heat flow is the opposite for the conventional Peltier element because of the cooling high temperature objects.

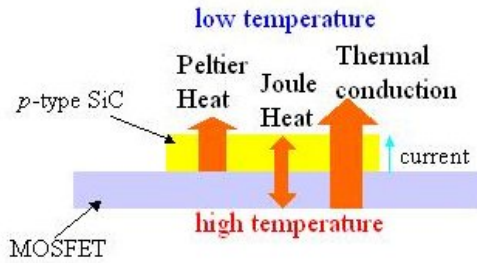


Fig. 2. Heat flux flows in the self-cooling device.

Considering a power device including a thermoelectric element in the circuit, the Peltier heat can increase the cooling efficiency to remove the Joule heat from the device. Since the Peltier heat is proportional to the current, the Peltier cooling by its self-current can be more effective in the high current power devices for cooling. In this system, Joule heat is generated isotropically and thermal conduction heat is transported by Peltier effect from power MOSFET to lower side. It is clear that SiC with high electrical conductivity, thermal conductivity and Seebeck coefficient is one of the suitable materials for the self-cooling device.

In this paper, we investigated the polysilastyrene (PSS) and Au addition effects on thermoelectric properties in  $\beta$ -SiC/Si composites, where PSS is one of the sintering additives in this system. The purpose of this study is to investigate the thermoelectric properties and crystal structure for SiC/Si/Au composite in order to apply the self-cooling semiconductor device.

## Experimental

Polycrystalline samples were prepared under the following condition by a conventional solid-state reaction.  $\beta$ -SiC (average particle size 0.15 $\mu$ m, Mitsui Toatsu, 99.7%, MSC-20), Si (Kojundo Chemical Laboratory, 99.99%) and Au (150 $\mu$ m pass, Kojundo Chemical Laboratory, 99.9%) powder were used as starting material, and PSS (Nippon Soda, PSS-100) was mixed as sintering aids. Excess PSS thermally decomposes and evaporates during the sintering process and pores remain in the sample. Slurries were made from mixed powder of (i) 60 wt.% SiC and 40 wt.% Si (SiC/Si=60/40), (ii) 60 wt.% SiC and 40 wt.% Si and added PSS 10 wt.% (SiC/Si=60/40+PSS) and (iii) 55 wt.% SiC, 40 wt.% Si, 5 wt.% Au and added PSS 10 wt.% (SiC/Si/Au=55/40/5+PSS). Then the slurries were mixed in polyethylene jars with nylon coated iron balls as the grinding media and with xylene solution as the mixing agent. After mixing for 24 hours and passing through 75 mesh sieve, these slurries were dried. After drying the slurry, their powder granulated using 500 $\mu$ m mesh sieve.

The mixing powder were formed pellet type of 20mm  $\phi$   $\times$  5mm by uniaxial pressing at  $1 \times 10^6$  kg/m<sup>2</sup>. Then the pellets were conducted the Cold Isostatic Pressing (CIP, Nikkiso, CL3-22-60) at  $3.5 \times 10^7$  kg/m<sup>2</sup> after sealed into an evacuated vinyl packing. Each pellet sample during the sintering process was muffled the iso-composition powder, and placed a carbon crucible which was anointed boron nitride at internal surface. Sintering was carried out in vacuum and atmosphere furnace heated by carbon. The sintering procedure employed the radio frequency induction furnace (Fujidenpa, FVPHP-10). First, the furnace was heated up to 1000 $^{\circ}$ C at a rate of 20 $^{\circ}$ C/min in vacuum, and Ar gas was introduced at normal pressure. The furnace temperature was further raised to 2100 $^{\circ}$ C at a rate of 10 $^{\circ}$ C/min, and the sample was kept at this temperature for 2 hours and cooled naturally down to room temperature. At last, furnace was cooled down to 1600 $^{\circ}$ C at the rate of 100 $^{\circ}$ C/min and then naturally cooled down to room temperature. Sintered bodies were cut to rectangular shaped specimens of 2 $\times$ 10 $\times$ 10 mm<sup>3</sup> in dimensions for measurement of the thermoelectric properties and using the self-cooling device.

The measurements of electrical resistivity and Seebeck coefficient were carried out in the temperature range from 80 to 960 K. The electrical resistivity  $\rho$  was measured by a van der Pauw technique with a current of 10mA in He atmosphere (Toyo Corp, 8350LH/CD) at low temperature (80-400K) and by a home made device in Ar atmosphere at high temperature (300-960K). The Seebeck coefficient  $S$  measurement was carried out on a sample placed between two blocks of oxygen-free high-conductivity (OFHC) silver. The Hall coefficient  $R_H$  measurement was performed on flat square pieces of materials with a current of 100mA in a magnetic field of 8500Oe by a van der Pauw technique at room temperature. The Hall carrier concentration  $n$  was determined from  $R_H$  using  $n=1/eR_H$ , where  $e$  is the electron charge. Furthermore, the Hall mobility  $\mu$  was determined from  $\rho$  and  $R_H$  using  $\mu=R_H/\rho$ . The thermal diffusivity  $A$ , sample density  $D$  and specific heat  $C$  were measured at room temperature, where the thermal conductivity  $\kappa$  was calculated from  $\kappa=ACD$ . The thermal diffusivity  $A$  was measured by a laser flash method (TC-3000, ULVAC). Sample density  $D$  was measured by Archimedes's method by using purified water as solvent.

The microstructures of the samples were examined by a scanning electron microscope (SEM: VE-8800, KEYENCE) and simultaneously elementary distributions of Si, C and Au in microstructure after sintering process were clarified by using an electron probe x-ray micro analyzer (EPMA: JXA-8900R, JEOL). The chemical composition of aurum and carbon in Sintered SiC samples was examined by using Inductively Coupled Plasma (ICP) analysis, the direct combustion-gasometric method and the gravimetric technique, respectively. SiC content was calculated from detection amount of carbon, and remaining amount of silicon was fixed up as additional Si. In consequence, additional Si and Au amount was detected 10-20wt.% and 2.5wt.% as against 40wt.% and 5wt.% respectively before it sintered, i.e., (i) SiC/Si=90/10, (ii) SiC/Si=80/20+PSS and (iii) SiC/Si/Au=80/17/3+PSS, respectively. It was confirmed that content of respective

elements was reduced above 50%, nevertheless sintering temperature was below the boiling point of Si and Au.

## Results and Discussion

The thermoelectric properties of the polycrystalline specimens of (i) SiC/Si=90/10, (ii) SiC/Si=80/20+PSS and (iii) SiC/Si/Au=80/17/3+PSS were measured in the temperature range from 80K to 960K. Figure 3 shows the discontinuous change at around 450K and metallic behavior in the temperature range from 80K to 450K. In particular, the sample of (i) shows a broad maximum and a metal-semiconductor transition at around 600K. Figure 4 shows the temperature dependence of the Seebeck coefficient  $S(T)$  of the samples in the temperature range from 80K to 960K. It can be observed clearly that the Seebeck coefficient shows positive which means main carrier is hole. Besides, all the samples show a broad maximum at around 600K. In particular, the sample of (i) shows maximum value at low temperature, on the other hand, the sample of (iii) shows maximum value at high temperature.

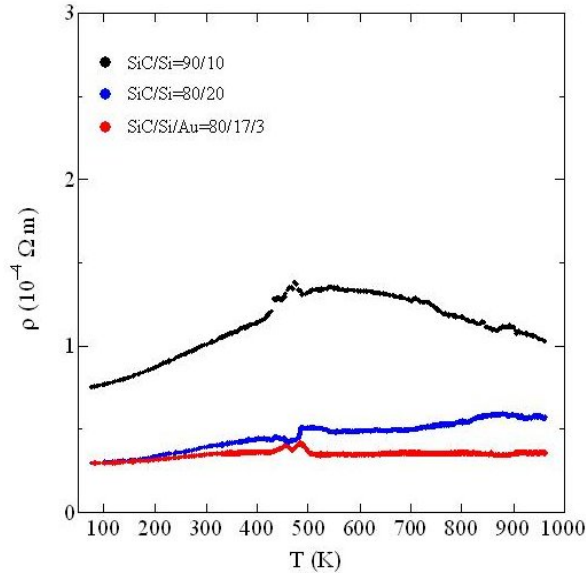


Fig. 3. Temperature dependence of electrical resistivity  $\rho$

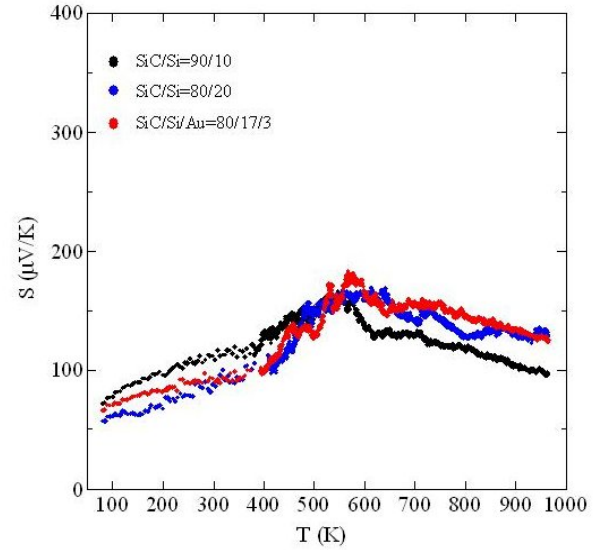


Fig. 4. Temperature dependence of Seebeck coefficient  $S$

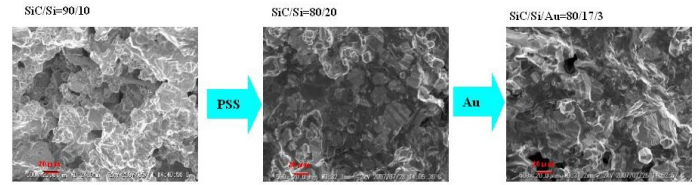


Table 1. electrical resistivity  $\rho$ , electrical conductivity  $\sigma$ , Hall coefficient  $R_H$ , carrier concentration  $n$ , hole mobility  $\mu$ , Seebeck coefficient  $S$ , power

	SiC/Si=90.10	SiC/Si=80.20	SiC/Si/Au=80.17.3
$\rho$ ( $10^{-4}$ $\Omega\text{m}$ )	0.997	0.382	0.336
$\sigma$ ( $10^4$ $\Omega^{-1}\text{m}^{-1}$ )	1.00	2.62	2.98
$S$ ( $10^{-6}$ V/K)	109	88.0	92.4
$S^2/\rho$ ( $10^{-8}$ W/mK <sup>2</sup> )	1.19	2.03	2.54
$R_H$ ( $10^{-8}$ m <sup>3</sup> /C)	3.09	1.99	1.93
$n$ ( $10^{26}$ /m <sup>3</sup> )	2.02	3.14	3.23
$\mu$ ( $10^{-4}$ m <sup>2</sup> /Vs)	3.10	5.21	5.74
$A$ ( $10^3$ kg/m <sup>3</sup> )	1.69	2.10	2.17
$B$ ( $10^{-4}$ m <sup>2</sup> /s)	0.32	0.38	0.37
$C$ ( $10^3$ J/kgK)	0.68	0.68	0.66
$\kappa$ (W/mK)	36.6	53.7	52.4

factor  $S^2\sigma$ , sample density  $A$ , thermal diffusivity  $B$ , specific heat  $C$  and thermal conductivity  $\kappa$  at room temperature.

Fig. 5. SEM observations of the cut surface in the samples with (i) SiC/Au=90/10, (ii) SiC/Si=80/20+PSS and (iii) SiC/Si/Au=80/17/3+PSS.

The results of thermoelectric properties of all samples at room temperature are illustrated in Table 1. We can see the minimum in resistivity  $\rho$ , maximum in power factor  $S^2/\rho$ , maximum in hole mobility  $\mu$  and maximum in sample density  $A$  of the sample (iii). Besides, one can see that Seebeck coefficient  $S$  decreases and thermal conduction  $\kappa$  increases as a function of PSS concentration, on the other hand,  $S$  increases and  $\kappa$  decreases as a function of Au concentration.

These results suggest that both the samples of (ii) and (iii) may be the candidate material of the self-cooling device.

Figure 5 shows the SEM observations of the cut surface in the samples with (i), (ii) and (iii). It can be observed clearly that the grain size increases and porosity decreases with adding PSS. The packing density of the samples in this work is about 53% for without PSS sample (i) and about 66~68% for adding PSS samples (ii) and (iii).

Figure 6 shows the time dependence of the average surface temperature distribution in the self-cooling device. As shown in Fig.1, the self-cooling device consists of commercial power MOSFET connected with the sample of (iii). The sample of (iii) perform the good Peltier cooling of which the bottom side is connected to the Source of MOSFET. Since the electric current (0.6A) is transferred from Drain to Source when the voltage (8V) energizes between Gate and Source, both electric current and the heat flux flows from the bottom to the upper direction in the sample of (iii). It can be observed clearly that the self-cooling device cools down the power MOSFET in Fig.6. The static Drain to Source on resistance of the commercial power MOSFET (e.g. FQP3N90, FAIRCHILD SEMICONDUCTOR Corp.) is 3.3  $\Omega$  [9]. Since the power MOSFET is continuously ON and its current between Drain and Source is 0.6A, Joule heat is generated about 1.19W. On the other hand, the Peltier heat flux of the bottom side of the sample (iii) at 150 second later is given by

$$Q_p = S \times T_H \times I = 92.4 [\mu \text{ V/K}] \times 350 [\text{K}] \times 0.6 [\text{A}] = 19 [\text{mW}]$$

However, the thermal conduction from the bottom side to upper side of the sample (iii) is given by

$$K \Delta T = 524 [\text{mW/cmK}] \times 1 [\text{cm}^2] / 0.2 [\text{cm}] \times 18 [\text{K}] = 47.2 [\text{W}]$$

, where K and  $\Delta T$  are the thermal conductance and the temperature difference between the bottom side and upper side of the sample (iii) at 150 second later. In this case almost all Joule heat generated in power MOSFET can be transferred outside by both Peltier cooling and thermal conduction.

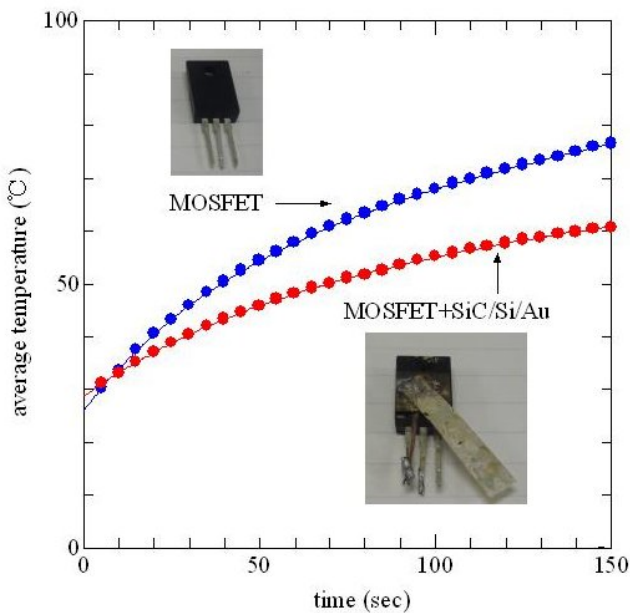


Fig. 6. Time dependence of the average surface temperature distribution in MOSFET (blue solid circles) and MOSFET + (iii) SiC/Si/Au = 80/17/3+PSS (red solid circles).

## Summary

The thermoelectric properties of  $\beta$ -SiC/Si composites are improved by employing PSS sintering additives. The grain size increases and the porosity decreases with adding PSS. As a result, sample density, hole mobility and thermal conductivity increase as a function of PSS concentration. The electrical resistivity at room temperature decrease to about 1/3 with adding PSS. The Seebeck coefficient increases with added PSS at high temperature. The thermal conductivity at room temperature increases with added PSS concentration to about 1.5 times larger than without PSS sample. These results suggest that both the samples of (ii) SiC/Si=80/20+PSS and (iii) SiC/Si/Au=80/17/3+PSS may be the candidate material of the self-cooling device. In fact, we have demonstrated that the self-cooling device using the sample of (iii) cools down the commercial power MOSFET by Peltier effect.

## Acknowledgments

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