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New Segregation Method of Sic During Carbo-Thermal Silica Reduction

Abderahmane Boucetta

Abstract: ***The focus on SOG-Si for solar cell production have been increased in the recent years. A low cost Si requires a production with less electric energy used, less impurities and fast process. Reducing C presence in Si ingots generally is necessary. In the present work, we attempted to develop a new method to segregate SiC from the bulk Si by means a new shape of ingots that we designed. We have used a conventional induction furnace for this process. A comparison study between 2 shapes of graphite ingots was performed. We have succeeded in segregation of SiC from Si during the carbo-thermal reduction process of silica. the results revealed that The first ingot contained 34.3% of SiC while the second ingot had 100% of Si from about 13mm distance from the ingot bottom.***

Keywords **:** ***SOG-Si, Ingots, SiC, Impurities.***

I. INTRODUCTION

Further cost reduction of solar cells requires fundamental improvement of the silicon production process [1]. There are two strategic paths to the silicon production processes for solar cells, that is, solar-grade silicon (SOG-Si), which has the order of 6N-graded purity: One is the way to improve the carbo-thermic reduction process for metal-grade silicon using purified silica and carbon, while the other is the way to improve the yield of the Siemens process of the silicon for the current electronics [2,3]. The former process, which is the direct reduction process, includes only one reduction process by carbon, instead of two reduction processes, by carbon and hydrogen. The direct reduction process, which has the only simple reduction process, is suitable for more production process of silicon, because of its low energy consumption in general. Impurities in SOG-Si is one of the biggest issues involving this process and mainly carbon presence that will effect conductivity [4,5,6]. Using carbon ingots for our induction heating laboratory scale furnace, affect directly the final products and will require another segregation process of SiC from the produced silicon. To reduce the presence of SiC, we have designed a new shape of ingots to use for the induction furnace and a comparison study between the 2 ingots will discussed.

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***** Correspondence Author
Abderahmane Boucetta*, Graduate School of Engineering and School of Engineering, Nagoya University, Nagoya, Japan.

II. MATERIALS AND METHODS

Fig. 1 shows the setup used for the comparison between the 2 ingots, (a) shows the conventional ingots and (b) the new proposed design. The idea is to check the effectiveness of segregations vertically based on weight mass of SiC and Si.

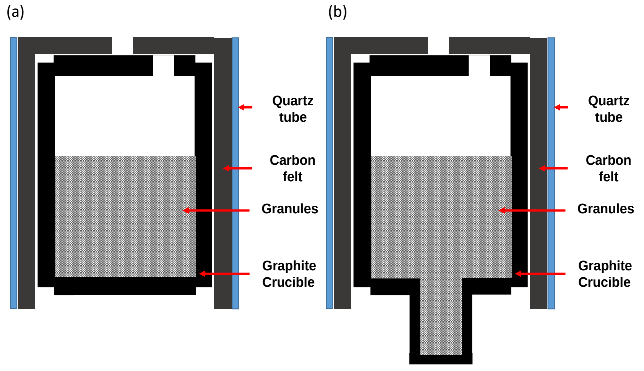

Fig. 1 Schematic design of the 2 ingots used for the reduction

Same amount of granulated Silica and Carbon were used. The feed powders for all the granulation were prepared by mixing with a ratio of 4 parts silica (diameter 20~100 µm, Taiheiyo cement corporation, Japan) to 3 parts carbon (diameter 20 µm, Tokai Carbon, Ltd) by molar. Polyvinyl alcohol (PVA) $[CH_2CH\ (OH)]_n$ (Kuraray Co., Ltd. Japan) was used as binders as shown in Table 1.

Table. 1: quantities used for the experiments

	Volume (cm^3)	Granules (g)	m/V
Experiment	408.67	410.2	1.0037

The granules and 6H-SiC powder were alternatingly supplied in layers into a high-purity graphite crucible (inner diameter 70 mm, height 175 mm). The crucible was capped with a graphite plate, which has one hole of 10mm diameter to release the generated CO gas. The whole crucibles were covered with a 10mm-thick carbon felt as thermal insulators.

The reaction mechanism of carbothermal reduction of SiO2 by carbon is explained by the Eqs. (1)–(7), [7]

$$SiO_2 + C = SiO + CO \quad (1)$$
$$SiO_2 + 3C = SiC + 2CO \quad (2)$$
$$2SiO_2 + SiC = 3SiO + CO \quad (3)$$
$$SiO + 2C = SiC + CO \quad (4)$$
$$SiO + SiC = 2Si + CO \quad (5)$$
$$SiO_2 + Si = 2SiO \quad (6)$$
$$3SiO_2 + 2SiC = Si + 4SiO + 2CO \quad (7)$$

All of the Si production can be reached only through SiC form.

So, in the stationary state of the Si production, a constant quantity of SiC exists in the furnace. In order to emulate the furnace of the stationary state, the reduction process was carried out by adding SiC powders into the furnace. A partial amount of SiC is consumed on the Eqs. (7), (5) and (3), while a partial amount of SiC is generated on the Eqs. (2) and (4). [8,9,10]

The reduction process was carried out by an induction heating furnace of a 40kW power (Toei Scientific Industrial Co., Ltd.) as shown in Figure 2.

Fig. 2 the induction furnace used for carbo-thermal reduction

The evacuation system of this apparatus is composed of a rotary pump and a diffusion pump, which can reach to 10-3Pa in vacuum. During the reduction, the top and side temperatures of the crucible were monitored by high sensitive 2-color type infrared thermometers with a temperature range from 773.15K(500°C) to 3773.15K(3500°C) (IR-CAQ, CHINO Corporation, Japan) through the glass window of the chamber, and the chamber gas was analyzed by a quadrupole mass spectrometer.

Figure 3 shows the heating time was 20 minutes and the maximum temperature of the top of the crucible was around (2373.15 K) 2100°C. It is noted that below the target temperature of 736.15K (463°C) the thermometer cannot measure it .

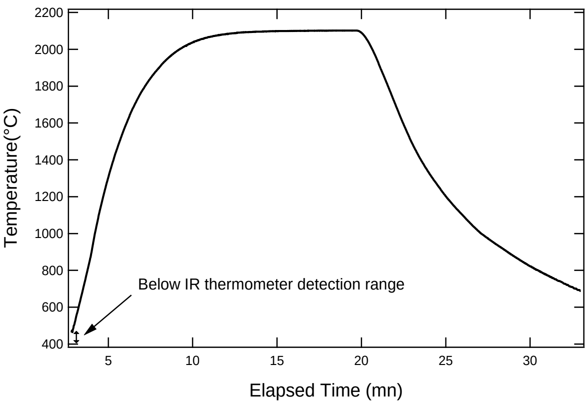

Fig. 3 Temperature profile used for both experiments

After the reduction, the products were milled using a planetary micro mill pulverisette 7 classic line (Fritsch, Germany). The analysis of the X-ray diffraction (XRD) with Cu-*Kα* (λ= 1.5405 Å) radiation source over the angular range of 20°≤ 2θ ≤ 140° and a scan rate of 10°/min was employed by an X-Ray Diffractometer (SmartLab, Rigaku Corporation) to examine the phase composition.

III. RESULTS AND DISCUSSION

Figure 4 shows the images of the final produced Silicon after the reduction process. Slightly different colors can be noticed between both products. In experiment (B), we have cut the additional bottom part of the ingot for vertical analysis of the carbon and SiC presence.

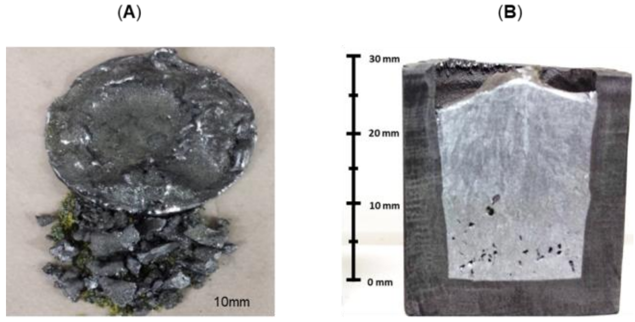

Fig. 4 the produced Silicon from both experiments

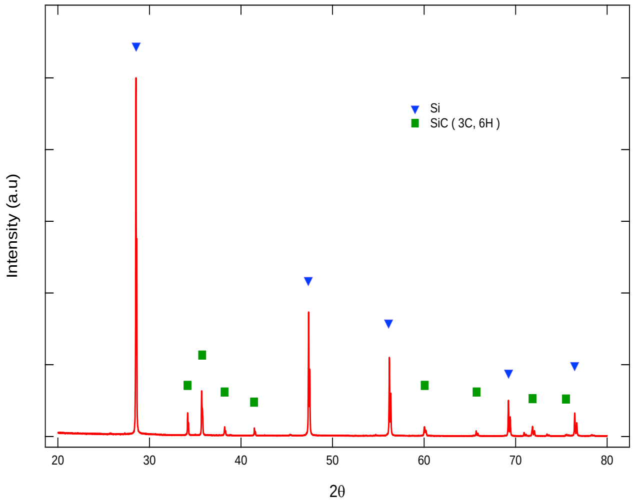

Fig. 5 XRD patterns of experiment (A)

XRD results of the products from experiment (A) shows the presence of SiC with both its types 3C and 6H. Table 2 presents the percentage of SiC in the Si. About 34% of SiC was found in the bulk.

Table 2. Percentage of SiC and Si in experiment (A)

Si (%)	SiC (%)	3C-SiC (%)	Si Yield (%)
65.70	28.08	6.20	33.90

Table 3. Percentage of SiC and Si in experiment (B)

	1	2	3	4	5
Si (%)	41.39	69	72.2	100	100
SiC (%)	58.6	31	27.8	0	0

Table 3 and Figure 6 shows the quantified products percentage from XRD patterns of the obtained products after the milling. It is observed that major SiC started to decrease after 13mm from the bottom and reached to 100% SiC segregations .

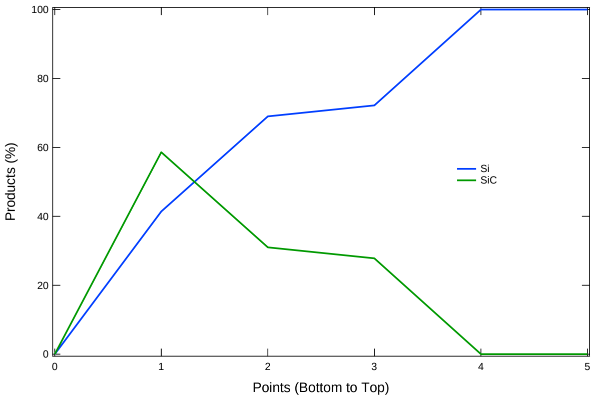

Fig. 6 distribution of SiC and Si along the vertical growth of the ingot of experiment (A)

The products are including two phases of silicon carbide, which are 6H-SiC and 3C-SiC. It is difficult to identify the silica peaks that we have seen in the figure 7. The results of XRD measurement are presented in figure 7. The silicon yield was 33.90% for the PVA experiment. The remained silica is about 3.4% of weight in the experiment where CMC used. The presence of the two SiC phases is explained by two possibilities: firstly, the formation of 3C-SiC is reported to be around 1500°C [11, 12] and it remain stable up to near 2100°C. the 3C-SiC start to change it's phase to 6H-SiC from 2100°C. In the present study, the temperature was around 2100°C in the inner bottom of the crucible. Therefore, this is more realistic in our experiments. Secondly, the 6H-SiC could be remained from the initial amount loaded [13]. It is reported that the segregation of this SiC phases is possible by imposition of high frequency magnetic field to the molten silicon [14].

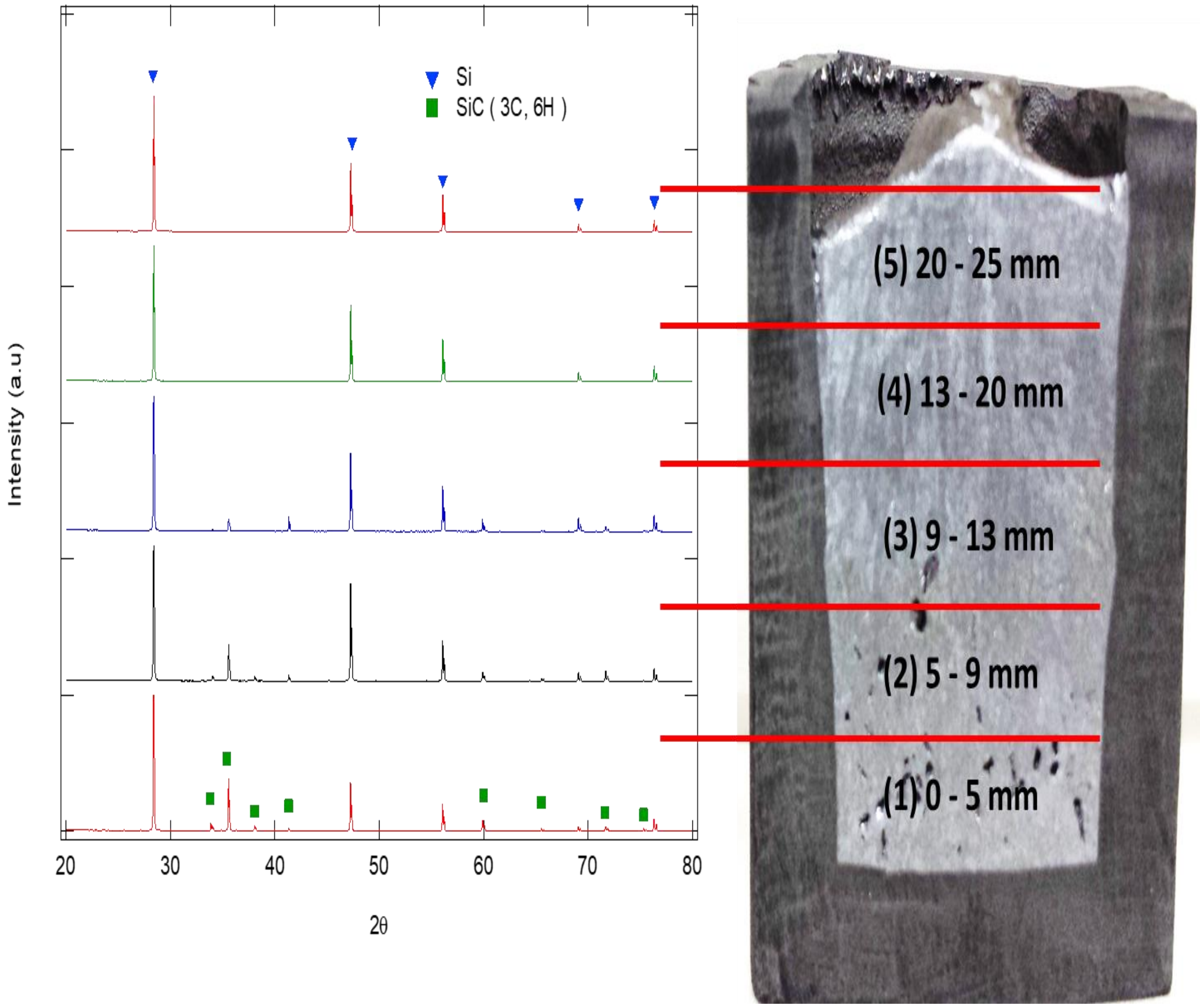

Fig. 7 XRD patterns of locations along the vertical growth of the ingot of experiment (A)

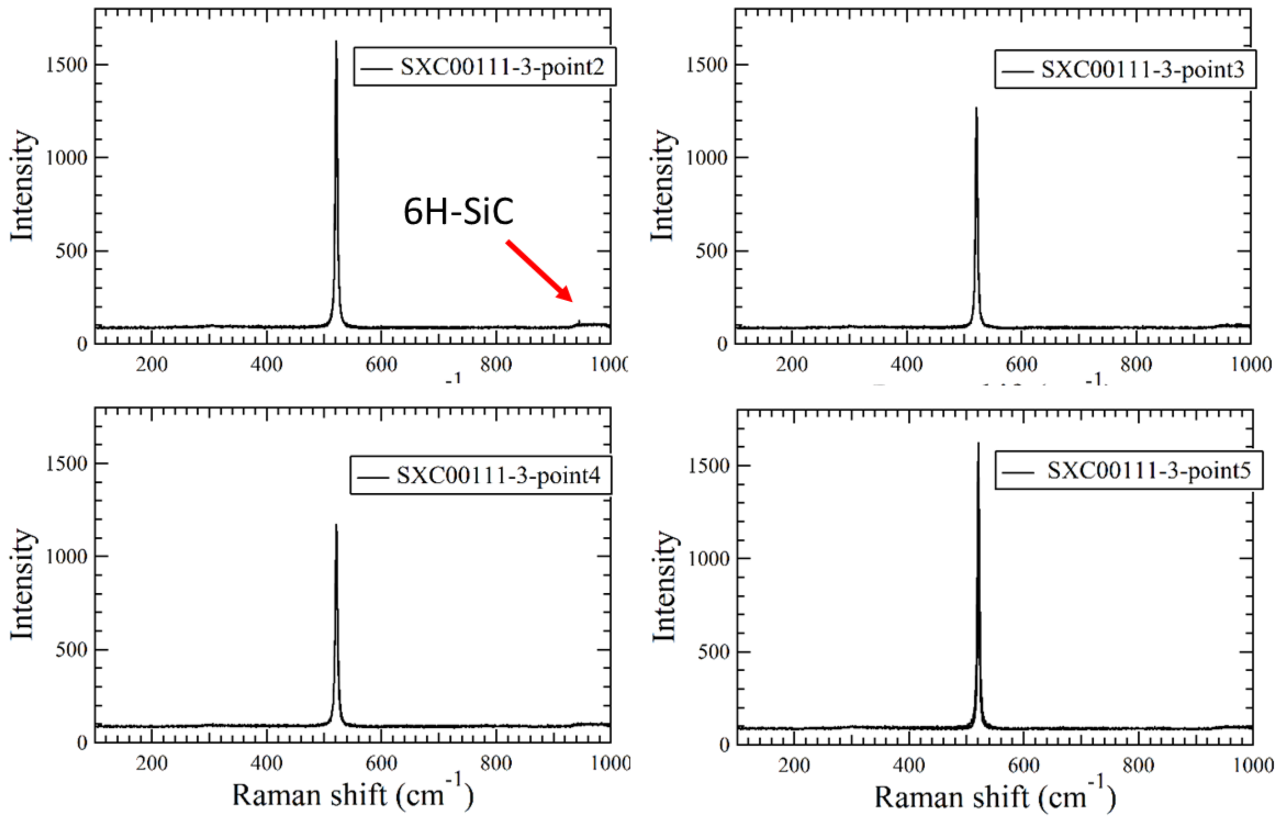

Fig. 8 Raman spectroscopy of locations along the vertical growth of the ingot of experiment (A)

Figure 8 shows Raman spectroscopy results of experiment (B) have confirmed the XRD ones and only the peaks of Silicon are present in the upper parts of the ingots. This have revealed that the new design has successfully segregated SiC from bulk silicon.

IV. CONCLUSION

We successfully produced silicon in the present study, in a laboratory induction-heating furnace. This is based on a new method using pure SiO_2 and carbon. The silicon yielding is about 33.9 % .

The new design of the crucible have successfully helped us segregate SiC from Si during the reduction process.

the results revealed that The first ingot contained 34.3% of SiC using conventional ingots while the new design of ingot had 100% of Si from about 13mm distance from the ingot bottom. This method will be very useful for industrial segregation processes

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AUTHORS PROFILE

Dr Abderahmane Boucetta received his PhD in Science from Hirosaki University, Japan. He worked on Silicon production for solar cells and involved to develop new techniques for low cost SoG-Si. He published several papers on the carbothermal reduction process of silica to produce silicon. Currently working at Nagoya University as a Researcher and working on using data science to study dislocations in silicon ingots and growth optimizations.