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SiC formation and SiO reactivity of methane at high temperatures

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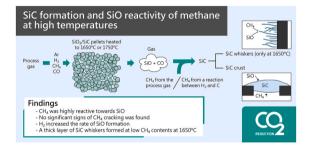
HIGHLIGHTS

G R A P H I C A L A B S T R A C T

- \bullet The presence of hydrogen increased the reaction rate of: $2SiO_2+SiC=3SiO+CO.$
- Methane appeared to be highly reactive towards SiO gas.
- A large amount of SiC whiskers formed in experiments at 1650 °C.
- Methane could be added to the process gas without causing significant thermal cracking.

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ABSTRACT

Methane (CH₄) is a carbon source currently not in use in the production of silicon. Using a gaseous carbon source instead of conventional solid carbon sources presents an opportunity to rethink silicon production. Preliminary research into this topic has shown CH4 to have a very high SiO reactivity and it might even be a step on the way towards a closed silicon furnace. In this study SiC formation from SiO gas in CH4 containing atmospheres is investigated. A reference gas of pure Ar was compared to H2, CH4 and CO gases. SiC was produced by passing CH4 containing process gases through a layer of SiO producing pellets gas at 1650 $^\circ$ C and 1750 $^\circ$ C. A thermocouple, which measured the process gas temperature, was used to look for signs of thermal cracking of CH₄. CH₄ contents up to 8% was tested, the lack of a correlation between CH₄ content and temperature showed that CH₄ does not crack in the current setup despite the temperature being in the range 1650 $^\circ$ C–1750 $^\circ$ C. With a CH₄ containing process gas, most of the SiC formed around the gas inlets and within the SiO producing raw material layer. The reaction between SiO and CH₄ occurred instantaneously when the two gases met, and appeared to be favored over thermal cracking of CH₄. At 1650 °C in H₂ or CO containing process gases a thick layer of whiskers formed around the rim of the crucible. The whiskers were examined with STEM using EDS and EELS, which determined the whiskers to be made of SiC. These results suggest that relatively high CH₄ pressures can be metastable at temperatures far away from the thermodynamic equilibrium. They also indicate CH₄ to exhibit a very high SiO reactivity, which makes it a promising alternative for current carbon sources in the production of silicon

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1. Introduction

$$\mathrm{SiO} + 2\mathrm{C} = \mathrm{SiC} + \mathrm{CO} \tag{1}$$

Commercial silicon production is done in submerged arc furnaces. During the process, carbon materials and quartz is transformed to silicon carbide and SiO gas, which is then transformed into elemental silicon. Reducing the quartz requires large amounts of electric energy in addition to carbon. The main carbon sources used in silicon production is coal, char, charcoal and woodchips. The initial role of carbon is to react with SiO gas according to Reaction (1). A high reactivity between SiO gas and the carbon materials are important because losing SiO from the charge results in lower energy efficiency [1]. How well carbon reacts with SiO gas is termed SiO reactivity, and is an important property of carbon in silicon production.

Silicon production currently only utilizes solid carbon sources, but recently an interest has developed in using carbon containing gases such as natural gas and bio-methane [2]. A gaseous carbon source has several advantages compared to the currently used solids. They are low in metallic impurities, which makes refining easier. They do not contribute heat to the top of the furnace, because Reaction (2) is endothermic, and may lower SiO loss. An added effect of using natural gas or bio-methane is that it moves silicon production from the Si–O – C system, into the Si–O – C–H system, and research thus far has shown that the presence of H₂ tends to speed up the silicon producing reactions [3]. H₂ also presents itself as a new valuable byproduct, which can either be sold or converted into electric energy.

$$2CH_4 + SiO = SiC + 4H_2 + CO$$
⁽²⁾

At the necessary temperatures for silicon production it is suggested that methane (CH_4) has an extremely high carbon activity [4,5]. This indicates that CH₄ likely also has a very good SiO reactivity. The challenge is harnessing this potential. When CH₄ is heated it decomposes into hydrogen and carbon in a reaction referred to as thermal cracking. As a result, some studies faced difficulties getting the CH₄ to react with SiO [6,7]. Various strategies have been employed to suppress cracking. One is using a gas mix of CH_4 and H_2 in an attempt to shift the CH_4/H_2 equilibrium towards CH₄ [6,7]. Because the temperature for CH₄ cracking is surface dependent, careful material selection is another possible approach [8]. Using cracking as part of the process has also been attempted, for example by depositing carbon on quartz and using that as the raw material, or through densification of charcoal [9,10]. Despite the challenges, several researchers have successfully produced SiC from a CH₄ containing gas [7,11]. One of the main reported challenges was blockage and subsequent pressure buildup within the setup due to cracking or build-up of a SiC layer. Using CH₄ as the carbon source in silicon production is not straightforward and likely requires altering the current process or designing an entirely new one. However, there is interest in finding a new process. For example, the idea of a closed silicon furnace is often discussed, but it faces challenges with handling the SiO gas [12]. In this regard, use of CH₄ is a new way of capturing SiO gas, which might have a role to play.

$$SiC + 2SiO_2 = 3SiO + CO$$
(3)

This study aims to investigate how a CH₄ containing process gas interacts with SiO and forms SiC at 1650 °C and 1750 °C, and will investigate the reaction products that forms. SiO₂ and SiC pellets were used to produce SiO and CO gas according to Reaction (3). Ar and CO was added in addition to CH₄. Ar as a reference, and CO because it is present during silicon production. Because this study uses CH₄ at a high temperature, it will also look for signs of cracking to verify that the gas is able to reach the reaction chamber without cracking. To mitigate cracking, H₂ was mixed with CH₄ prior to being injected into the setup, and only CH₄ contents of up to 8% was used. 10% CH₄ was tested but completing the holding time before the setup clogged was not possible.

2. Experimental

2.1. Raw materials

SiO₂ and SiC was used to produce SiO and CO gas according to Reaction (3). The raw material came in the form of pellets with a 1:2 M ratio of SiC and SiO₂. The SiC powder was provided by Washington Mills, it was at least 95% pure with the mayor impurities being SiO₂, Al and Fe. The quartz was the same as "Quartz type A" used by Jusnes and contains more than 99.9% SiO₂ [13]. A Turbula Type T2C Shaker Mixer was used to mix the powders for 6 h. Afterwards, they were pelletized in a steel drum using water as the binder. Finally, the pellets were dried at 120 °C for 6 h and the heated to 1200 °C for 30 min to increase strength.

2.2. Setup

Fig. 1 is a drawing of the experimental setup. It is made of graphite, with the exception of the alumina gas lance. The top section is the condensation chamber, and the bottom section is the reaction chamber. The crucible was placed in the reaction chamber. The raw material was placed on a disk 1 cm above the bottom of the crucible. The gas lance goes through a central hole in the disk and injects the gas below the raw material. After flowing through the reaction chamber, the gas leaves the setup through the condensation chamber where the leftover SiO is transformed to SiO₂ and SiC. A thermocouple was placed within the gas lance at the height of the graphite disk to monitor the temperature.

The setup was heated in a vertical graphite tube furnace. The heating step lasted for 30 min. It was then held at either 1650 °C or 1750 °C for 1 h or until a blockage occurred. During both the heating and the cooling step 1 slpm (litres per minute at 1 atm of pressure and 25 °C) of Ar was passed through the lance. During the holding step 1 slpm of the process

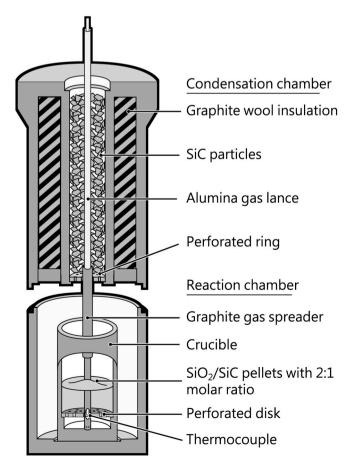


Fig. 1. Experimental setup used during the high temperature experiments.

gases shown in Table 1 was used. The experiments that terminated prematurely due to blockage are written in a parenthesis in Table 1, and as a cross in Fig. 2.

2.3. SEM and STEM

A SEM (Zeiss - Supra 55VP, Scanning Electron Microscope) was used to examine the crucibles for reaction products post experiment. The SEM was equipped with an EDS (Energy Dispersive X-ray Spectroscopy) detector, which performed spot analysis to determine the chemical composition of the reaction products.

SiC whiskers was examined in a STEM (Jeol - JEM ARM200F, Scanning Transmission Electron Microscope). The whiskers were obtained from an experiment at 1650 °C using 100% H₂ as the process gas. They were removed from the crucible with a tweezer and prepared for STEM analysis by dispersing the whiskers in isopropanol using an ultrasonic bath. The dispersion was then dripped onto a TEM grid. In the STEM the whiskers were imaged and the chemical composition were examined with EDS and EELS (Electron Energy-Loss Spectroscopy).

3. Results and discussion

3.1. Effect of CH₄ on weight loss

Adding CH₄ to the reaction chamber resulted in SiC formation through Reaction (2) when it reacted with SiO gas. Weight loss was caused by the SiO and CO generating raw material being consumed. This means that a greater gas production leads to a greater weight loss. Fig. 2a shows the total weight loss with an Ar process gas compared to H₂/CH₄ process gases. H₂ resulted in increased gas production, and only small amounts of SiC formation, which has also been seen elsewhere [3, 14]. This is believed to be the result of small amounts of CH₄ forming from the carbon and hydrogen in the system, which in turn improves the kinetics of reaction (3). Further increasing the CH₄ content by adding CH₄ directly to the process gas decreased the weight loss at 1650 °C. When coupling this with visually observing SiC reaction product it shows that CH₄ promotes SiC formation. Fig. 2b shows that adding 25% CO to the process gas suppressed the increased gas production caused by H₂. However, the increased SiC formation at 1650 °C caused by CH₄ persisted.

The location and type of SiC deposition depended on the CH₄ content and temperature. At 1650 °C the SiC crust formed mainly within the pellet layer, close to the bottom of the raw material or along the lance. Additionally, a layer of SiC whiskers formed around the top of the crucible. It was up to several millimeters thick, still it only made up a small amount of the SiC mass due to the its low density. The main part by weight of SiC formation was the SiC, which formed within the raw material layer, and directly above. At 1750 °C SiC formation was limited to the regions on the perforated disk surrounding the gas inlets. This is in contrast to what was seen earlier [14], where large amount of SiC formation occurred at the crucible wall at both 1650 °C and at 1750 °C. However, the experimental setups differed in how gas was injected. The current setup injects gas from below the raw material, while the one used previously injects the process gas above the raw material. When the

Table 1

Experimental matrix for high temperature experiments. Numbers without parenthesis is the number of experiments that completed the full holding time, numbers in parenthesis is the number of experiments that were terminated before completing the holding time.

	Argon	100% H ₂	5% CH ₄ 95% H ₂	8% CH ₄ 92% H ₂	25% CO 75% H ₂	5% CH ₄ 25% CO 70% H ₂
1650 °C	2	2	2	1+(1)	2	2
1750 °C	2	2	3	(1)	(3)	(2)

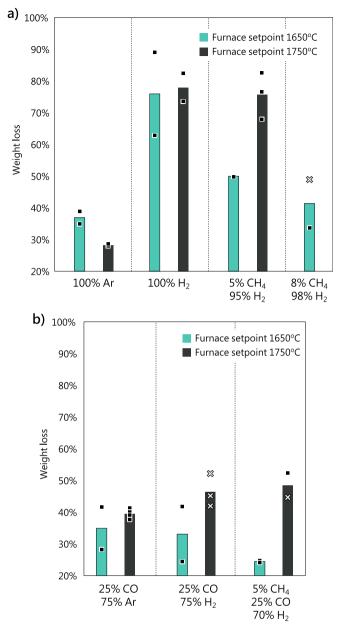


Fig. 2. Weight loss relative to the starting raw material weight (a) for Ar and H_2 with up to 8% CH₄, and (b) Ar, H_2 and H_2/CH_4 in the presence of CO. The datapoints show the measurements for each experiment, the datapoints marked with squares completed the full temperature program, while the crosses are from experiments that were aborted before completing the 60 min holding time.

process gas was injected in this manner SiC mainly deposited on the lance and on the crucible side. In contrast, when gas injection comes from below the raw material CH₄ and SiO reacts within the pellet layer. Earlier work showed formation of SiC whiskers to depend on the CH₄ partial pressure being within a certain range. A low amount of SiC whiskers formed when using 100% H₂ or H₂ with 5% CH₄, whereas a large amount of SiC whiskers formed in H₂ with 2% CH₄ [14]. However, the current setup did not have this behavior, at 1650 °C a layer of SiC whiskers formed for all ratios of CH₄ and H₂ tested, including 100% H₂. The reason the layer of SiC whiskers formed with 100% H₂ might be because the graphite lance and H₂ reacts and generates a sufficient CH₄ pressure for SiC whisker growth. In fact, it was seen visually that some graphite parts were partially consumed by the flowing H₂. The reason SiC whiskers still formed up to 8% CH₄ could be that the CH₄ gas quickly

reacted upon entering the pellet layer, so the gas leaving the raw material was mostly depleted of CH₄, ad as a result had a suitable CH₄ content for SiC whisker growth.

3.2. SiO reactivity of CH₄ and cracking

The temperature measurements and SEM analysis suggests that CH_4 will, if given the chance, react with SiO instead of cracking. All the experiments with CH_4 in the process gas experienced SiC formation surrounding the gas inlet. Fig. 3 shows the SiC that formed around the gas inlets. Because this did not occur in the absence of CH_4 , the SiC must have formed through Reaction (4).

$$2CH_4 + SiO = SiC + CO + 4H_2$$

$$\Delta H_{\rm T} = 1700 \, {}^{\circ}{\rm C} = 101.03 \, \, {\rm KJ} \tag{4}$$

 $CH_4 = C + 2H_2$

$$\Delta H_{\rm T} = 1700 \,^{\circ}{\rm C} = 87.71 \,\,{\rm KJ} \tag{5}$$

The temperature within the lance was measured during most experiments, Fig. 4a shows these temperature measurements. At equilibrium, there should be virtually no remaining CH4 in these conditions due to cracking, but this was not the case. Because Reaction (5) is endothermic, the CH₄ cracking in the lance should result in a lower temperature when more CH₄ is added, which is not seen in Fig. 4a. To confirm that the CH₄ did not crack higher up in the lance, a temperature gradient within the lance was also measured. It was measured with and without process gas flow and found no indication of CH₄ cracking. Fig. 4b shows temperature measurements from earlier experiments where the temperature was measured in the volume where SiO and CH₄ reacted [14]. It shows a trend of lower temperature for increased CH₄ content. This temperature drop is a result of Reaction (4) which is endothermic. Thus, in these conditions CH₄ is metastable and cracking does not deplete the process gas of CH₄ before reaching the SiO containing zone, then when CH4 reaches the region with SiO the gases react immediately.

The weight loss can be used to estimate how much CH₄ and SiO reacted. This is plotted in Fig. 5. It is calculated by assuming that the weight loss in 100% H₂ shows the weight change when no SiO is retained as SiC. Then a line is drawn to the weight loss that would have been observed if all SiO was retained as SiC. Finally, the measured weight loss from CH_4/H_2 atmospheres is plotted on that line. Likewise, for each CH₄ content a line is drawn from the weight loss where no SiO is retained to the point where 100% CH₄ or 100% SiO reacts, then the measured weight losses in CH₄/H₂ atmospheres are plotted on those lines. The line for 8% CH₄ plateaus at a certain point, this is due to there

being in total more CH₄ than needed to convert all the SiO to SiC.

This shows that when the process gas contained 5% CH₄ approximately 44% of the SiO reacted with CH₄, and 51% of the CH₄ reacted with SiO. At 8% CH₄ content 73% of the SiO reacted with CH₄, and 53% of the CH₄ reacts with SiO is 53%. These results suggest it is possible to capture a large amount of SiO gas by maintaining a CH₄ containing atmosphere. Because the SiO capture increases with the CH₄ content it is probable that a setup capable of using higher CH₄ contents without being blocked could capture an even greater amount of SiO.

3.3. SiC whiskers

Process gases containing H_2 , and to a lesser extent CO, promoted whisker formation. The layer of whiskers that formed on the crucible wall was up to several millimeters thick, which is shown in Fig. 6a. It was examined in a SEM, Fig. 6b shows that the whiskers are long and appear intertwined. By using a STEM it was possible to resolve individual whiskers, Fig. 6c shows a STEM image of a whisker. The STEM showed the whiskers to be hexagonal with a diameter from 60 to 200 nm, with an irregularly oscillating thickness. In addition, the whiskers had a number of stacking faults, which is shown by the lines in Fig. 6c. The STEM also performed chemical analysis of the whiskers using EELS and EDS to confirm that the whiskers were composed of SiC. The simplest SiC producing reaction from CO is Reaction (6). Whereas, in the H₂ containing process gases Reaction (4) can also occur. However, in the case where H₂ is added on its own, CH₄ must first form through Reaction (5).

$$SiO + 3CO = SiC + 2CO_2$$
(6)

$$2\mathrm{SiO} + 2\mathrm{CO} = \mathrm{SiC} + \mathrm{SiO}_2 + \mathrm{CO}_2 \tag{7}$$

$$CH_4 + 2SiO = SiC + SiO_2 + 2H_2$$
(8)

An issue with the simple SiC whisker formation mechanism through Reaction (4) or Reaction (6) is that it does not explain why these whiskers only formed at 1650 °C. Work by Broggi et al. and Jayakumari et al. also saw similar SiC whiskers and noted that they did not form at 1750 °C and higher temperatures [15,16]. It is a possibility that formation of this kind of SiC whisker depends on the temperature being below the melting point of SiO₂, which is 1710 °C. In oxide-assisted growth, a SiC core grows simultaneously with an outer SiO₂ shell with a molten tip. This mechanism is known to occur in the formation of white condensate, which forms through Reaction (3) [17]. It is proposed that for atmospheres enriched in CO or containing CH₄, Reaction (7) and Reaction (8) can occur instead of Reaction (3), this produces SiC–SiO₂ core shell whiskers with 1:1 ratio of SiC and SiO₂. Then the SiC core and the SiO₂ shell may react according to Reaction (3) which consumes 2 mol of SiO₂ for every 1 mol of SiC, which results in the SiO₂ layer being

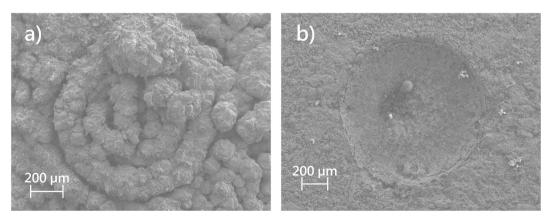


Fig. 3. Gas holes where the process gas first meets SiO are plugged if the CH_4 content is too far away from equilibrium. The type of SiC that forms depends on temperature (**a**) is from 1750 °C where the gas holes are blocked by a solid layer of SiC, (**b**) is from 1650 °C with 10% CH_4 where the gas holes are blocked by a layer of whiskers.

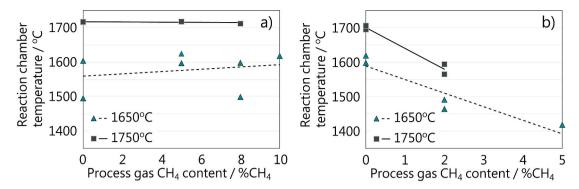


Fig. 4. The reaction chamber temperature from (a) the current study where the process gas was injected below the raw material, and (b) from earlier experiments where the process gas was injected above the raw material [14].

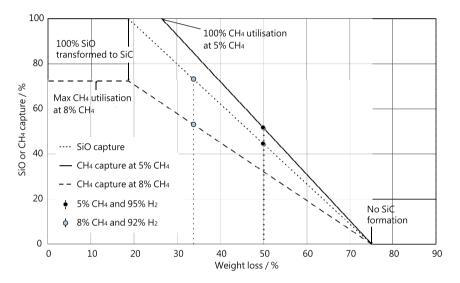


Fig. 5. Amount of CH₄ and SiO that produced SiC at 1650 °C. The SiO formation is calculated based on experiments in 100% H₂, and is used to determine how much of the SiO and CH₄ that reacted for each CH₄ content. The vertical lines indicate the data-points that come from the same experiment.

stripped from the SiC core, which leaves behind a partially reacted SiC whisker.

3.4. Condensation

Condensation, which is the reversed Reaction (3), occurred within the reaction chamber when using 5% CH₄, 25% CO and 70% H₂ as the process gas. The condensate was found deposited on the inner crucible wall. Fig. 7a shows an overview of the condensate, which is a network of treads with a thickness of roughly 1 µm. Fig. 7b shows the condensate at a higher magnification. The condensate grows through the oxide assisted growth mechanism and is made up of a central SiC whisker surrounded by an SiO₂ shell layer. Further down in the crucible, where the temperature is higher, surface tension makes the shell layer contract into roughly spherical globules, this is shown in Fig. 7c. EDS analysis determined the chemical composition of the shell layer to be SiO₂. While the core whiskers were made of SiC. The reason condensation occurred with this process gas might be additional cooling provided by CH₄, as Reaction (4) is endothermic. This may lower the temperature sufficiently for condensation to occur. Because condensate was not seen when only CH₄ and H₂ was used, the increase in driving force for condensation caused by the increased CO content in the process gas may also have played a role.

4. Conclusions

CH₄ containing process gases resulted in formation of a SiC layer where the process gas first encountered SiO generated by the raw material. This left the gas flow exiting the raw material layer mostly depleted of CH₄. This low CH₄ content was suitable for growth of SiC whiskers. The whiskers formed as a layer around the top of the crucible. The SiC whisker layer could grow to a large thickness, but due to its low density it only made up a small part of the overall SiC mass. SiC whiskers only formed in H₂ atmospheres, or atmospheres enriched in CO at 1650 °C, but not at 1750 °C. It is suggested that the temperature limit for SiC whisker growth is the melting point of SiO₂ which is 1710 °C, and that the SiC whiskers initially formed as SiC-SiO2 core shell whiskers before the SiO₂ shell layer was stripped away. Temperature measurements indicate that significant thermal cracking of CH4 did not occur, and the process gas reached the raw material without losing much CH4 due to cracking. The results indicate that CH4 is extremely reactive towards SiO and when the two gases came into contact, they reacted rapidly and produced SiC. Condensation within the crucible were only confirmed for the process gas containing 5% CH₄, 25% CO and 70% H₂, and only at 1650 °C. The condensate formed with a SiC-SiO₂ core shell structure. This indicates the role of the gases are as follows: H₂ allows improved mass transport and increased SiO content. CO provides a greater driving force. CH₄ provides cooling as it reacts with SiO.

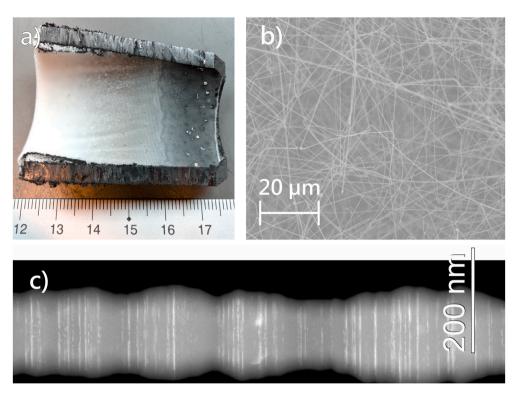


Fig. 6. SiC whiskers formed at 1650 $^{\circ}$ C with a 100% H₂ process gas (a) shows an overview of the crucible and the thickness of the SiC whisker layer, (b) shows an overview image of the whiskers taken with a SEM, (c) shows a BF-DF image of a SiC whisker.

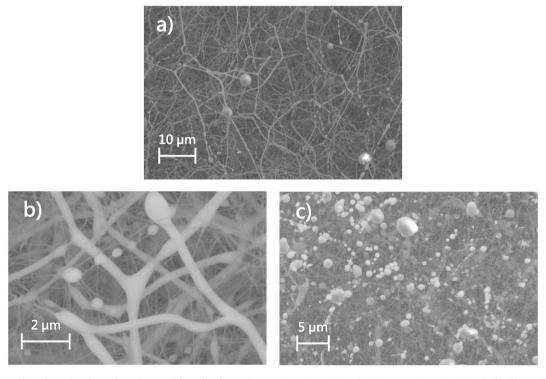


Fig. 7. SEM images of condensation formed on the crucible wall when using 5% CH_4 , 25% CO and 70% H_2 at 1650 °C. (a) and (b) shows SiC–SiO₂ core shell whiskers, (c) is lower in the crucible where the temperature is higher and the shell layer contracted and formed SiO₂ globules.

Credit author statement

T.S.A. performed experimental work, wrote the manuscript and prepared all figures, with feedback from M.T. and E.R. All authors reviewed the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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