

POSSIBLE USE OF NATURAL GAS FOR SILICON OR FERROSILICON PRODUCTION

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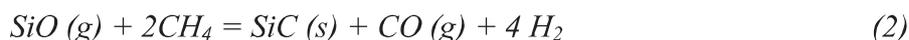
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ABSTRACT

The possibilities for using natural gas in silicon or ferrosilicon production were investigated in a Norwegian competence building project (GASFERROSIL). The overall purpose was to reduce energy consumption and CO₂ emissions. Silicon carbide is an important intermediate product in silicon production, and is formed from hot SiO gas and solid carbon according to reaction (1) above 1512 °C. The basic idea in GASFERROSIL is to utilize natural gas in the production of silicon carbide, according to reaction (2).

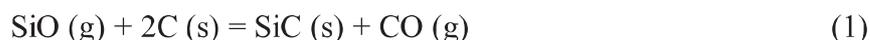


Enabling hot SiO gas to meet cold CH₄ before it cracks into C and H₂, a special water-cooled injection lance was constructed and inserted directly into a reaction chamber at 1600-1650 °C where hot SiO gas is flowing. The efficiency of reaction (2) has been verified at these temperatures. Both the thermodynamics and kinetics of the reaction have been studied for different molar ratios of CH₄/SiO. Excess CH₄ will crack to C and H₂ while the temperature drops. In this way natural gas can also act as a cooling agent, while carbon and hydrogen are being simultaneously produced.

KEYWORDS: Natural gas, methane, SiO gas, SiC formation, Si production.

1. INTRODUCTION

The production of silicon (Si) and ferrosilicon (FeSi) is conducted in semi-closed furnaces allowing air to enter the furnace just above the charge. The oxygen in air sustains the combustion of CO gas from the reduction process as well as oxidation of the gaseous bi-product, SiO. Heat generated from these reactions form the basis for subsequent energy recovery and the fine silica dust is collected in bag filters and sold. Most of the SiO gas generated in the lower part of the furnace reacts with carbon in the charge to form silicon carbide, which in turn reacts with quartz to form silicon, SiO and CO. Hot SiO gas and solid carbon react and form silicon carbide as an important intermediate product in silicon production, according to reaction (1) above 1512 °C:



In GASFERROSIL we have had a closer look at the options available if the off gases are combined with natural gas, but new processes for silicon production have also been considered. A common challenge for all the examples of possible industrial usage of natural gas has been that cold

natural gas must meet hot SiO gas. This challenge has been addressed in the present work together with thermodynamic calculations using FactSage. The main question was: "Is it possible to experimentally investigate what happens when cold CH₄ (g) meets hot SiO (g), or will the methane crack on its way into the furnace and plug the equipment?" A lot is known about how SiO reacts with carbon materials and how it will decompose into silicon and silica at temperatures lower than ~1100°C, but little is known about possible reactions with methane.

Methane, on the other hand, decomposes easily to carbon and hydrogen during heating, starting at around 400 °C while the conversion should be complete at 1000°C, according to thermodynamic calculations by Wærnes et al [1]. In order to enable hot SiO gas to meet cold natural gas before it decomposes to carbon and hydrogen, a special water-cooled injection lance was constructed. The lance was inserted directly into a reaction chamber at 1600-1650 °C where hot SiO gas was flowing.

2. EXPERIMENTAL

The furnace and most of the experimental set-up is the same as normally used for the SiO-reactivity test at SINTEF [2, 3]. A water-cooled lance with 10 mm outer diameter was made for CH₄-injection. Cold CH₄ will meet hot SiO gas in the reaction chamber (red colour) in figure 1. The lance tip is positioned in the middle of the reaction chamber in most experiments.

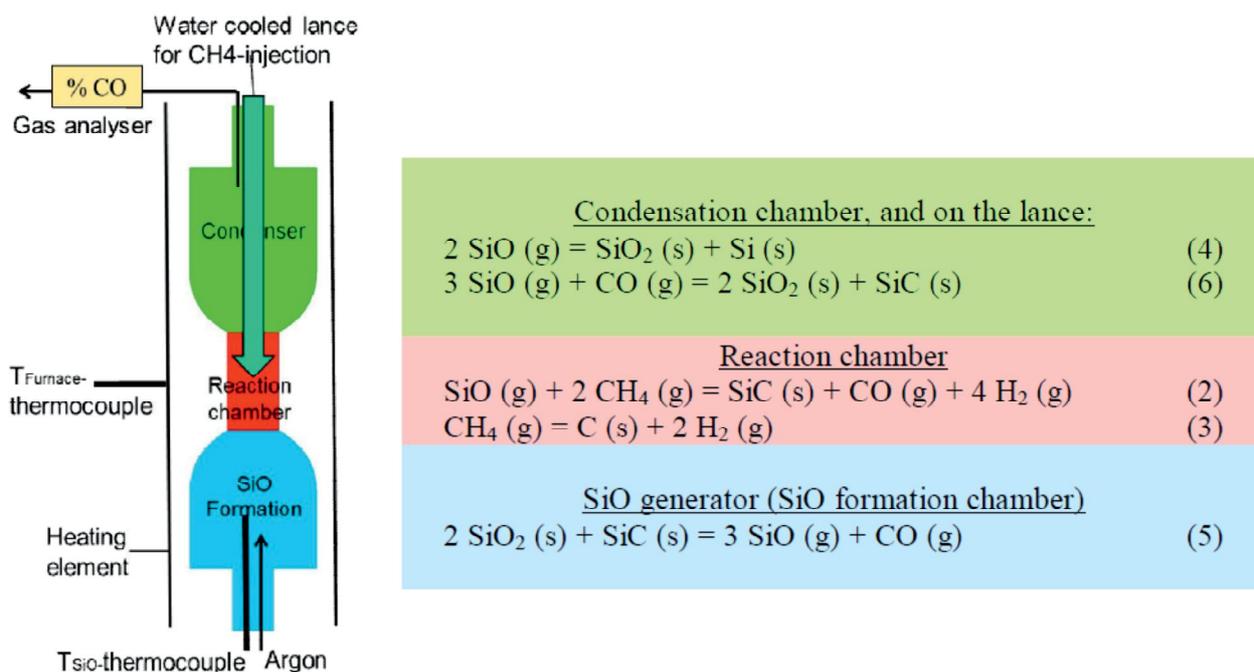


Figure 1: Experimental principle for cold CH₄ to meet hot SiO

The SiO-gas generator (blue chamber) is filled with pellets from which SiO and CO is formed according to equation (5) and Argon (19.2 l/h) is used as carrier gas, as in the SiO test. The hot SiO gas is formed around 1650 °C and flows into the reaction chamber. The weight of pellets before and after each experiment was registered. The amount of SiO gas generated can be calculated from the pellets weight loss or by measuring the CO-level in the off-gas without methane injection.

The reaction chamber (red chamber) is a little wider than the one used in SiO-reactivity tests in order to get enough space for both the water cooled lance and products. The inner tube diameter is 28 mm and the height is 60 mm. Here will cold methane meet hot SiO gas and the reactions (2) and (3) can be studied by measuring the off-gas CO-level and by analysing excavated products. The condensation chamber (green chamber) is not filled with alumina Raschig rings, as in the SiO reactivity test. Condensation takes place on the cold lance, mainly on the part inside the reaction chamber according to reactions (4) or (6).

The temperature was measured in the middle of the SiO generator (T_{SiO}) as in the SiO-test. Both the reaction chamber and the surface of the SiO-pellets may be cooled down by the water cooled lance. Therefore, T_{SiO} was kept stable in the range 1644-1651 °C during methane injection while the temperature in the SiO-test usually is around 1630–1640°C. The furnace temperature ($T_{Furnace}$) was around 1473-1483°C, which is 30-40°C lower than in SiO reactivity tests. In the SiO-test the temperature in the reaction chamber is around 1650 °C. The temperature at the top of the SiO generator was measured to 1600 °C at the start of the injection period, using a longer thermocouple (run 12), while the furnace temperature was kept the same as in the previous run. Towards the end of the injection period the temperature at the top of the SiO generator increased to 1624 °C, while the temperature in the middle had increased to 1650 °C. It is believed that the product formed at this position is insulating the thermocouple and the top of the SiO pellets.

Experimental procedure: The basis of the SiO-reactivity test is that a gas mixture containing 13.5 % SiO and 4.5 % CO is passed through a coke-bed with argon (19.2 l/h) as a carrier gas at around 1650 °C. In the present set-up there is no coke-bed, but a product-bed did build up during methane injection and consequently the pressure increased a little. Methane injection was started when the temperature in the SiO chamber had reached 1555-1625°C and lasted for 60 minutes in most experiments. The furnace was turned off after the injection was stopped. The products in the reaction chamber and on the lance were excavated after the reactor was cooled down. Experiments were also carried out without methane injection. Run 11 and 12 were carried out with pre-used SiO-pellets in order to avoid the CO-peak at the start of the injection period.

Off-gas measurements: The CO-level in the off-gas was measured by a CO gas analyser, which was calibrated before the start of each experiment. A μ -GC was used in some experiments in order to measure hydrogen, CO, CO₂, unconverted CH₄, and to see if other species were formed.

Chemical analyses of products were carried out by MOLAB. These analyses were total carbon (combustion, IR), free carbon (temperature controlled combustion) according to ISO 9286:1997 chemical analysis of silicon carbide, and silicon that was determined by titration according to Fedorovs method. SiC was calculated and SiO₂ was by difference (100 - SiC - C_{free}).

$\% \text{ SiC} = (C_{\text{total}} - C_{\text{free}}) \cdot m_{\text{SiC}}/m_{\text{C}} = (C_{\text{total}} - C_{\text{free}}) \cdot 40.1/12.01 = (C_{\text{total}} - C_{\text{free}}) \cdot 3.339$. The relative standard deviation is approximately 1.0 % for total carbon (medium values) and 5 % for low values of free carbon. Examples are (30.0±0.03) % total C and (1.00±0.05) % free C.

Identification of phases was done by XRD (X-ray diffraction analyses). Semi-quantitative XRD were carried out at NTNU, with a User D8 ADVANCE using a DIFFRAC^{plus} SEARCH software and quantitative XRD was carried out using mathematical modelling. Each sample was milled in sling mill using a WC mortar until all the material passed a 45 μ m sieve. Milling time was only 10 seconds each time in order to avoid that the material becomes semi-amorphous by over-crushing. Samples that were analysed both by XRD and chemical analyses were split.

3. REACTION EQUILIBRIA BETWEEN CH₄ AND SiO

The reaction equilibria between streams of SiO (g) at 1650 °C and CH₄ (g) at 25 °C at arbitrary pressure (1 bar) have been simulated for various molar ratio of CH₄ to SiO at adiabatic

condition (no heat added) as shown in figure 2 using thermochemical data from the FACT databases [4]. Increasing the CH_4/SiO ratio results in lower adiabatic temperature because the carbon black producing reaction (3) is endothermic. For $\text{CH}_4/\text{SiO} = 1.0$ the adiabatic temperature was $1614\text{ }^\circ\text{C}$ and for $\text{CH}_4/\text{SiO} < 0.8$ the adiabatic temperature increases to more than $1670\text{ }^\circ\text{C}$ because of the exothermal condensation reactions (4, 6). An optimal SiC production occurs at a molar ratio of $\text{CH}_4/\text{SiO} \sim 1.26$ for which 0.75 mole SiC is formed at a temperature close to $1380\text{ }^\circ\text{C}$. It is well known that the lowest temperature for SiC production by reacting SiO gas with carbon is around $1512\text{ }^\circ\text{C}$. Consequently, SiC can be produced at a lower temperature by reacting CH_4 with SiO. This is due to the high carbon activity when methane reaches the hot zone which can be of great technical significance for a new SiC or Si process. The gas mixture produced in the SiO generator is close to the $3\text{SiO}-\text{CO}$ mixture. Therefore reaction equilibria at fixed temperatures in the range $1200 - 2000\text{ }^\circ\text{C}$ were also simulated for various molar ratios of $\text{CH}_4(\text{g})$ and streams with $0.75\text{SiO}(\text{g}) + 0.25\text{CO}(\text{g})$ as seen in figure 3.

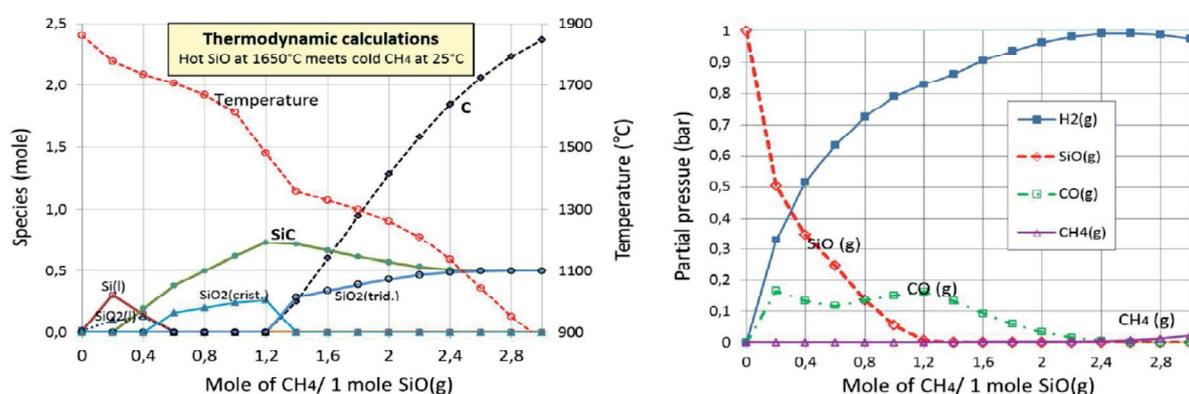


Figure 2: Thermodynamic calculations in the system Si-O-C-H for reactions between cold methane (CH_4 at $25\text{ }^\circ\text{C}$) and hot silicon monoxide (SiO at $1650\text{ }^\circ\text{C}$) for various molar ratios of $\text{CH}_4:\text{SiO}$. No energy added, adiabatic temperature, product species (left) and partial pressures (right) calculated

4. EXPERIMENTAL RESULTS AND DISCUSSION

Table 1 shows chemical analyses and the amount of different products formed in the reaction chamber and on the lance. A mixture of SiC, C, SiO_2 and Si was formed in all the experiments. Lumps were formed at the bottom of the reaction chamber in most experiments with methane injection. In all experiments there were also a lump formed on the lance tip.

The lump on the lance tip usually had a hole in the centre which was formed by methane injected through the lance. This gas-hole was usually larger for the highest flow rates. Pictures of some selected products are shown in figure 4. Notice the crystals grown on the inside of the wall in the reaction chamber in reference experiment 5, without methane injection. Brownish looking crystals seem to grow out from single spots on the wall.

At the left in figure 3 is shown the SiC contours (mole SiC) as a function of both temperature and the $\text{CH}_4/(0.75\text{SiO}+0.25\text{CO})$ mole ratio. The heat changes of isothermal reactions (DH) were also calculated and are shown in the same figure at the right. Dark blue area indicates exothermic reactions, whereas other areas are endothermic reactions.

However, the SiO-gas condensate was mainly found on the water-cooled lance, as expected. There is no gas-hole in the lump from the lance tip of the reference run.

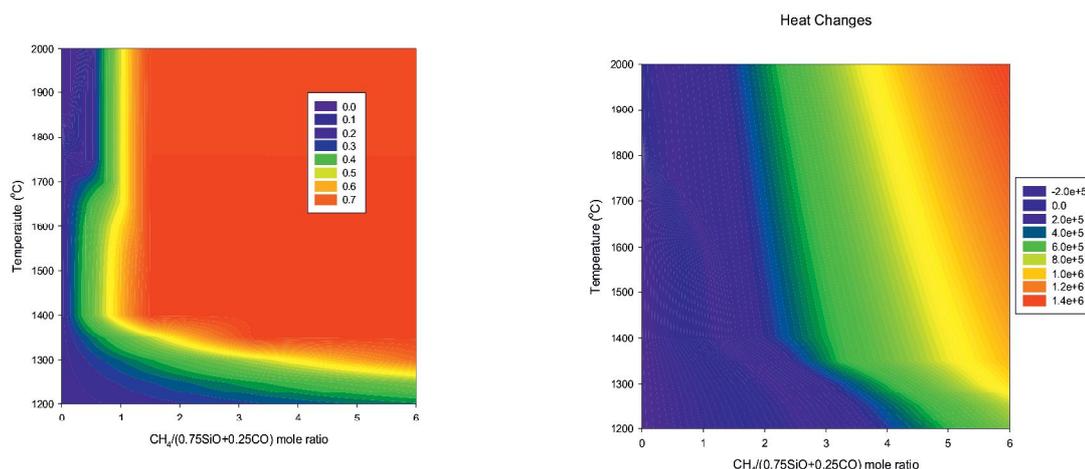


Figure 3: Equilibrium SiC (mole SiC) contours at left and energy changes (DH, kJ) of isothermal reactions at right, both as a function of the $\text{CH}_4/(\text{0.75SiO}+\text{0.25CO})$ mole ratio at 1200-2000°C

Table 1: Chemical analyses and amounts of the products excavated

Run	CH ₄ - injection		Product analysed		Chemical analyses (wt%)			Calculated analyses (wt%)	
	(l/h)	(min)	Position	weight (g)	C total	C free	Si	SiC	SiO ₂
5 ref.	0		Total, analysed	3,13	2,41	0,97	21,4	4,8	72,8
11	2,4	60,0	Rx-chamber	2,49	23,2	12,7	8,3	35,1	43,9
			Lance	2,33	21,1	13	10,8	27,0	49,2
			Total, calc.	4,82		12,8	9,5	31,2	46,5
6	3,8	60,0	Total, analysed	5,83	25,6	14,5	8,6	37,1	39,8
12	6,0	60,0	Rx-chamber	3,86	22,4	10,9	6,7	38,4	44,0
			Lance	2,63	48,9	41,9	5,2	23,4	29,5
			Total, calc.	6,49		23,5	6,1	32,3	38,1
4	9,1	63,0	Rx-chamber lump	2,65	29,5	16,8	2,2	42,4	38,6
			Lance tip lump	2,27	77,5	73,9	1,7	12,0	12,4
			Rest (rx+lance)	1,60	27,2	21,3	6,2	19,7	52,8
			Total, calc.	6,52		37,8	3,0	26,3	33,0

Figure 5 at the left shows the effect of methane flow rate on the total rate of formation (g/h) for the different species (sum of products in the reaction chamber and condensed on the lance). Maximum SiC content was 37.1 wt % for 3.8 l/h methane, corresponding to a production rate of 2.2 g SiC per hour.

The condensed SiO₂ is formed at a rate more or less constant around 2.0-2.5 g/h. Carbon is formed at a steady rate for an increasing methane flow rate while the rate of silicon formation is decreasing. Figure 5 (at right) shows the corresponding molar production rates.

The SiC producing reaction (2) is efficient at low methane flow rates, consuming 2 moles of CH₄ for each mole SiC produced. Condensation reactions (4, 6) take place, but the CB forming reaction (3) dominates at the highest methane injection rates.

However, the competing reactions take place in various degrees depending on the location. The composition of the products grown at the bottom of the reaction chamber and on the lance differs considerably as shown in figure 6.

SiC is mainly deposited in the reaction chamber while carbon is mainly deposited on the lance, especially for the highest methane flow rates. The production rates of both SiC and SiO₂ goes through a maximum around 1.5-1.6 g/h close to 6 l/h methane for the products at the bottom of the reaction chamber where hot SiO-gas is entering. The molar ratio SiC/SiO₂ is close to 1.2-1.3. The carbon production rate is much lower, slightly increasing for increasing methane flow rate to 0.5 g/h, while the silicon production rate is decreasing.

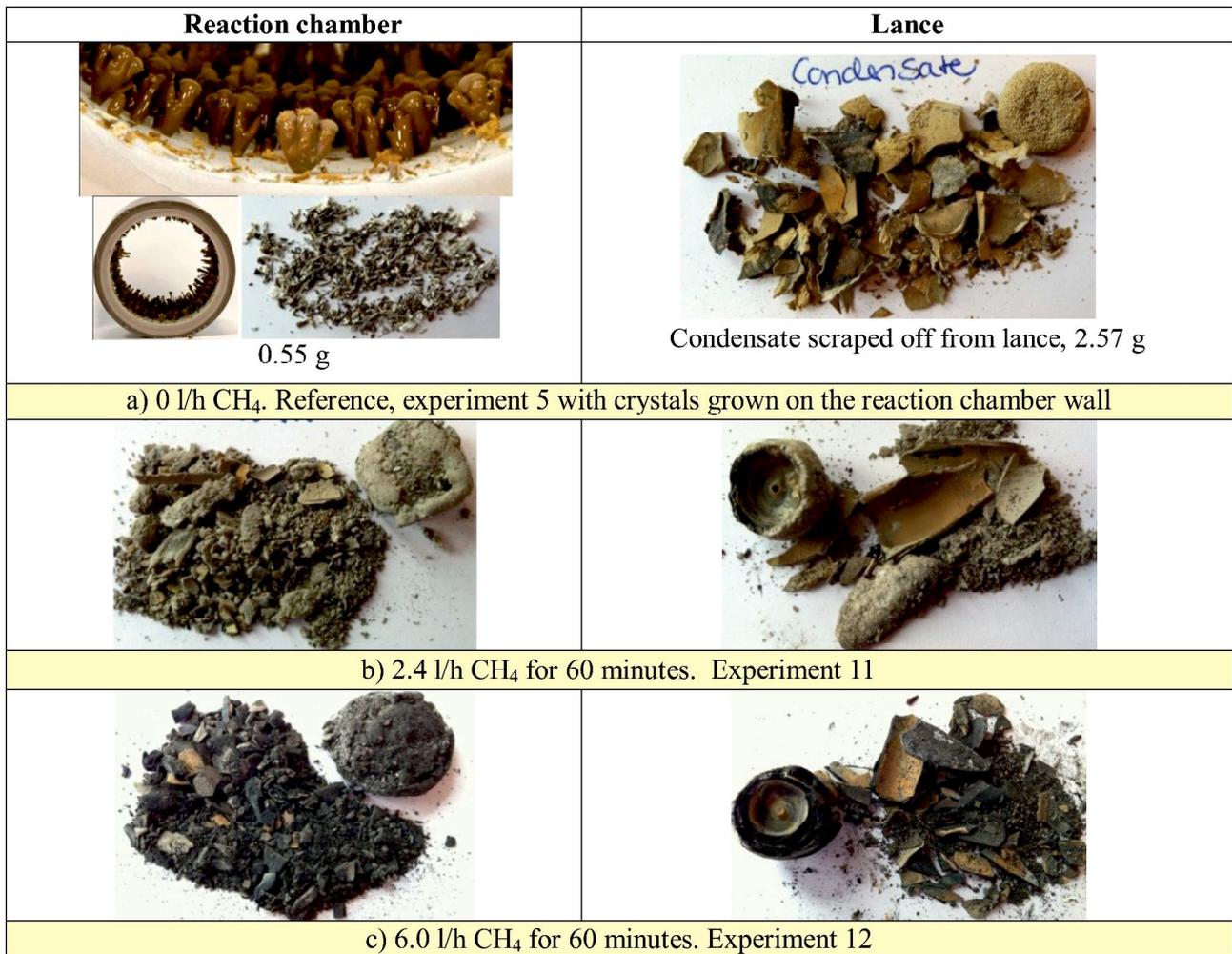


Figure 4: Pictures of excavated products for increasing methane injection

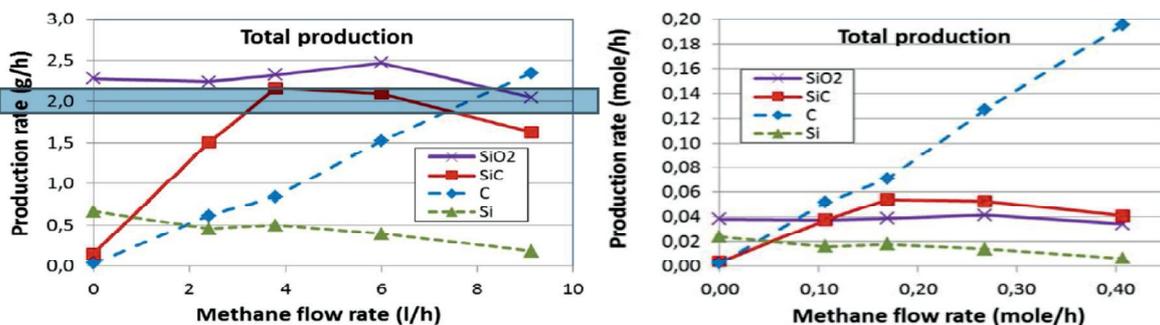


Figure 5: Production rates at the frontier of hot SiO gas at 1600-1650 °C and cold methane

Silicon is formed in the reaction chamber through condensation reaction (4) but at high CH₄ flow rates silicon seems to react with carbon to SiC or condensation reaction (6) is favored. All the samples contained SiC, also references, which was somewhat surprising. The condensation reaction (4) was thought to be dominating when methane was not injected, producing equal amounts of SiO₂ and Si on a molar basis. However, the molar ratio SiO₂/Si is higher than 1.0 (1.6 for run 5). The reference contained 5 % SiC. This may be explained by the pellet back-reaction (6), producing SiC and SiO₂ from the SiO gas by consuming some of the CO.

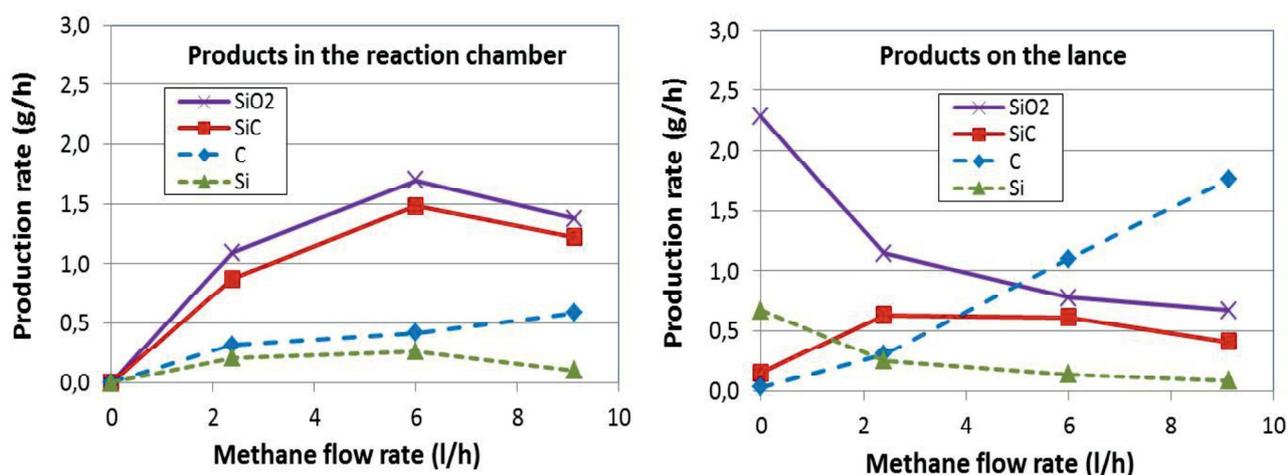


Figure 6: Production rate for products in the reaction chamber (at left) and on the lance (at right)

5. VERIFICATION OF RESULTS BY XRD ANALYSES

Figure 7 shows XRD diagrams of the lumps formed at the highest methane flow rate. SiC was easily identified in the reaction chamber lump and carbon was easily identified in the lance lump! SiC exists in many different modifications, and 3C, 6H, 4H, 2H and 15R were detected.

The results by quantitative XRD (XRD-Q) and chemical analyses were usually rather close, but XRD-Q was rather time-consuming. Amorphous silica was underestimated by XRD Semi-Q, while silicon generally was overestimated. Referring to a comparison of the different analyses of the sample from run 6 in figure 8, XRD-Q may be too low in carbon, while SiO₂ may be overestimated.

This is supported by a material balance. Of these reasons, it was decided to use chemical analyses, combined with the necessary identification of phases by XRD.

6. CO ANALYSES

Figure 9 shows that the flow rate of CO (l/h) is clearly different for different injection rates. The reference stabilises at 0.64 l/h CO after 20 minutes (corresponding to 3.2 % CO, 1651 °C in the SiO generator), while the rate is 3 times higher (1.7-1.8 l/h CO) when 6-9 l/h CH₄ is injected. The rate is twice as high (1.2 - 1.4 l/h CO) for 2.4 - 3.8 l/h CH₄, but it is declining towards 1.0 l/h CO at the end. The flow rate of CO was calculated from % CO taking into account that 2 moles of H₂ always will be created from each mole of CH₄ and that the flow rate of argon was constant (19.2 l/h).

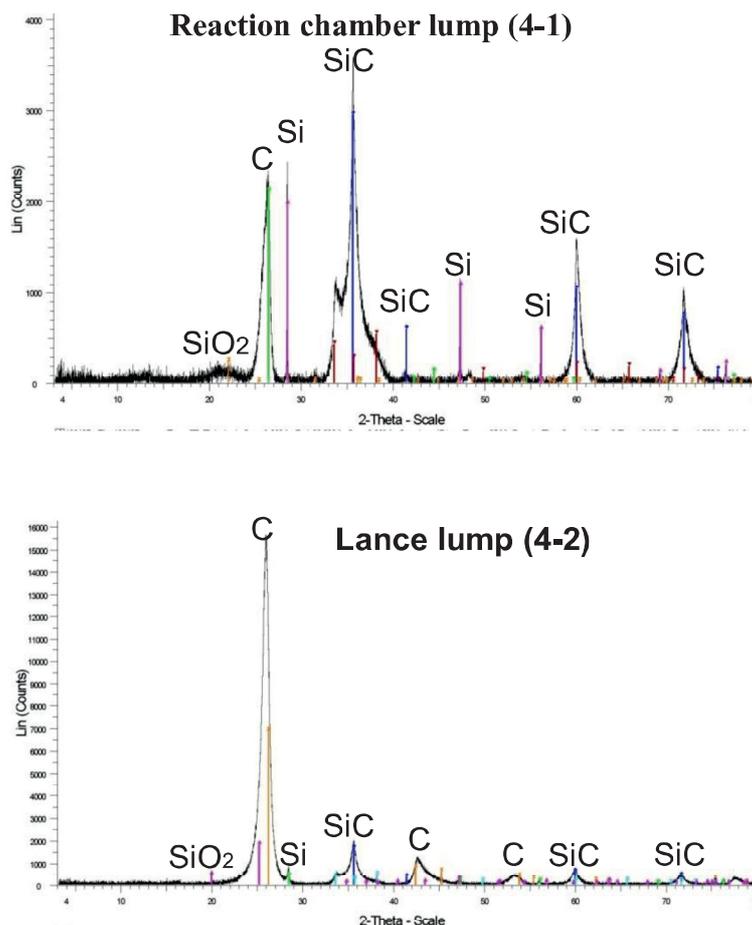


Figure 7: XRD diagrams of the lance tip lump and the reaction chamber lump. Run 4 (9.1 l/h CH₄)

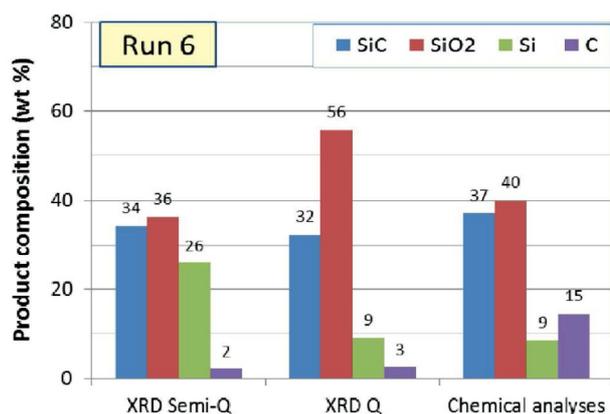


Figure 8: Comparison of different analyses for the total sample, using 3.8 l/h methane

Table 2 sums up these results by listing the flow rates of CO (l/h) which were calculated from % CO measurements. Both the mean CO flow rates realized (CO_{mean}) and the values towards the end of the experiments are steadily increasing with increasing methane flow rate. The table includes the gas formed in the SiO-generator (3 SiO + CO_{pellet}, reaction 5), based on the measured pellet weight loss in each experiment.

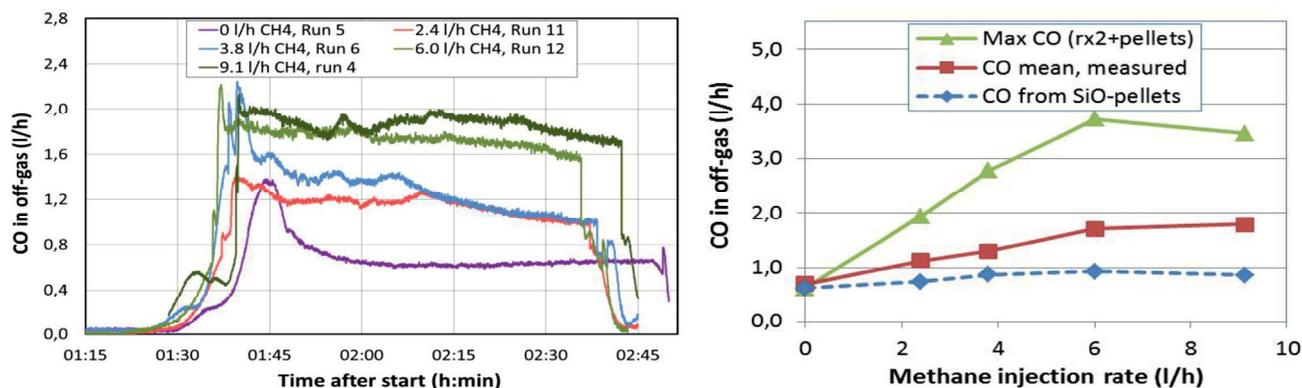


Figure 9: At left: Flow of CO in the off-gas during CH₄ injection, compared to the reference. At right: Realized flow rate CO (l/h) (mean values) compared to CO generated from the SiO-pellets and the maximum flow rate for complete conversion of reaction 2 (rx2)

Calculated theoretical off gas composition (mole/h) for maximum CO formation (CO_{max}) is also shown for complete conversion of SiO or CH₄ according to reaction (2), which is dependant on the molar ratio of CH₄/SiO. Figure 9 at the right shows the increasing CO_{mean} for increasing methane flow rate, compared to CO_{pellet} and CO_{max} which seems to level off at an injection rate of 6 l/h methane. The share realized (x) of the maximum CO formation was found in the range 0.46-0.58, or in average 52 %. Clearly, SiC has been formed!

$$x = \text{CO}_{\text{mean}}/\text{CO}_{\text{max}} = \text{CO}_{\text{mean}} / (\text{CO}_{\text{pellet}} + \text{CO}_{\text{max rx2}})$$

CH₄/SiO=0: Reference;

CO = CO_{pellets}, generated from SiO-pellets, based on pellet weight loss.

CH₄/SiO<2: Run (6, 10, 11);

CO = CO_{pellets} + ½ CH₄ (molar basis). CH₄ limiting for reaction (2).

CH₄/SiO>2: Run (4, 12);

CO = CO_{pellets} + SiO_{pellets} (molar basis). SiO is limiting for reaction (2).

Table 2: Off-gas flow rates measured and calculated provided a complete conversion of reaction (2). Different ways of calculating moles CO formed, dependant on molar ratio of CH₄/SiO

Run	CH4-injection		CO from %CO-log		Gas from SiO generator				CH4/SiO molar ratio	Off-gas calc.: If max conversion of reaction (2)							Share of max CO x = a/b	
	rate	time	mean	stable	Wt loss	SiO	CO	CO		Flow rates (mole/h)				b (l/h)				
	l/h	(mole/h)	(min)	(l/h)	(l/h)	(mg/min)	mole/h	mole/h	l/h	CO	H2	Ar	Sum	CO	%CO	%H2		
5	0	0	0	0,70	0,64	75	0,08	0,03	0,63	0,0	0,03	0,00	0,86	0,88	0,63	3,2	0,0	
11	2,4	0,107	60	1,12	1,13	89	0,10	0,03	0,75	1,1	0,09	0,21	0,86	1,16	1,94	7,5	18,4	0,58
6	3,8	0,170	60	1,30	1,03	104	0,12	0,04	0,87	1,5	0,12	0,34	0,86	1,32	2,77	9,4	25,7	0,47
12	6,0	0,268	60	1,70	1,63	111	0,12	0,04	0,93	2,1	0,17	0,54	0,86	1,56	3,72	10,7	34,4	0,46
4	9,1	0,407	63	1,79	1,74	103	0,12	0,04	0,86	3,5	0,15	0,81	0,86	1,83	3,46	8,4	44,6	0,52

There is a peak in the measured % CO in experiments without methane injection, just before the intended temperature is reached in the middle of the SiO generator. However, the CO-peak is distinguished smaller when methane is injected, probably due to a cooling effect of the methane. The off-gas analyses with μ-GC and the CO-analyser compares very well, nearly identical % CO was measured by the two methods. The μ-GC did neither detect any unconverted CH₄ nor other

species formed, apart from hydrogen, for low injection rates. A small amount of unconverted methane (0.2-0.7 %) was detected at start and at the end (0.03 %) for the highest flow rate (9.1 l/h), but only minor amount of side products were detected (0.04 % C₂H₂ or C₂H₄). The two latter species cannot be distinguished by this method.

7. MOLAR RATIO OF CH₄/SiO

The correct molar ratios of CH₄/SiO in these experiments are somewhat uncertain due to the way the SiO gas was formed from the pellets. The reason is temperature gradients due to the water-cooled lance and the endothermic reactions taking place at high CH₄/SiO molar ratios. The SiO formation rate by reaction (5) has been calculated in different ways either based on pellet weight losses in each of these experiments, or compared to different kind of references such as pellet weight losses in SiO-reactivity tests, pellet weight loss in the reference experiment (run 5) or measured CO in the off-gas (run 5). The two latter methods give nearly identical numbers for the molar ratios of CH₄/SiO, since measured CO in the off-gas gives nearly the same CO flow rate as by calculations based on pellet weight loss. Hence the SiO flow rate, being 3 times the CO flow rate, will also be very close.

The preliminary conclusion is that the pellet weight loss in each experiment gives the best estimate for the SiO flow rate and subsequently the best estimate for the molar ratio of CH₄/SiO. The pellet weight loss in experiments with methane injection (89-128 mg/min) is closer to that of the SiO reactivity test (123 – 130 mg/min) than the reference experiments (48-78 mg/min). This is explained by the products that were formed at the bottom of the reaction chamber, probably insulating the top of the SiO-pellets from the water-cooled lance.

An increasing temperature was measured at the top of the SiO-pellets during an experiment (run 12). Such products were not formed in the reference experiments. Hence, the top of the SiO-pellets might be colder in reference experiments, reducing SiO-gas formation.

Figure 10 at left shows the production rates (moles/h) for SiC, SiO₂, C and Si, while at the right is shown the product composition (wt %) for various molar ratios of CH₄/SiO. The generation of SiO gas is based on the SiO-pellets weight loss in each experiment.

The experimentally determined SiC production rate in figure 2 resembles the thermodynamic calculated equilibrium at adiabatic conditions shown in figure 2. The curves and relative amounts of SiC, SiO₂ and C are very similar at high molar ratios of CH₄/SiO.

At low ratios the experimentally determined SiO₂ is higher, probably due to the cooling effect of the lance.

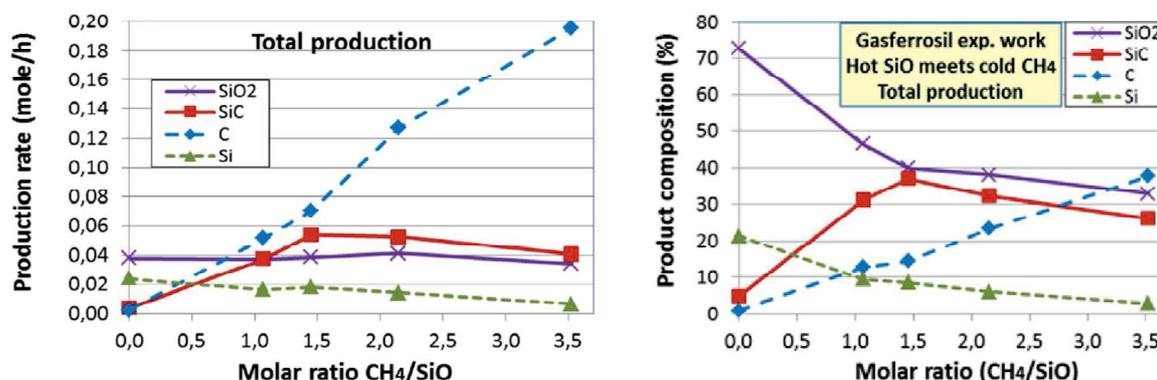


Figure 10: Production rates on a molar basis (at left) and product composition (wt %) at right. Molar ratio with SiO-generation is based on pellet weight loss in each experiment

8. MATERIAL BALANCES

A material balance for C, Si and O is shown in table 3. The input is based on the injected methane flow rate and the SiO and CO gas generated from the SiO-pellets, determined by the pellet weight loss in each experiment. The output is based on the chemical analyses and the amounts of products excavated, together with off-gas analyses of % CO (mean values were used). Argon will be the same for input and output, and 2 moles H₂ will be formed from each mole CH₄, which is not included. There is reasonable agreement between the percentages found in output relative to input. For Run 6 we can see that 88 % of the carbon was found. Lower C and SiC analyses, as determined by the quantitative XRD for run 6, would not help in improving the material balance.

Table 3: Material balances for C, Si and O for the selected experiments

Run	CH ₄ -injection		Sum IN (mole/h)			Sum OUT (mole/h)			% Found		
	(l/h)	mole/h	Si	C	O	Si	C	O	Si	C	O
5 ref.	0	0,00	0,084	0,028	0,112	0,066	0,038	0,107	78 %	134 %	95 %
11	2,4	0,11	0,100	0,140	0,133	0,091	0,139	0,124	91 %	99 %	93 %
6	3,8	0,17	0,117	0,208	0,156	0,110	0,182	0,135	95 %	88 %	87 %
12	6	0,27	0,125	0,309	0,166	0,108	0,255	0,158	86 %	82 %	95 %
4	9,1	0,41	0,116	0,446	0,154	0,081	0,316	0,148	70 %	71 %	96 %

9. CONCLUSIONS

The reaction equilibria between hot SiO-gas (1650°C) and cold CH₄ (25°C) were simulated for various molar ratio of CH₄ to SiO at adiabatic condition. Thermodynamic calculations show that maximum SiC production occurs at CH₄/SiO~1.3 at 1380°C, while the lowest temperature for SiC production by reacting SiO-gas with C is around 1512°C. Consequently, SiC can be produced at a lower temperature by reacting CH₄ with SiO. This is due to the high carbon activity when methane reaches the hot zone which can be of great technical significance for a new SiC or Si process. Excess natural gas will crack to carbon and hydrogen while the temperature drops. In this way natural gas can also act as a cooling agent, while carbon and hydrogen are produced.

The efficiency of silicon carbide formation at these temperatures was verified experimentally for various molar ratios of CH₄/SiO. A water-cooled lance for CH₄ injection was inserted directly into a reaction chamber at 1600-1650°C where hot SiO gas was flowing. In this way it was avoided that methane cracks on its way into the furnace and plugs the equipment. Hence, it was possible to experimentally investigate what happened. SiC was formed to a large extent, confirmed by chemical analyses, and verified by XRD analyses. Maximum SiC was formed close to CH₄/SiO = 1.5. The CO flow rate provides a continuous method for monitoring the process. Experimental and thermodynamic results resemble each other.

10. ACKNOWLEDGEMENT

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