SYSTEM ANALYSIS OF COMPLEX REACTOR BEHAVIOR - A CASE STUDY.

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Abstract: The paper examines the nonlinear behaviour between carbon coverage (input) and silicon production (output) in a submerged arc silicon furnace. A silicon furnace is a highly endothermic chemical reactor. Carbon coverage is the main input for control of the silicon yield and is determined manually. This is a difficult task since the dynamic response varies with the carbon level in the furnace. Internal interconnections between the reactions in the process cause the dominant zeros and poles in the response to move. For high input values and high silicon yield, inverse responses and finally unstability result. The paper describes how the dominant poles and zeros move and identifies the internal interconnections in the reactor that cause the change in dynamics.

Keywords: Reactor dynamic behaviour, zero and pole drift. Submerged arc silicon furnace.

1. INTRODUCTION

In this paper we take a reactor perspective on the silicon furnace, and try to explain and untangle its highly complex and changing dynamic behavior. The focus is the dynamic response between carbon coverage and the tap-rate of silicon. Many interconnected phenomena inside the silicon furnace make the behavior change dramatically from low to high carbon coverage values. By applying the views often used to analyze the dynamic behavior of integrated process systems such as distillation columns and/or chemical reactors, this paper contributes with new insight into the dynamic behavior of the silicon furnace.

The production of silicon metal is a large industry world wide. Elkem has approximately 20% of the market share for silicon metal world wide with net sales of 2.2 BNOK (210 BTons) in 2002, (*Elkem Annual Report 2002*, 2002). Elkem has many plants in Norway, Iceland, Canada and the United States, each containing several furnaces. Silicon metal is used for the production of silicones in the chemical industry, and as an important additive to other metals in the casting industry. Silicon metal is also further refined for production of solar cells and electronics. A valuable by-product of silicon metal production is microsilica which is used as an additive to concrete, among other things.

All data in this paper are obtained using "Simod" a dynamic simulator for submerged arc silicon furnaces developed by Elkem, NTNU and Sintef. Simod is especially detailed with respect reaction kinetics and the thermodynamics of the furnace. The model is a nonlinear differential algebraic equation system, see (Foss and Wasbø, 2001). Simod was originally developed for teaching and

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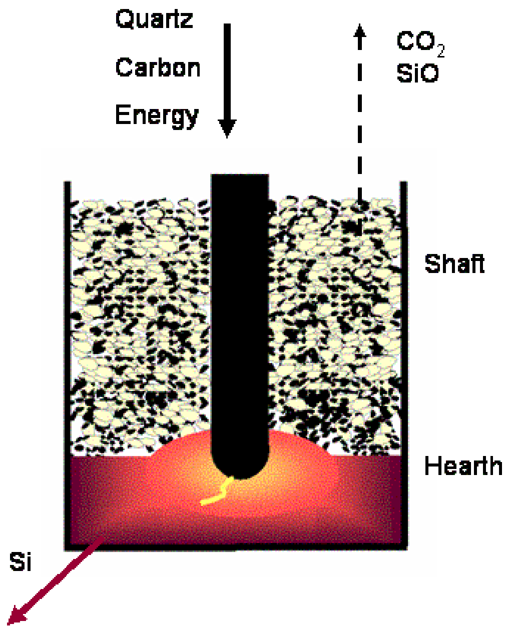

Fig. 1. Silicon furnace.

training purposes of plant operation personnel in Elkem. It has also provided a valuable means to collect and consolidate Elkem's knowledge about the silicon furnace behavior. Simod has been used over several years, and its behavior has been verified by metallurgists and other process personnel.

The contents of the paper are as follows. An introduction to the fundamentals of the silicon production and chemistry is given in section 2. Section 3 describes and quantifies the properties of the steady state and dynamic response between carbon coverage and silicon tap-rate. The changing dynamic behavior of the process has been shown using small perturbations around a steady state for different carbon coverage levels. This can be considered a linear approximation to the nonlinear behavior. The time constants and gain of the linear model approximation have been estimated in order to quantify the changing dynamic behavior of the process. An analysis of the results is given, in section 4. A discussion and conclusion is given in section 5.

2. SILICON PRODUCTION

A silicon furnace is basically a large pot (3-4m high, 3-5m in diameter) in which carbon materials and quartz (SiO_2) are fed at the top and silicon metal (Si) is tapped at the bottom, see figure 1. Some of the SiO gas produced in the furnace escapes over the furnace top, oxidizes and forms microsilica which are captured in the off-gas filter.

The silicon furnace is highly endothermic. The main energy source to the process is electricity supplied through carbon electrodes. Depending on the size of the furnace, the electric power supply typically lies between 10 and 25MW. Most of this energy is released through an electric arc in the in the lower part of the furnace ("hearth") where silicon is formed and tapped (Valderhaug, 1992). The upper, cooler part of the furnace is called the "shaft".

The measurements of the mass transport of carbon, quartz into the furnace and amount of tapped silicon out of the furnace are registered as aggregated values on an hourly, shift (8 Hours) or day (24 Hours) basis. Microsilica production is in some plants measured for individual furnaces.

2.1 Silicon furnace chemistry

In essence, silicon production implies removing the oxygen atoms from the quartz molecule, SiO_2. The overall, gross reaction for the whole furnace can be written:

$$SiO_2+(1+x)C=xSi+(1\text{-}x)SiO+(1+x)CO \quad (1)$$

where x is the ratio of silicon metal recovered from the silicon in the quartz feed.

In reality a number of reactions take place in order for Si to be formed. The "engine" of the reactions in the furnace is silicon oxide gas, SiO. This gas is formed in the hearth in a highly endothermic reaction between silicon metal and melted SiO_2:

$$Si+SiO_2=2SiO \quad (2)$$

Some of the SiO is consumed within the hearth to form silicon metal according to the reaction:

$$SiC+SiO=2Si+CO \quad (3)$$

This reaction requires a sufficiently high partial pressure of SiO as well as a temperature of approximately 2000^{o}C, (Schei *et al.*, 1998) which is obtained by the electric arc. Most of the silicon carbide required in reaction (3) is formed in the shaft in a reaction between SiO rising up from the hearth and the carbon feed in the shaft:

$$SiO+2C=SiC+CO \quad (4)$$

SiC is a solid, and travels down to the hearth. Reaction (4) may also take place in the hearth if unreacted carbon reaches the hearth.

Some of the SiO that rises up through the shaft condenses, and travels down with the other solids to the hearth or lower shaft as Si/SiO_2 condensate. The SiO condensation is a significant energy supply to materials in the shaft.

The component flows between the reactions and the hearth and shaft are shown in figure 2.

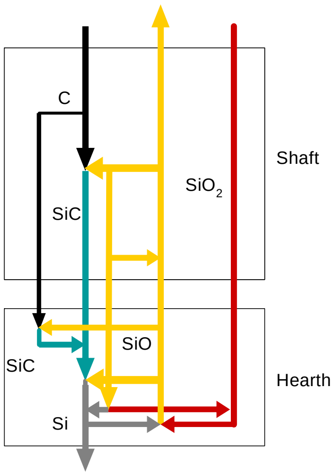

Fig. 2. Component flows in the silicon furnace shaft (upper) and hearth (lower).

3. CARBON COVERAGE TO SILICON TAP-RATE RESPONSE

This section gives a description of the dynamic behavior of the silicon furnace with an emphasis of the response with respect to changes in carbon coverage to the silicon tap-rate. The reasons for focusing on this particular input output pair is that carbon coverage is the main variable used for controlling the silicon yield. Also, silicon is the main product of the process is one of the key quantities for process operation personnel to control the process and evaluate its performance. We first will take a look at the steady state gain and dynamic responses at various carbon coverage levels. Next the changes in the dynamics are quantified.

3.1 Dynamic behavior and steady state gain

The steady state gain for carbon coverage to silicon tap-rate is plotted in figure 3. Carbon coverage is defined as a percentage value of the stoichiometric ratio between the silicon in the quartz feed and the carbon in the carbon materials. A carbon coverage of 100% would be consistent with 100% silicon recovery. Due to some loss of silicon over the furnace top, the average carbon coverage value should lie below 100%. In the curve below, the carbon coverage has been varied from 93% to 99%. These values include a 3% loss of carbon at the furnace top, and 3% should therefore be subtracted to get the net carbon coverage value entering the reactions of the furnace. The silicon tap-rate is the amount of silicon (measured in kg) tapped during one hour.

The simulations have been made with a net 22MW power supply and a reactivity parameter for the carbon of 0.56. A feed controller in the simulator ensures a constant material level of the furnace,

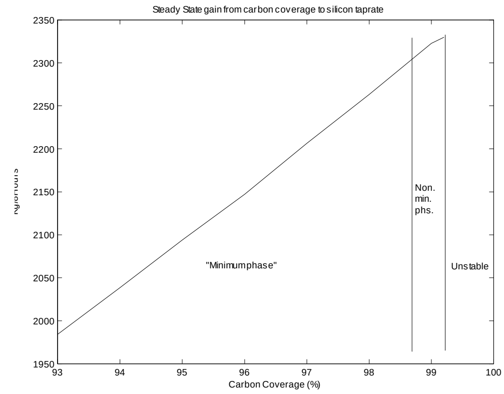

Fig. 3. Steady state gain between carbon coverage and taprate silicon.

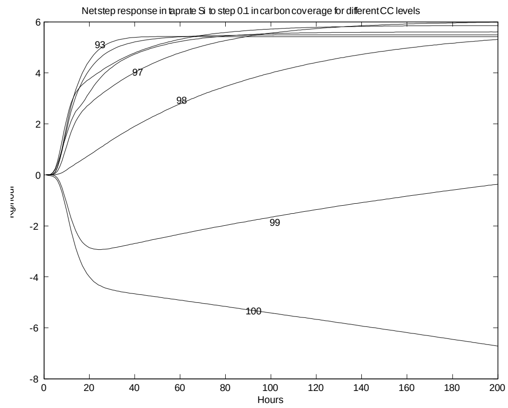

Fig. 4. Net step responses in taprate Si to a 0.1 step in carbon coverage (CC) at different carbon coverage values.

and a tapping controller maintains a constant Si level in the hearth.

We see that the steady state gain is relatively constant all up to 99%. Above 99.5% there is a buildup of silicon carbide in the hearth. This creates an integrating effect, and no steady state gain can therefore be defined for such high carbon coverage values.

The following method has been chosen to see how the dynamic response evolves for different carbon coverage values. For the carbon coverage values (including a 3% loss): 93, 94, 95, 96, 97, 98, 99,100 we have run the simulator to reach a steady state value (not obtainable at 100%). At each of these steady state situations, we have made a small positive perturbation (step) by adding 0.1 to the carbon coverage value at time 1 Hours and registered the silicon tap-rate response. In order to compare the dynamic response more easily, we have for each carbon coverage level, subtracted the steady state silicon tap-rate value so that each response starts from zero. As we can see, the responses are much faster for small carbon coverage values than for the higher values. The dominant time constant at a carbon coverage value of 93% (net 90%) lies around 8 Hours

Carbon coverage	K	T1	T2	T3	T4
93	54,4	*	*	4,9	4,9
94	55,5	31,2	34,6	2,7	7,8
95	56,2	28,5	34,2	0,5	14,1
96	59,3	17,1	33,3	2,7	2,7
97	61,7	16,9	46,7	3,7	3,7
98	58,2	5,1	78,4	5,0	5,0
99	56,0	-306,7	688,8	3,1	3,1

Fig. 5. Identified poles and zeros (in Hours).

whereas for 98% it is approximately 100 Hours. We also can observe some nonlinear effects in the responses corresponding to 92% and 93%. At 99% carbon coverage we observe an inverse response in the tap-rate, thus we have indicated in figure 3 that this is a non-minimum region. Since we have included only 200 Hours, the figure does not reveal the fact that this response has approximately the same steady state gain as the other curves. For 100% (net 97%) we see that there is a net negative integrating effect, and no steady state gain can be defined for this input value. The negative gain makes the process is unstable in this region.

3.2 Quantification of the change in dynamic behavior

The following approximation to the linearized behavior of the process has been used

$$H(s) = \frac{K_1 e^{-\tau s}(1 + T_1 s)}{(1 + T_2 s)(1 + T_3 s)(1 + T_4 s)} \qquad (5)$$

Even though we have said the process is nearly minimum-phase for small carbon coverage values, we still have included a small transport delay to capture some of the propagation time for the materials down through the furnace. This delay is estimated to approximately 2.5 Hours for all responses, and is not included in the table of figure 5. The parameters have been estimated using a least squares method ("lsqcurvefit" in Matlab). All the time constants are in hours. As we can see from the table, the gain is relatively constant. The two right-most columns (T_3 and T_4) are the fastest poles of the process. The process seems to have a double pole which in the range 2.5-5.0 Hours. T_1 represents a zero and T_2 a pole. For 93% the response is better identified with an approximation model with only two poles. For higher carbon coverage values the time constant of the pole (T_2) increases. The time constant of the the zero (T_1) decreases, and finally goes negative. For a carbon coverage value of 99%, the zero is faster than the pole, and since the time constant is negative, the result is an inverse response. The negative integrating effect observed for carbon coverage equal to 100% can be interpreted as the time constant for the pole T_2 having become infinitely large. The development of the dominant time constant in the pole (T_2) and zero (T_1) have been plotted against the carbon coverage level in figure 6.

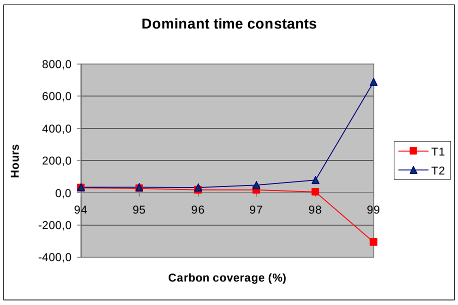

Fig. 6. Development of the dominant time constants.

4. ANALYSIS OF THE CHANGE IN DYNAMIC BEHAVIOR

The changing dynamic behaviour of the carbon coverage to silicon tap-rate response seen in figure 4 and identified in figure 5 has a dramatic effect on process operation, especially since carbon coverage is determined manually. At low silicon yield it is possible to have a "fast" process, and the effects can be observed within a shift. With high carbon coverage the response is extremely slow and difficult to relate to in manual operation. Our aim in this section is to explain the development of the dominant zero and pole by looking at the internal interconnections and component flows in the process.

4.1 Process interconnections and plant dynamics

The basis for a system theoretic analysis, is the literature dealing with the changing dynamics of reactors due to interconnections in process plants, see for instance (Morud and Skogestad, 1996). The paper classifies different process interconnections and gives an overview over the effect each type has on the plant dynamics. On the overall level they distinguish between external and internal interconnections. By external interconnections are meant interconnections between subsystems associated with different processing equipment. Internal interconnections mean interconnections between phenomena within one process vessel. Within the internal interconnection "class" they subdivide into recycle, parallel paths and series interconnections. Series interconnection is the simplest kind of interconnection and implies that there is a one-way flow between subsystems of material and/or energy. In a system with recycle, mass and/or energy flow is fed back in the process. Recycle generally moves the poles of the process. Which way, generally depends on whether the feedback is positive or negative. In most systems, recycle leads to positive feedback, according to (Morud and Skogestad, 1996). The third form of internal interconnections is parallel paths, or process "feedforward". The existence of parallel paths creates or moves the zeros of the process. If the effects of the parallel paths have opposite signs, they are competing effects which may give unstable zero dynamics, or inverse responses for linear systems. An example of this can be found in (Kuhlmann and Bogle, 1997) who describes a van de Vusse process in which there are competing effects causing the zeros of the process to move. In the van de Vusse process the unstable zero dynamics also cause a sign change in the steady state gain (input multiplicity).

To apply the ideas presented in (Morud and Skogestad, 1996) on the silicon furnace, we take a look back at figure 2 and try to identify the types of internal interconnections that we might have in the component flows in the silicon furnace from carbon feed to tapped silicon in order to explain the changing dynamics. There are many types of interconnections in this process, and the different types cannot be easily distinguished.

4.2 Steady state conversion levels of the reactions

In order to determine which are the dominant phenomena within the silicon furnace, we need to quantify the steady state conversion rates in the different reactions at different carbon coverage levels. In figure 7 we have plotted the steady state conversion rate of the Si producing reaction (4) and the SiO producing reaction (2). The difference in generated kmol/h of SiO is approximately 4 (twice the increase in the reaction) from the highest and to the lowest carbon coverage level, whereas the difference in consumed SiO in the Si producing reaction is almost 10 kmol/h. Since

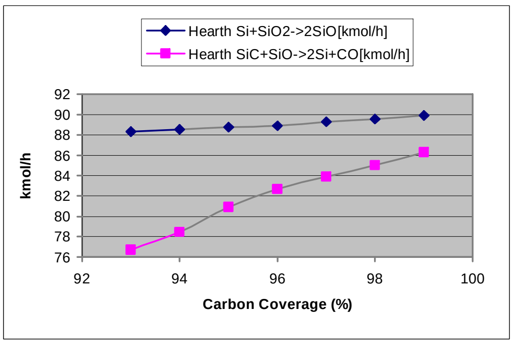

Fig. 7. Stationary conversion rates for SiO and Si producing reactions in hearth.

the SiO consumption in the hearth increases more than the production, the SiO level in the shaft will be lower for higher carbon coverage levels.

Next we look at the SiC generating reaction (4) which also consumes SiO. In figure 8 we have plotted the conversion rates for this reaction in the shaft (upper) and hearth (lower). For low carbon coverage rates, most of the SiC is formed in the shaft and little in the hearth. For higher carbon coverage rates less SiC is formed in the shaft and more in the hearth. The explanation lies the increased SiO consumption in the hearth due to increased production of Si, as seen in figure 7. In addition, the increased production of SiC in the hearth reduces the SiO in the shaft further. The last reaction we need to quantify is

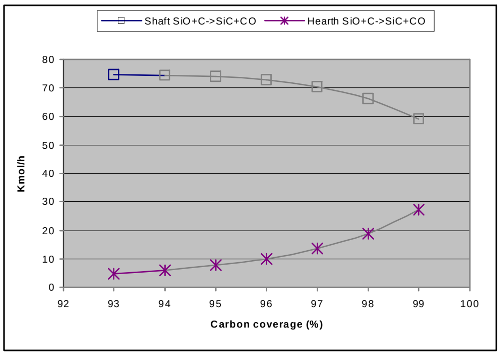

Fig. 8. SiC generation in the shaft (upper) and hearth (lower).

the recycling through condensation of SiO. The simulations show that most of this recycling takes place within the shaft itself, and that very little condensate reaches the hearth (0.01-0.02 kmol/h). This is a very small rate compared to the rates of the reactions in the figures above, and the "material recycle" of Si through SiO/SiC therefore dominates. We therefore ignore the effect of recycle through condensation of SiO in the shaft in the following analysis.

4.3 Analysis of the dominant mechanisms

By ignoring the recycling of SiO through condensation in the shaft, we can now redraw figure 2 emphasizing the response from carbon to tapped silicon. The boxes in figure 9 indicate reactions.

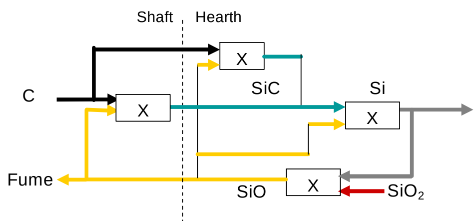

Fig. 9. Mass flow between reactions in the hearth and shaft.

The Si level in the hearth is kept constant by a level controller, and any surplus of Si is tapped. Si is also recycled through as SiO in the lower right box. SiO is consumed by the SiC generating and Si generating reactions in the hearth. What is not consumed in the hearth rises up through the shaft and is consumed by the SiC generating reaction there. The rest of the SiO gas escapes as fume over the furnace top.

One reaspm for the slower response for high carbon coverage values is that the carbon in the hearth needs time to react to SiC, whereas for low carbon coverage values, the SiC is formed in the shaft and comes down to the hearth ”ready” to enter the Si producing reaction. Another reason for the slower response at high carbon coverage rates is the decreased amount of SiO gas in the furnace, see the steady state profiles for SiO in figure 10. Elements 1 to 10 on the x-axis correspond to the top to bottom elements of the shaft, element 11 corresponds to the hearth. Since the partial pressure of SiO (above an equilibrium pressure) drives the SiC and Si generating reactions, less free SiO in the furnace will clearly give a slower response in all reactions. When looking for a source

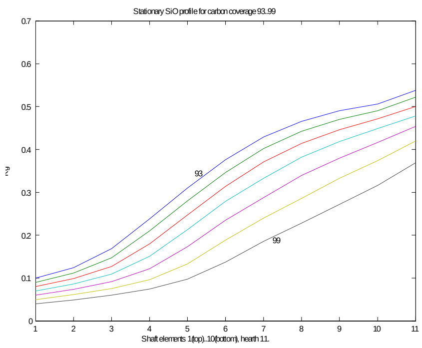

Fig. 10. The steady state SiO profile for carbon coverage values from 93% to 99%.

of the moving zero, (Morud and Skogestad, 1996) indicate that we should look parallel and possibly competing phenomena. The two reactions in the hearth producing SiC and Si are obvious candidates since they are exposed to the same partial pressure of SiO. SiC production in the hearth is insignificant for low carbon coverage values according to figure 8, but amounts to almost half of the SiC production for 99% carbon coverage. In the low carbon situation there is a relatively large surplus of SiO in all parts of the furnace. For a high carbon coverage level, the general level of SiO in the furnace goes down, more unreacted carbon will enter the hearth, and the SiC generating reaction in the hearth will take a more and more significant part of the SiO in the hearth.

5. DISCUSSION AND CONCLUSION

This paper has shown that speed of the response in silicon tap-rate to a change in carbon coverage is an indication of distance from maximal silicon production. A slow response indicates that the furnace operates very close to maximal production, whereas a fast response indicates that it is possible to increase the carbon coverage level. The carbon coverage level is in most furnaces determined manually. This result can be applied in the current manual determination of the carbon coverage level to the furnace.

There are however other factors and inputs influencing the silicon furnace behaviour which have not been treated in this paper. Firstly, the reactivity of the carbon material is an obvious candidate for further studies since it enters linearly in the SiC generating reaction. A higher reactivity would cause more of the carbon to react in the shaft and less in the hearth.

Another effect not included in the simulations of this paper is the possibility for formation of a particulary hard and non reactive form of SiC. This effect may occur when there is a sufficiently high amount of carbon in the hearth (at maximal production), and may cause build up and severe tapping problems in the furnace. Inclusion of this effect would lower the maximal production somewhat, in the range of 1% carbon coverage.

Other inputs not considered in the paper are the electrical energy supply level, and vertical energy distribution in the furnace. In (Lund *et al.*, 2003) it was shown that the energy supplied to the furnace affects the production level of silicon. Investigation of the effect of the reactivity and the power level on the dynamic behavior of the furnace should therefore be considered for future work and before an automatic controller structure is suggested.

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