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DYNAMICAL MODEL FOR THE HIGH-TEMPERATURE PART OF THE CARBOTHERMIC SILICON METAL PROCESS.

Birger Andresen and Johan Kr Tuset.
Division of Metallurgy, The Norwegian Institute of Technology, The University of Trondheim, Trondheim, Norway.

ABSTRACT

A 2-dimensional, cylindersymmetric dynamical model for the high-temperature part of the carbothermic silicon metal process has been developed. One out of three electrodes is modelled. The model combines submodels for DC electric arcs, fluid flow, heterogeneous chemical reactions taking place at temperatures above $1850°C$ and other prominent phenomena in the process. Simplifications are made to reduce the overall complexity of the model, such as assuming inert furnace walls and an inert electrode, neglecting electric currents and any effects thereof outside the electric arc and assuming that the metal pool is a rigid body rather than a fluid undergoing continuous movement. The results from the model, such as gas and temperature distributions, furnace and particle geometries, fluid flow fields etc., are presented graphically in an informative way. The model may prove to be an efficient research tool, improve the general understanding of the high-temperature part of the process and finally serve as the basis for a complete model of the process.

INTRODUCTION

The carbothermic process for making silicon or ferrosilicon carried out in submerged arc furnaces is a complex process where a number of interconnected subprocesses take place simultaneously and at very high temperatures as well. Temperatures around 2300 K have been recorded at the walls of the gas filled cavity surrounding the lower end of the electrodes [1], but elevate to much higher levels in the free burning electric arc, extending from the lower tip of the electrode to the metal pool or the cavity walls. Knowledge of the kinetics and the thermodynamics of the reactions involved at these elevated temperatures are limited. The same is true also for important data on properties affecting the exchange of mass and heat. The metallurgy of the process is thus only partly understood.

The prevailing temperatures and the practical difficulties involved in mounting sensors and devices deep inside the furnace make the process unsuited for most direct measurements. Important parameters like local temperatures and compositions that could be used to

characterize the state of the process are therefore unavailable, and must be estimated from measurements of other observable variables or from educated guessing.

Large time delays caused by slow transport of materials in the furnace and sometimes by slow responses in measured variables to other changes in the process also add to the difficulties involved in monitoring the state of the process. It takes hours rather than minutes from a change is done in the raw material mixture on top of the furnace until the effect of it is observed as a change in the state of the furnace close to the high temperature, metal producing zone around and underneath the electrodes.

The highly complicated metallurgy of the process combined with the severe problems in measuring the state of the furnace and the sometimes large time delays involved accentuate the need for simulation programs which may help the metallurgist in chosing the correct strategy for the furnace operation. Such simulation programs are quite different from the computer control systems widely used in the ferrosilicon and silicon metal industry at present. These control systems do not model the actual process, but are instead helping the operator to monitor the process and equipment by collecting and processing historical data and then presenting them in a quick and suitable way. They also regulate input variables automatically to keep the furnace state close to that which is believed to be the most favourable. Such systems have been used for several decades and have paid off mainly by making it easier to keep the furnace operation steady and running, and by increasing the general understanding of the process as well.

Even with the improvements achieved in recent decades, there are still room for significant improvements. Simulation programs modelling some of the most prominent physical and chemical phenomena of the process may contribute to this even though such programs are not likely to be used directly in furnace control in the near future. Their main contribution will probably be to improve the overall understanding of the process and to provide a tool for checking how the process, or parts of it, is likely to respond to certain changes in the control parameters. Better furnace operation may be obtained from this.

The only successfully implemented dynamical simulation program for the silicon metal process known to the authors is the unidimensional dynamic model developed and used by Elkem a/s [2]. This model describes a vertical shaft extending from an unspecified cavity up to the top where fresh raw materials are charged. Partial differential equations are formulated for mass and energy transport and chemical conversion due to chemical reactions and phase transformations are modelled. The model is currently being used by the Elkem company, and has proved useful for educating operators and for research purposes.

THE HIGH-TEMPERATURE MODEL

General description.

Excellent presentations of the metallurgy of the ferrosilicon and silicon metal process are given by Schei [1] and by Schei and Larsen [3]. Their descriptions of the high-temperature zone of the process form the basis of the present model.

The model is dynamical and concentrates on the electric arc and its surroundings, i.e. the gas filled cavity, the metal pool and the lower parts of the materials above the cavity.

Chemical reactions taking place in these areas at temperatures higher than approximately $1850°C$ are included. Sub models for the electric arc, fluid flow, energy transport and heterogeneous chemical reactions are combined in a way that has not earlier been done for the silicon metal process. Energy transfer by radiation from the electric arc to the surface of the metal pool and to the cavity walls is included, whereas the radiation between surfaces of different temperatures is neglected. The colder parts of the process are also modelled, but chemical reactions taking place below $1850°C$ are omitted at this stage. The topochemical conversion of C to SiC and the strongly exothermic condensation of SiO-rich gas in the upper parts of the furnace are the most important chemical reactions that are omitted.

The furnace is modelled as a two-dimensional, cylinder-symmetric system containing one single vertical electrode with its center at the axis of symmetry as indicated in Figure 1. The electrode and the furnace walls are assumed to be inert. Direct current is assumed in the electric arc. The period between two successive stokings is simulated.

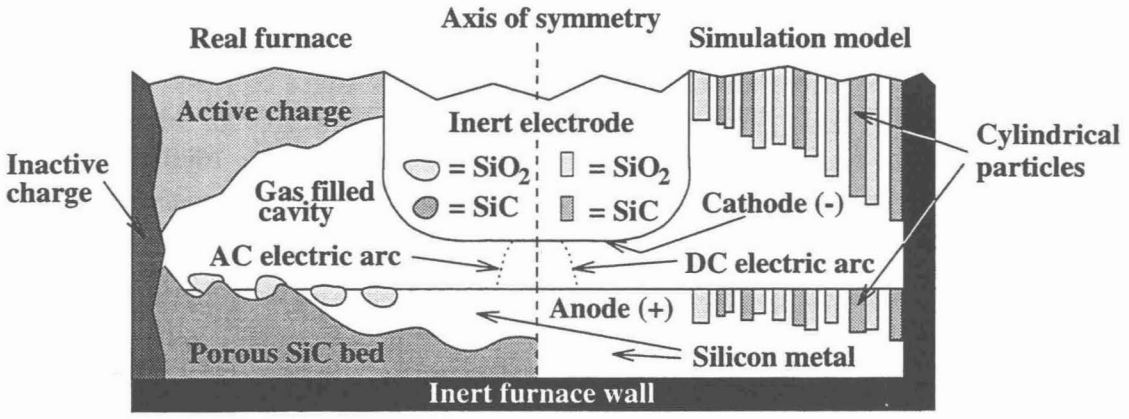

Figure 1: Outline of the lower part of the furnace. Real furnace to the left and the same situation as represented in the simulation model to the right.

The main features of the model are described below, whereas the details are given in [4].

Phases, reactions and kinetics.

The raw materials used in the silicon metal process are quartsite (SiO_2) and carbon. Both are usually charged as lumpy materials. They contain a certain percentage of impurites which are all neglected in the simulation model. Si, C and O are consequently the only three chemical elements present in the system. The most important chemical reactions taking place between these elements at the prevailing conditions inside the furnace are believed to be :

$$SiO_2 + C \overset{R_1}{=} SiO + CO \tag{1}$$

$$2SiO_2 + SiC \overset{R_2}{=} 3SiO + CO \tag{2}$$

$$SiO_2 + Si \overset{R_3}{=} 2SiO \tag{3}$$

$$SiO + SiC \overset{R_4}{=} 2Si + CO \tag{4}$$

$$SiO + 2C \overset{R_5}{=} SiC + CO \tag{5}$$

The carbon in the raw materials is assumed to be completely converted to SiC while reacting with SiO-gas at temperatures below $1850°C$ in the upper parts of the furnace. The fraction of the carbon in the raw materials that do actually enter the hot zone of the furnace (about 50%) is thus replaced by a corresponding amount of SiC in the model. The rest of the carbon leaves the furnace as CO-gas mainly produced by reaction (5). Free carbon is consequently not present in the model as a chemical active compound because the electrode is assumed to be inert.

Liquid SiO_2 is highly viscous, and the silica is assumed to preserve its original shape also when appearing in its molten state in the hot zone of the furnace. Molten SiO_2 is thus treated formally just like solid SiO_2 in the model.

The condensate, which is known to form in the upper parts of the furnace when SiO-rich gas cools, consists mainly of SiO_2 and Si produced by reaction (3) proceeding to the left (Schei and Sandberg [5]). Some C and SiC may also form by the reversed reactions (1) and (2), respectively. The Si in the condensate is expected to form droplets that leave the condensate and drips down to the metal pool at temperatures below $1850°C$. The extremely small particles of C and SiC in the condensate are in intimate contact with the surrounding SiO_2. As there are no kinetic hindrances for these to react with the matrix of SiO_2, reactions (1) and (2) proceed to the right at temperatures for which the reaction pressure exceeds the ambient pressure. The carbon reacts at $T \geq 1680°C$ and the SiC at $T \geq 1813°C$ for a total pressure of 1 atm. The condensate is therefore not likely to contain C and SiC above $1850°C$, and is treated as pure SiO_2 in the model.

In accordance with these assumptions, SiO_2 and SiC are the only materials present in the shaft of the model except for gas. They are modelled as individual concentric rings of SiO_2 and SiC around the symmetry axis. These particles appear as vertical bars in Figure 1. They may or may not be seperated by gas channels or by other materials. In the current version of the model, they may react with gas only; SiC with $SiO(g)$ (reaction (4)) and SiO_2 with $Si(g)$ (reaction (3)).

SiO_2, SiC and Si are the only materials that are present in the metal pool. Here, the SiO_2-particles are only allowed to react with silicon from the metal pool or with $Si(g)$ that has evaporated from the metal pool, both according to reaction (3). The SiC-particles can only react with $SiO(g)$ (reaction (4)). This means that reactions (3) and (4) are the only reactions that are assumed to proceed in the model, both in the forward direction.

Silicon evaporates from the upper surface of the metal pool. The evaporation rate is, below the boiling point of silicon, controlled by diffusion of $Si(g)$ away from the surface through a turbulent boundary layer :

$$\dot{J}_{Si} = k_{Si} \cdot (p_{Si,eq} - p_{Si,bulk}) \;\; ; \;\; \dot{J}_{Si} \geq 0.0 \tag{6}$$

where $p_{Si,eq}$ is the equilibrium pressure of silicon vapour at the surface, $p_{Si,bulk}$ is the bulk pressure and k_{Si} is a function of the turbulent boundary layer thickness and the Schmidt number as proposed by Johansen [7]. The rate controlling mechanism is gradually changed to the equation developed by Langmuir (Gu [8, page 94]) above the boiling point.

In the real furnace, the evaporated silicon will partly react with other gases and partly condense in the gas phase or at the surfaces of solids or liquids, giving off energy and mass. A fairly simple approach is used in the model to account roughly for the real energy transport and associated effects caused by the evaporation and condensation of silicon.

Energy is removed from the place of evaporation and redistributed partly to the gas and partly to the crater wall and the metal pool. Si in the metal pool and SiC surfaces will in the model be heated, but less so as their local temperatures approach the boiling point of silicon. SiO_2 surfaces may, on the other hand, experience either a net heating or a net cooling depending on its local temperature. This is a result of two competing phenomena; heating caused by the condensation of $Si(g)$ and cooling caused by the strongly endotermic reaction (3) between condensing $Si(g)$ and SiO_2. There will be a net heating if the local temperature of the SiO_2 surface is below that which gives $p_{SiO} = 1.0$ atm for reaction (3), and cooling otherwise. The magnitude of the cooling/heating increases with larger deviations from this reference temperature of $T = 1859°C$. The practical calculations related to the evaporation/condensation do not involve any physical transfer of Si to or from the gas phase. The details explaining how this is done are presented in [4].

Reaction (3) also takes place at the Si/SiO_2 interfaces in the metal pool when the reaction pressure exceeds the ambient pressure ($p_{reac} \geq 1$ atm when $T \geq 1859°C$), in which case there is assumed to be no kinetical hindrances involved. The rate is in this case controlled by the net energy transfer by heat conduction through the reacting materials to the reacting surface which is assumed to remain at $T = 1859°C$.

The SiO produced by these reactions at different locations on the surface of a given SiO_2-particle is distributed evenly (same kg/m^3) to the gas phase just above the particle.

Reaction (4) takes place at surfaces of SiC-particles exposed to furnace gas. The reaction can thus proceed both in the metal pool and in the furnace shaft. The reaction rate is controlled by mass transport of $SiO(g)$ to the reacting surface through a turbulent boundary layer as described above for the evaporation of silicon.

The effective surface area of particles participating in chemical reactions can be adjusted by operator defined correction factors to allow for deviations between the specific surface areas for the chosen model geometry and the situation in a real furnace.

The furnace shaft.

The bars representing the SiO_2 and the SiC-particles in the furnace shaft do not move in the model. This corresponds fairly well with observations on production furnaces, and may partly be explained by the formation of condensate which glue the different particles together to a sticky mass. However, some material is in the model transferred from the lower edges of the particles in the shaft to the upper edges of corresponding particles of the same type straight below in the metal pool. This simulates material dripping down from the cavity wall due to the increased fluidity of SiO_2 as the temperature increases. The amount of mass that is transferred from each SiO_2-bar is calculated from the temperature distribution of the bar. The total amount of SiO_2 dripping down is used to calculate the corresponding amount of SiC which is transferred to the metal pool together with the SiO_2. The mass ratio of SiO_2 to SiC in the material that drips down is chosen so that the same ratio in the shaft remains as unchanged as possible. The ratio in the shaft may, however, change slightly with time due to chemical reactions taking place in the shaft.

The total mass that drips down from a given particle (bar) is removed from the lower edge of the particle as an equally thick slice over its entire cross section. The same is true for material consumed in chemical reactions, regardless of which parts of the particle that actually do react. All particle surfaces thus remain smooth at all times. The slices that are

removed from the SiO_2-particles due to dripping are generally of different thickness, while the slices removed from the SiC-particles due to dripping are all of the same thickness. The overall result of chemical reactions and dripping is that the lower edges of the particles move upwards with time. The cavity thus increases in volume during the simulation.

Electric conduction and any effects of currents passing through the materials in the charge are neglected in the model.

The metal pool.

The density of SiO_2 at relevant temperatures is somewhat lower than that of Si, which in turn is lower than that of SiC. SiO_2 therefore floats fairly deep at the top of a possible metal pool in a furnace producing silicon metal, whereas the SiC sinks to the bottom of the pool. Both will float in the metal pool, however, when smelting ferrosilicon. Observations on furnaces producing silicon metal indicate that SiC forms a sort of highly irregular bed with cracks and holes at the bottom of the furnace. The silicon metal flows down into these cracks and holes together with molten SiO_2. Both SiO_2 and SiC in the metal pool are thus partly exposed to the gas in the cavity and partly covered by silicon metal.

This rather complicated situation is simplified in the model by assuming a rigid metal pool of pure silicon in which particles of SiO_2 and SiC float with their upper surfaces alligned with the top of the metal pool. Each particle is partly exposed to the furnace gas above the metal pool and partly in contact with the silicon metal in the pool. The gas flow above the metal pool then becomes fairly simple as the surface of the metal pool is smooth. See also Figure 1.

The upper and lower surfaces of the metal pool are both kept at constant levels throughout the entire period of simulation. The amount of silicon in the metal pool changes, however, with the amount of SiO_2 and SiC present at the given time. To adjust for this, and to keep the upper edge of the metal pool at the prescribed constant level, silicon is transferred to or from an external silicon reservoir (outside the furnace). This silicon reservoir can be looked upon as a perfectly insulated ladle with no temperature gradients. The reservoir changes its temperature when new silicon of a different temperature is added from the furnace.

The particles in the metal pool change their geometry as a result of chemical reactions and addition of materials dripping down from above. Simulating the removal of mass both in the vertical direction and along the sides of the particles require rather complicated and time-consuming computer algorithms without prospects of gaining much in terms of a more realistic model. A simplification is therefore used where the particles are allowed to change their sizes in the vertical direction only. The total mass that drips down to a given particle from above is added on top of the particle as an equally thick slice of material over its entire cross section. Mass consumed in chemical reactions is likewise removed as an equally thick layer from the upper surface of SiC-particles (since they react at the top with the gas in the cavity) and from the lower surface of SiO_2-particles (since they react with the silicon in the metal pool at its lower edge and its sides). The particle is then moved vertically to allign its new upper surface with the top of the metal pool. Details on how this is handled in the model and how the metal is redistributed in the metal pool are given in [4].

Electric conduction and any effects of currents passing through the materials in the metal pool are neglected in the model. Any fluid flow inside the metal pool or possible deformations of the metal pool caused by the electric arc or by other phenomena and any associated effects are also neglected.

The electric arc.

The electric arc is modelled according to the equations given for DC arcs by Ramakrishnan et al. [6]. This model assumes a parabolic current distribution over the cross section of the arc, and a known expansion of the arc as a function of the distance from the electrode spot. The diameter of the electrode spot is a parameter in the model as well as the total current of the arc. The model is believed to be well suited for electric arcs of the size and power levels relevant for furnaces producing silicon metal. The volumetric radiation density of the plasma is, in abcense of data for Si-C-O mixtures, calculated for pure argon or Ar-Si mixtures and selected effective arc radii in the range 1cm – 15cm (Gu [8]). The transport properties for the gas (Gu [9]) correspond to a mixture of 60 mole precent SiO and 40 mole percent CO. Data for other Si-C-O mixtures can easilly be added. Both the radiation data and the transport properties are calculated in the range T=[1000K, 30000K]. Radiation from the electric arc to the solid/liquid surfaces in the furnace is included. The power level of the electric arc can be changed at any time during the simulation. Heat transfer from the arc to the metal pool is included.

The fact that the AC arc of the real furnace is substituted by a DC arc in the model may be of basic importance, especially as the flow fields are fundamentally different in the two cases.

IMPLEMENTATION.

The model is implemented in FORTRAN-77 code, using version 2.97 of the fluid flow simulation program FLUENT [10] as a basis. FLUENT is widely used for fluid flow simulation in various parts of the world. The basic version of FLUENT contains a number of relatively sofisticated and well tested standard models for laminar and turbulent flow and phenomena like heat conduction and heat transfer from gases to condensed materials etc. FLUENT basically follows the ideas presented by Patankar [11]. Turbulent flow according to the K-ϵ model [10] is assumed in the simulations.

FLUENT was developed for fluid flow simulations at low and moderate temperatures. Applying the program to high-temperature metallurgical processes requires major modifications. Such modifications have been made by research personell at SINTEF/NTH over the last decade. Further modifications were neccesary while developing the present model. The major contributions are the introduction of heterogeneous chemical reactions and associated changes of particle sizes, dripping of materials from one particle to another due to melting and modifications of models for the electric arc and radiation from it.

SIMULATIONS.

Selected results from an arbitrary test case are presented in figures 2-4 in order to indicate how the results from the simulations may be visualized and to show some general features

of the model. See [4] for further results and discussions. The colour plots actually obtained are much more informative than the black and white plots presented here.

The results are for a 3.0 meters high, 7kA furnace with a radius of 1.2 meters corresponding to the crater dimensions of a real furnace with an electrode radius of 0.63 meters and an assumed arc length of 10cm. The computational domain has 200 and 100 grid points in the vertical and radial directions respectively. The calculated power level is 8.7MW, the radiation loss from the arc is about 2.1MW and the average net production of silicon is about 3.75 kg/minute. This is lower than expected in a real furnace with the same power level, and may partly be explained by the absence of the condensation reactions in the upper parts of the shaft, but are also due to simplifications in the model and possibly ill-tuned model parameters. About 30% of the total $SiO(g)$ production was caused by $Si(g)$ condensing on SiO_2 surfaces in this special case.

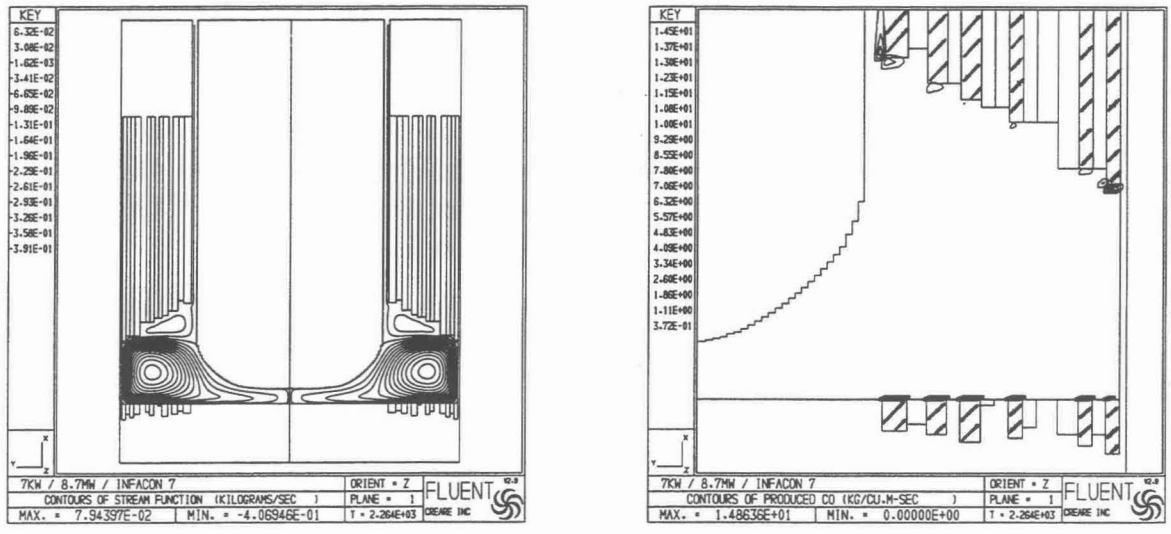

Figure 2: Furnace geometry and stream lines (left). Production of $CO(g)$ (right). SiC is hatched in the plot to the right.

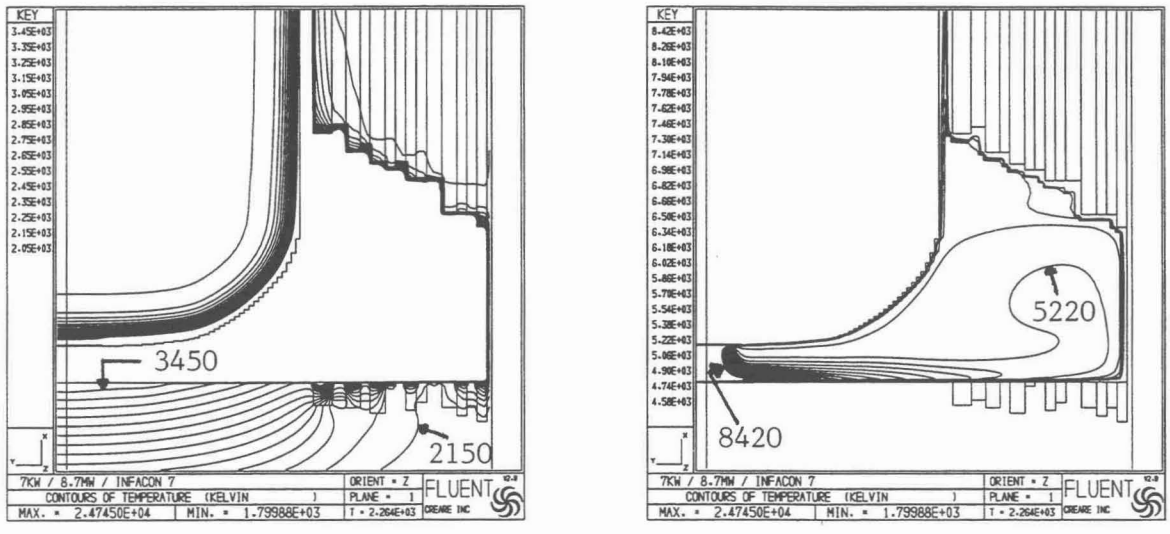

Figure 3: Temperature profiles in the metal pool (left) and in the gas up to 8500K (right). The prominent cooling effect caused by reaction (3) at the SiO_2/Si interfaces is difficult to see in this plot, but is clearly visible in closeup plots of the metal pool. $T_{max} \approx 25000$K.

The net production of silicon is very sensitive to the effective surface area of SiC. Other important model parameters are the effective heat conductivity of the silicon metal, the distribution of enthalpy and mass from the evaporation, the geometries of the particles and the electric arc, the arc current and the properties of the gas/plasma.

The simulations are highly timeconsuming, and the simulation run presented here typically took about $1 - 1\frac{1}{2}$ hours realtime on a HP-Apollo 9000/755 computer for every minute (30 time steps of 2 seconds each) progressed in the model.

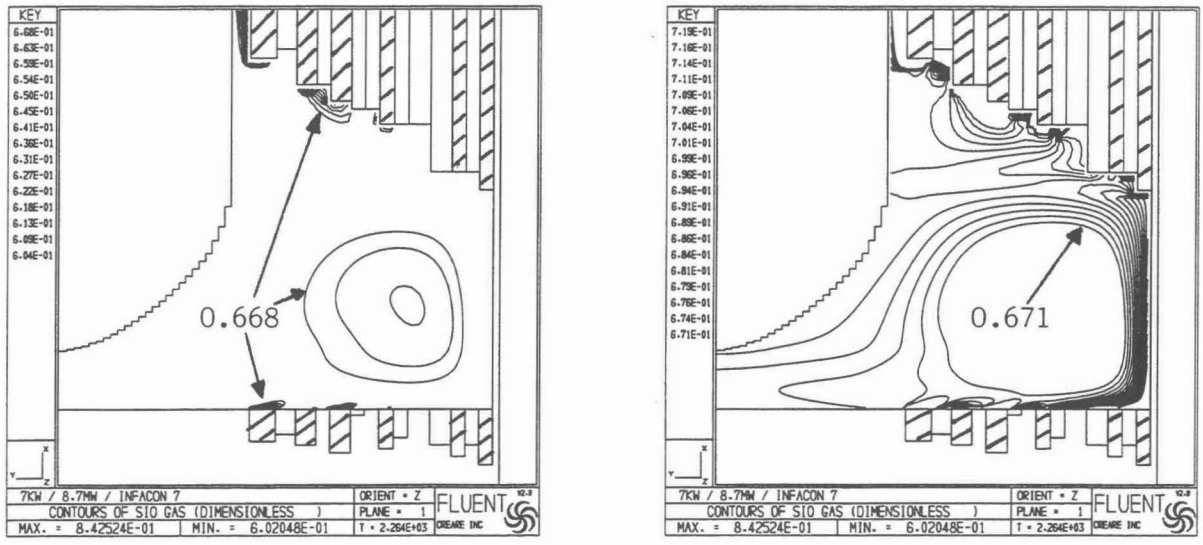

Figure 4: Mass fractions of SiO in the gas. Values from 0.6 to 0.67 (left) and 0.67 to 0.72 (right) are plotted separately to make it possible to distinguish between them even in the black and white plots. The SiO production around the SiO_2 is apparent as well as the SiO consumption (and CO production) around the SiC (hatched).

FURTHER WORK.

It is not yet decided if the model shall be improved compared to its current state in the near future. One strategy for such further work is to add chemical reactions proceeding below $1850°C$ to simulate the complete process. Another approach, which may prove more useful, is to improve one or more of the submodels already implemented or to concentrate on getting more accurate data for transport phenomena and other important input parameters. Advanced models for some of the submodels have already been developed at SINTEF/NTH. Gu [8] has developed a very advanced FLUENT based model for the interactions between electric arcs, a gas filled cavity and a silicon metal pool. Løken Larsen [12] is currently developing a FLUENT-based model for AC electric arcs. All these may prove useful for improving the model described in this paper.

CONCLUSIONS.

A 2-dimensional, cylindersymmetric dynamical model for the high-temperature part of the carbothermic silicon metal process has been developed. The results from the model,

such as gas and temperature distributions, furnace and particle geometries, fluid flow fields etc., are presented graphically in an informative and easilly accessible way. The model may prove to be an efficient research tool, improve the general understanding of the high-temperature part of the process and finally serve as the basis for a complete model of the process. Adjusting parameters and improving some submodels are needed before the model can provide realistic simulation results.

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