SIF 5036 Matematisk modellering

MATHEMATICAL MODELLING OF A GAS STIRRED SILICON REFINING LADLE

(Revised version, fall 2002)

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1 INTRODUCTION

The present note describes a complex of problems presented to us by the metallurgical company *Elkem A/S.* The case was originally proposed for the ECMI Modelling Week in 1996 and this note is a revised version of the note made in 1996.

During the production of Silicon (Si) the molten metal is poured from the furnace into a slightly conical *ladle* or *transfer vessel* (see Fig. 1) for the transportation to the moulds. During this transport oxygen-enriched air is blown into the melt through a porous plug at the bottom of the ladle. The gas flow stirs the melt and the oxygen reacts with impurities, basically aluminium (Al) and calcium (Ca), but also to a large extent silicon, forming a mixture of oxides called *slag.* This oxidation adds heat to the melt and helps to keep the temperature up during the transport. The slag is rather viscous and tends to stick to the walls and is left behind in the ladle when the content is finally poured into the moulds. The process of adding oxygen to the ladle is thus a *refining process,* reducing, *e.g.* the calcium content by roughly a factor of ten.

Elkem produces silicon this way and is interested in improving the lifetime of the ladles. The ladles degrade over time due to solidification of silicon metal and slag on their inside bottom and walls. Although the ladles are cleaned by scraping out the slag left in the ladle immediately after the ladle is emptied, this cleaning is not able to remove all solid which has been formed on the walls and the bottom of the ladle during the refining process (which typically takes of the order of one hour). The inner surface of the ladle, which in the beginning is quite smooth, gets gradually more and more rough and after 10-20 fillings, the ladle has to be extensively cleaned. This operation, and also the rough temperature conditions the ladle is experiencing, wear out the inner wall. After about 1000 fillings the heat insulation and the inner protecting coating of the ladle have to be rebuilt completely.

The ladle is equipped with a removable lid on the top to reduce heat loss by radiation. However, the lid has an elliptic opening where the melt is poured into the ladle. This hole remains open during the rest of the refining process. Near the end, the lid is removed and the content of the ladle is poured in the moulds.

The company is interesting in obtaining a better understanding of the degradation of the ladle, basically questions related to heat transfer and metal/slag solidification on the walls. The problems are quite complex and all details about the physical processes that take place during the operation are not very well known.

It has also been observed that a porous plug of slag and metal is formed near the gas inlet of the gas. The plug is permeable to the gas, and sometimes the plug apparently creates more favourable conditions for gas stirring that in other cases. The reason for this, and also the mechanism and the modelling of the plug appears to be unknown.

There seem to be extensive studies of the gas stirring in ladle systems, as discussed in the recent

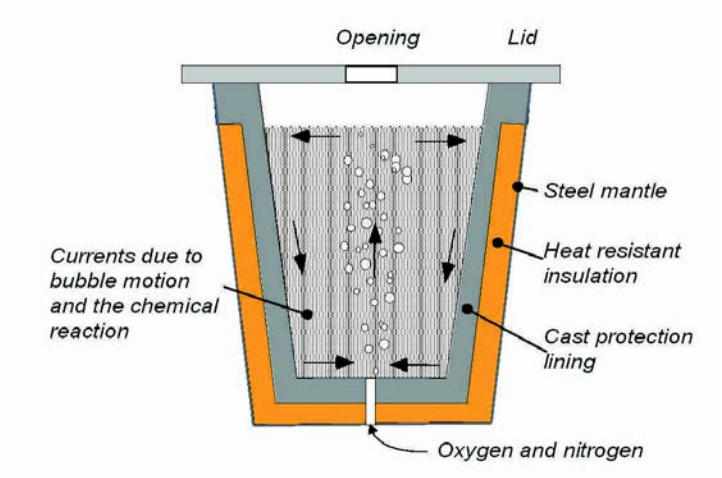


Figure 1: The principle of the silicon refining ladle.

review by D. Mazumdar and R.I.L. Guthrie (1995). This includes in particular the mechanism by which the gas bubbles create stirring of the melt, a mechanism not very different from what is observed in the familiar aquarium pump. The melt is set into turbulent motion which on the average, for gas injection at the centre of the ladle, forms an axi-symmetric flow as indicated in Fig. 1.

The properties of the slag, which basically is a mixture of oxides from Si, Ca and Al, are studied in a series of papers by Johan Kr. Tuset (Tuset, 1985). The density of the slag is almost the same as for the metal, but for slags with high SiO_2-content, the density will be less than for the metal, and the slag will flow on top of the melt. At the same time, such slags tend to be viscous. The melting point of the slag is not very well defined since this depends strongly on the composition, which is not necessarily uniform.

The basic objective of the modelling study will be to get an understanding of aspects related to heat conduction and cooling of the ladle, including the accumulation of solid metal (if this really occurs) and slag on the inner walls. It is also of interest to investigate how this accumulation will vary when the physical dimensions of the ladle are varied. In particular, is it possible to use a thinner insulation for the same steel mantle?

It is necessary get an idea about the heat budget for the process, *e.g.*, how much is lost by conduction through the walls, how much is lost by radiation, how important is the energy coming from the oxidization and from the solidification of the metal? Also, how important is the time aspect, since it takes time to fill the ladle and when it is filled, the refining process continues for some time before the content is poured into the moulds.

To get a grasp of these questions it will be necessary to solve a series of basic problems, like the so-called Stefan problem dealing with solidification of metal on the walls, the radiation loss for the given geometry, heat loss through walls of different compositions etc. This will then disclose what is important in the operation of the ladle, and will form the basis for further work.

The modelling of the forming and the behaviour of the porous plug represents another challenging exercise.

During discussions with Elkem, we have tried to obtain as much information about the pro-

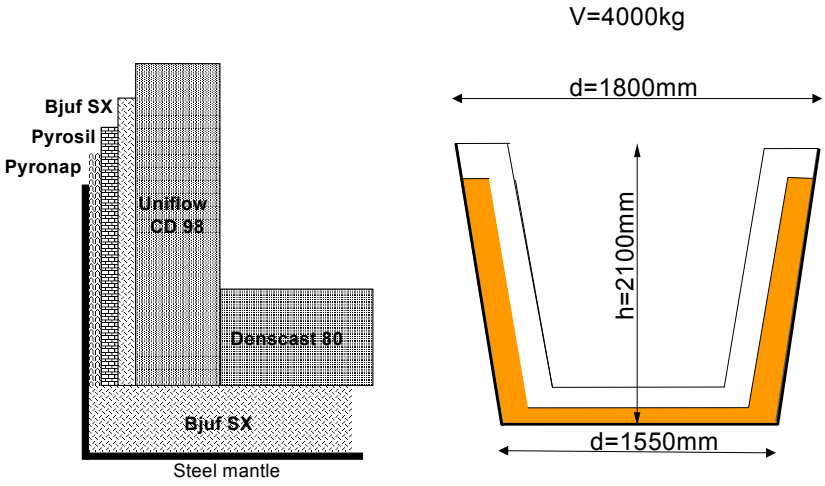


Figure 2: Dimension and construction of the ladle.

cess as possible, but with virtually no background in metallurgy it has not been easy to really understand all details.

The case is quite open ended and it is not at all possible to come up with a final and complete answer. However, in this respect the problem is typical for practical industrial mathematics where the main issue is to obtain a well-founded opinion of what matters and what is of less importance.

2 SOME BACKGROUND MATERIAL

Below we summarise information about the size and construction of the ladles and some of the physical constants that should be of interest for the modelling. Some constants are missing but should be available from the literature, whereas others have to be intelligent guesses.

2.1 THE PHYSICAL CONSTANTS

2.1.1 The ladle materials

The ladle is composed of several types of material as summarized in Table 1. The commercial names of the materials have been received from Elkem. For the steel mantle we have assumed the physical properties of plain carbon steel. *Pynonap* is an insulating mat with maximum operation temperature equal to 1150°C. The heat conduction varies significantly over the range of temperatures we may experience here. *Pyrosil* is an insulating brick, operational up to 1750°C. *Bjuf SX* is a heat and alkaline resistant brick operational up to 1710°C, but softens from about 1400°C. *Uniflow* CD98 and *Denscast* are heat resistant concretes. The dimensions and cross section of the ladle are shown in Fig. 2.

The composition of the walls and the bottom is listed in Table 2.

Material	Density	Heat capacity	Heat conduction constant
	kg/m^3	kJ/kgK	W/mK
Steel mantle	7860	0.48	40
Pyronap	250	1.04	400°C 0.07 700°C 0.13 1200°C 0.23
Pyrosil	870	0.84	200°C 0.28 600°C 0.34 1000°C 0.39 1200°C 0.44
Bjuf SX	2100	0.84	500°C 1.8 1250°C 1.7
UNIFLOW CD98	2800	0.84	600°C 2.0 1400°C 2.1
Denscast	2500	0.84	500°C 2.6 800°C 2.4 1250°C 2.5

Table 1: Physical constants for the ladle materials.

Wall composition (from outer to inner):	**Thickness (mm)**
Steel mantle	12
Pyronap	12
Pyrosil	25
Bjuf SX	25
UNIFLOW CD98	120
Bottom composition (from outer to inner):	
Steel mantle	12
Bjuf SX	128
Denscast 80	180
Top lid:	
UNIFLOW CD98	100

Table 2: Composition of the walls. The top lid has an elliptical hole of diamater 50 times 30 cm.

Silicon (Si)	Density	Heat Capacity	Heat conduction coefficient
	kg/m^3	kJ/kgK	W/mK
Solic metal	2330 @20°C	0.936	800°C 42.2 1000°C 31.2 1200°C 25.2
Liquid	?	0.908	N/A

Table 3: Physical properties of silicon

2.1.2 Properties of silicon (Si)

Properties of solid and melted silicon have partly been obtained from the company and partly from the literature. Table 3 summarises the findings. The melting temperature for silicon are 1410K, and the latent melting heat is 1.728×10^6J/kg

2.1.3 Slag

The composition of the slag that is formed during the refining process appears to be rather complicated. The main components in the melt that is tapped from the furnace seems to be

98%	Si
$\sim 0.5\%$	Fe
$\sim 0.3\%$	Ca
$\sim 1.0\%$	Al
$\sim 0.2\%$	Other

The refining process essentially leaves Fe unchanged, but reduces the Ca content to about 0.03%. It is the reduction of Ca that appears to be the main point of the refining, but the Al-content is also reduced.

The slag is then essentially a mixture of the three oxides CaO, Al_2O_3 and SiO_2. The physical properties of the such mixtures are very dependent on the relative amounts of the oxides, and metallurgists use to present this in the form of *nomograms* as the one shown in figures 3, 4 and 5. We leave to the reader to figure out how to read these nomograms by going in from each side, in a direction making 60 degrees with respect to the scale.

Slags rich in Si tend to float, and in the present process some Si is consumed in order to form a floating and, as seen in Fig. 4, a viscous slag.

2.1.4 The gas injection

The company has given us the following numbers for the heat released from the injection of Oxygen:

1 kg Ca + 0.28 Nm^3O_2 = 1.4 kg CaO + 4.4kWh

1 kg Al + 0.62 Nm^3O_2 = 1.9 kg Al_2O_3 + 8.6kWh

1 kg Si + 0.77 Nm^3O_2 = 2.1 kg SiO_2 + 4.4kWh

Heating of the nitrogen gas (which essentially just bubbles through the melt) from 25 - 1500°C requires .6 kWh/Nm^3.

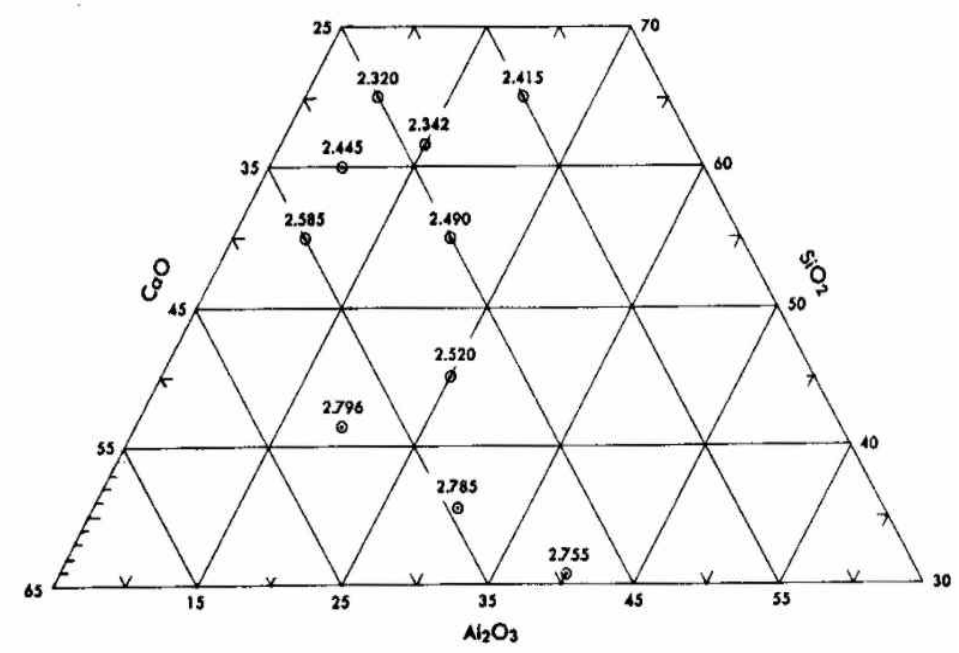


Figure 3: Density (kg/dm^3) of CaO-Al_2O_3-SiO_2 slags at 1500 ˚ C (Copied from the article by Tuset, 1985)

The oxygen gas starts reacting by the liquid metal immediately, and the gas injection is varied by adjusting the air and oxygen between the following limits:

Oxygen: 30 Nm^3/h - 50 Nm^3/h

Air: 35 Nm^3/h - 54 Nm^3/h

The "ideal" mixture appears to be about 80% O_2 and 20% N_2.

We have not been able to obtain characteristic reaction time scales, but this may probably be computed from the reaction kinetic constants. It is likely that the time of the refining period reflects the reaction time. However, the necessary refining time is also obviously dependent on the effectivity of the stirring.

3 SOME ADDITIONAL INFORMATION PROVIDED BY ELKEM

The ladle is filled by making a hole in the furnace and pouring the liquid metal into the ladle at a rate about 1–2 kg per second. This takes 15 to 30 minutes. It is not quite clear whether the ladle is completely cooled from the previous operation (about 3 hours ago) when this operation start. The temperature of the melt, 1500–1600°C, is well above the melting point of silicon (1410°C). However, one may expect that some metal solidifies when it comes into contact with the cold ladle. The oxygen-enriched air is injected from the moment the pouring begins. This is said to keep the temperature up during the pouring and the following *refining period* after the ladle is filled (strictly speaking, the refining also takes place during the filling). The length of the refining period is determined by taking a sample for analysis immediately after the pouring into the ladle

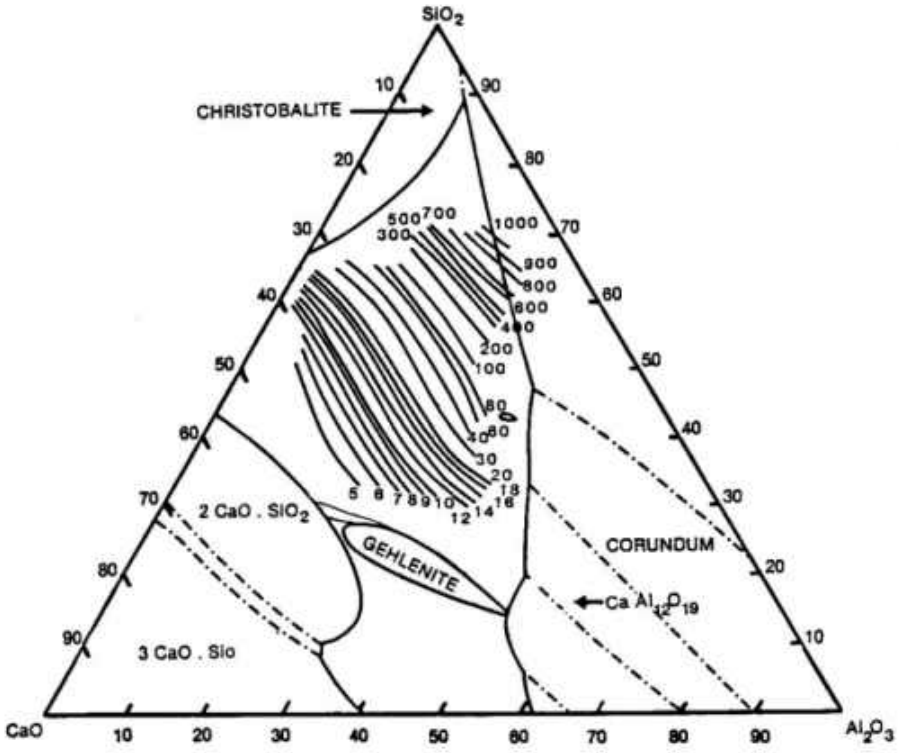


Figure 4: Iso-viscosity lines (poise) for CaO-Al_2O_3-SiO_2 slags at 1500°C (Copied from Tuset, 1985)

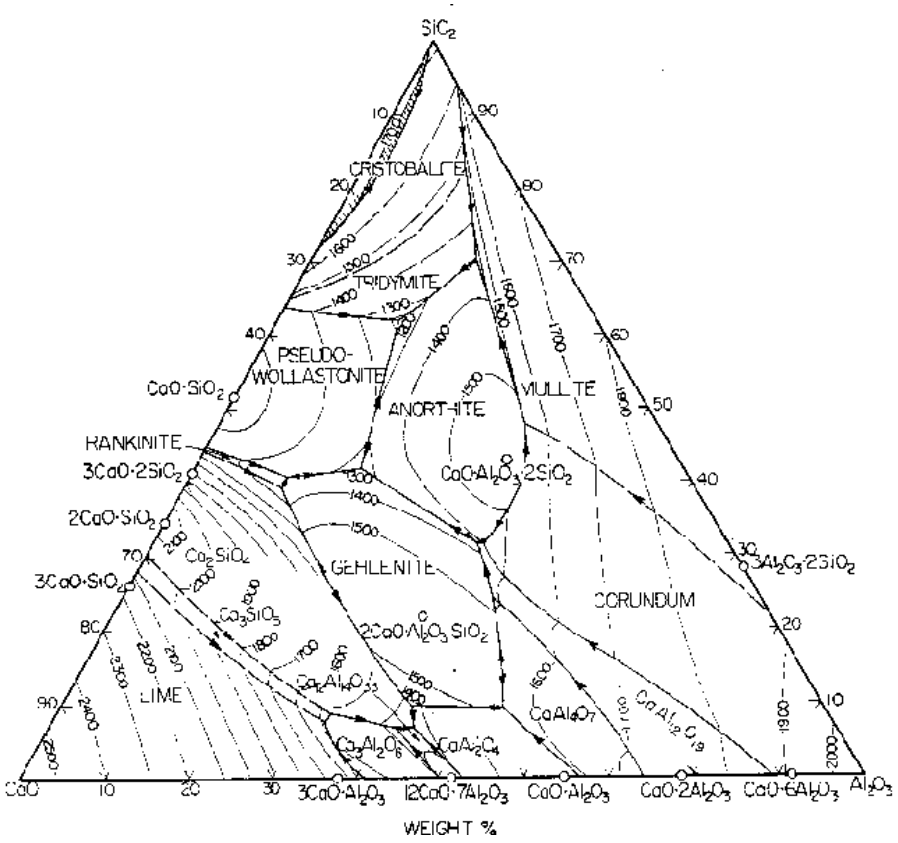


Figure 5: Melting temperature for CaO-Al_2O_3-SiO_2 slags

has started. The refining period may last from 0 – 15 minutes.

The temperature at the end of the process should ideally be about 1470 – 1480°C, and this is a temperature that is still above the melting point for silicon. As long as the temperature in the melt is about uniform (which should be true because of the stirring, it is therefore somewhat unlikely that we should have solid silicon in the ladle, at least below the level of the melt. However, when they scrape out the slag after the ladle is empty, the company says that this is mostly solid,- or very sticky. After about 10 fillings the ladle is cleaned, and when they then analyse what has been taken out altogether, they find about 50% slag and 50% silicon metal. If we do not think any solid metal is formed directly on the wall, one may well ask where the metal is coming from. Here the metallurgists at NTNU tell us that the slag, which may have considerably higher melting point that the liquid metal, may form a porous spongy solid (or at least nearly solid) which soaks metal into its pores, and this is the reason why we get so much metal in the material we scrape out from the ladle. (This is rather speculative at the moment. It has been impossible to verify that the capillary forces between liquid metal and slag are such that this is possible).

It has also been very difficult to quantify how much reaction heat from the oxidization that will be released to the melt. However, the following general description by the company should be of some help: "*About 3.5–4% of the metal disappears during the refining procedure*". This has to be found in the slag/metal mixture that is collected from the cleaning. It should therefore be possible to see how much slag that has been formed and from that be able to estimate the contributed reaction heat.

Another important number that is of interest is the heat transfer coefficient between the melt and the wall. This is dependent on the flow from the steering, and one of the metallurgists at Elkem said this number was probably in the range of 100–300 W/m^2K. It will be necessary to check whether this number is reasonable.

On the outside of the walls we have heat loss by natural convection, but this is well treated in the literature.

4 SUGGESTED MODELLING APPROACH

In the present case where we hardly know what is going on, it may seem to be very hard to get started. This is rather characteristic for many practical problems. However, the main goal of the modelling is not so much to come up with a complete model but rather get as much insight as possible.

The following approach may be reasonable, but should not be considered as the one and only way.

4.0.5 Discuss what is going on

Try to list the physical constants and mechanisms that are involved. Identify the basic conservation laws.

4.0.6 Consider a dimensional analysis

Try to identify dimensionless combinations of parameters. If we were to make experiments, what dimensionless combinations would be relevant?

4.0.7 Identify the scales

There are *e.g.*several time scales involved

- filling of the ladle
- heat loss by radiation
- heat loss by convection
- heat added by oxidization

The length scale are *e.g.*the diameter of the ladle and the wall thickness. Since it seems that the wall is "thin", comparing the thickness and the diameter, it should be sufficient to consider the heat flow trough the wall as a one-dimensional problem. Note that some of the heat conduction coefficients are rather non-linear.

It is clear that in order to find time scales for radiation and convection through the wall one may actually have to solve these "sub-problems", at least approximately.

4.0.8 Consider relevant sub-problems

Examples in addition to the problems mentioned above could be

- the heating by the oxidation
- solidification of metal on a cold wall
- temperature of the wall during the filling
- heating of the gas

4.0.9 Establish the total heat budget

Identify what really matters (*e.g.* radiation vs. conduction)

4.1 THE PLUG PROBLEM

The plug problem could be an interesting modelling problem, but unfortunately, it has not been possible to get a full explanation of what is really happening. Our current understanding is, however, as follows:

The gas enters the ladle through an injection plug. It seems to be common belief that where the gas meets the metal, an oxide layer is formed. In the present case, the oxide may have a melting point above the temperature of the metal such that the oxide forms kind of a solid cap above the plug. It seems that cap is not always stable and may break from time to time. As far as we have understood, the cap is porous such that the gas may pass through the cap, but it

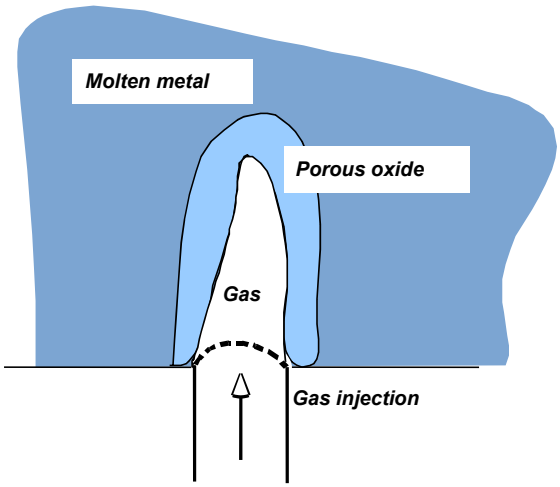


Figure 6: Formation of a sponge of porous material abond the gas inlet.

also seems to happen that cap gets rather solid such that the gas injection stops. It should be interesting make reasonable assumptions and try to model the generation and growth of such a cap, of say a spherical shape.

5 REFERENCES

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