HOW THE FERROALLOYS INDUSTRY CAN MEET GREENHOUSE GAS REGULATIONS

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ABSTRACT

For the Norwegian Pollution Control Authority (SFT) we have carried out a study on possible actions to reduce greenhouse gas emissions from Norwegian process industry including oil and gas landing. Both short term (to 2020) and long term solutions are addressed. Questionnaires answered by the industry are the basis for the study. The study will be one of the technological bases for an environmental announcement from the Norwegian government.

The present paper is confined to the ferroalloy industry, in Norway comprising the production of ferrosilicon, silicon, ferromanganese and silicomanganese. In 2007 this industry in Norway emitted about 2 million tons CO_2-equivalents.

Emission cuts and costs are given for various suggested actions. Substituting coal and coke with charcoal is an obvious solution. By improving the process conditions for silicon and ferrosilicon production, the minor emissions of other climate gases (CH_4 and N_2O) can be further diminished. Capturing CO_2 from the off-gas is another possibility. Silicon metal may be produced by electrolysis, but this is obviously a long-term project.

1 INTRODUCTION

Based on the reports from the Intergovernmental Panel on Climate Change (IPCC), most countries have accepted that greenhouse gases cause higher average temperatures world-wide. The ferroalloys industry, as all other industry, will face severe regulations on climate gas emissions.

The Norwegian Ferroalloy Producers Research Association (FFF) understood early that the industry would be accountable for their greenhouse gas emissions, and backed studies and research at the Norwegian university of Science and Technology (NTNU) and SINTEF. This first report in 1991 dealt with the calculation of emissions from ferroalloys production, giving generic CO_2 emission factors for ferroalloy production (ton CO_2/ton product) [1]. These factors were later used in the Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories [2]. Subsequent work among others described in [3] and [4] were the basis for the section on Ferroalloy Production in the 2006 IPCC Guidelines [5].

Greenhouse Gas emission data, as well as implemented and planned actions to reduce emissions were collected in 2002 for all Norwegian process industry [6]. A study on possible initiatives to reduce greenhouse gas emissions from Norwegian process industry including oil and gas landing were carried out in 2009 [7] for the Norwegian Pollution Control Authority (SFT). Both short term (to 2020) and long term solutions were addressed. Questionnaires answered by the industry were the basis for the study. The study will be one of the technological bases for an environmental announcement from the Norwegian government.

2 GREENHOUSE GAS EMISSIONS

2.1 Main Ferroalloys

This study is confined to the ferroalloys produced in large quantities, e.g. FeSi, Si, FeMn, SiMn and FeCr. These ferroalloys amount to more than 90 % of the world production. For these ferroalloys generic emissions factors are reported in the IPCC Guidelines [5].

2.2 Emissions from the ferroalloys industry in Norway.

Emission data from the Norwegian ferroalloy industry in 2007 are based on the industries reports to the Norwegian Pollution Control Authority (SFT).

Table 1: Greenhouse gas emission from Norwegian Ferroalloy production 2007

Product	Ton CO_2-eq/year
FeSi and Si	733 000
FeMn and SiMn	1 409 000
Sum	2 142 000

The greenhouse gases are primarily CO_2. CH_4 and N_2O account for only about 0.2 % for FeSi and Si-production and about 0.8 % for SiMn and FeMn production. The emissions from the ferroalloys industry account for about 4 % of the total CO_2-emissions in Norway.

2.3 Emissions from the ferroalloys industry in the world.

Based on the generic emission factors in 2006 IPCC Guidelines for National Greenhouse Gas Inventories [5] and data for world production of these ferroalloys, total emissions in the world can be estimated.

Table 2: Generic CO_2 emission factors for ferroalloy production (tons CO_2/ton product) [5]

Type of ferroalloy	Emission factor
Ferrosilicon 45 % Si	2.5
Ferrosilicon 65 % Si	3.6
Ferrosilicon 75 % Si	4.0
Ferrosilicon 90 % Si	4.8
Ferromanganese (7 % C)	1.3
Ferromanganese (1 % C)	1.5
Silicomanganese	1.4
Silicon metal	5.0
Ferrochromium	1.3 (FeCr including sinter plant: 1.6)

A compilation of world production figures for ferroalloys is found in Index mundi [8] and quoted in Table 3. Using generic emission factors (averages for ferrosilicon and ferromanganese, respectively), CO_2 emissions for each ferroalloy product are calculated. Both each figure and the sum are probably calculated too low because the generic emission factors represent fairly good operation.

The CO_2- emissions from the ferroalloys industry of 39 million tons thus amounts to about 0.1 % of all CO_2 emissions in the world (38 Gton/year according to IPCC [9]). Neither number includes CO_2-eq from other gases. Indirect emissions from the ferroalloys industry, which refer to extraction and production of raw materials (example coke oven) and production of electric energy are not included in this number. The total emissions will be several times higher if the electricity is based on coal power.

Table 3: World Production 2006 of ferroalloys (in electric furnace) and calculated CO_2 emissions

Type of ferroalloy	Production (tons)	Emission factor	CO_2-emissions (tons)
Ferrosilicon	4 210 000	4.0	16.8
Ferromanganese	2 750 000	1.4	3.9
Silicomanganese	4 360 000	1.4	6.1
Silicon metal	688 000	5.0	3.4
Ferrochromium	5 177 000	1.5	7.8
Ferromolybdenum, ferrovanadium etc,	904 000	1.1 estimate	1.0
Sum	18 100 000		39.0

3 INITIATIVES TO REDUCE EMISSIONS

The CO_2 emissions from the Norwegian ferroalloys production account for about 4 % of the Norwegian emissions. The Federation of Norwegian Industries where the ferroalloys industry are members entered in September 2009 a contract with The Norwegian Ministry of Environment to confine the emissions from the non-quota bond industry to a yearly average of 6.2 million tons for the years 2008-2012 [10] The ferroalloys industry is part of this contract. From 2013 this industry probably will be included in EU's quota system.

3.1 Improving efficiency

3.1.1 Ferrosilicon and silicon metal

So far improving efficiency and reduced specific consumption of reduction materials (coal and coke) have resulted in substantial reductions in CO_2 emissions in ferrosilicon and silicon metal production. The theoretical emission would be 3.15 kg CO_2/kg Si if it was possible to produce silicon from pure graphite with 100 % silicon yield. With 90 % silicon yield the emission would be 3.5 kg CO_2/kg Si. With coal, containing 30 % volatiles the CO_2 emission is increased to about 5 kg CO_2/kg Si, which actually is the generic emission factor for silicon metal used by IPCC [5]. Obviously the CO_2 emission from the plant can be reduced by 25-30 % if coal as reduction material is replaced by coke. Global emissions will not be changed, but the emissions are moved from a ferroalloy plant to a coke plant. Carbon lost by wind sieving in the furnace hood may add another 5 % of CO_2 emissions [11]. This loss is minimized using continuous charging instead of batch charging. However, by sealing the furnaces it may be possible to reduce the carbon requirements by 10 %, according to Dosaj et al [30].

The IPCC Guidelines have set operation specific emission factors for CH_4 from Si-metal production to 1.5, 1.2 and 0.7 kg CH_4/ton product for batch-charging, sprinkle-charging or sprinkle-charging and ≥750 ˚C in the hood [5]. The global warming potential (GWP) of CH_4 is 21[5]. By a change-over from batch-charging to sprinkle-charging it would thus be possible to decrease greenhouse gas emissions with around 17 kg CO_2/ton Si.

N_2O, with GWP of 310, is produced when the off-gas burns. Because of few data available from ferroalloys production, this gas was not included in the IPCC guidelines on ferroalloys. But the emission of N_2O in CO_2 equivalents is about 5 times the emission of CH_4, and the gas is included in the yearly reports to SFT. More measurements are needed before any action should be taken.

3.1.2. Ferromanganese and silicomanganese

The theoretical minimum consumption of carbon could be attained if CO gas from the reduction of MnO to Mn was exclusively used for pre-reduction of MnO_2 to MnO [12].

$MnO_2 + CO = MnO + CO_2$
$MnO + C = Mn + CO$

This can never be fully accomplished since some CO_2 will react with accessible carbon in the pre-reduction shaft (Boudouard reaction).

$C + CO_2 = 2\ CO$

The Norwegian FeMn and SiMn furnaces are closed and the off-gas therefore typically contains 60-80 % CO. They use metallurgical coke as the sole reductant. Kaczorowski showed that the ash content multiplied by the ash basicity is the most important variable causing high Boudouard reactivity [12, 13]. Theoretically it should be possible to reduce the CO_2 emissions with up to 40 % for a manganese production based only on MnO_2 ore. The Boudouard reaction in the FeMn and SiMn furnaces represent almost 500 000 tons of CO_2 per year (around 30 % of yearly emissions). With a duty on CO_2 emissions, an action should be to secure long-term supplies of coke with low Boudouard reactivity.

For standard SiMn produced in Norway the CO_2 emissions has been reduced with 230 kg CO_2/ton SiMn from 2000 to 2008 [7].

3.2 Using charcoal

SINTEF, Trondheim has directed and carried out several biocarbon research projects for The Norwegian Ferroalloy Producers Research Association (FFF) in the period 1997-2007, and some results are referred in the report to SFT in 2009 [7]. The goal was to elucidate the effects of using more charcoal on the furnace processes and calculate costs for this way of reducing emissions of greenhouse gas by replacing parts of the coal and coke with charcoal.

Conclusions from the feasibility studies in 1997-2001 are given in two open reports from 1997 and 1999 [14, 15] and a paper in 2001 by Eikeland, Monsen and Modahl [16]. The two first studies assume that 20 or 40 % of the fossil Carbon is replaced by charcoal, while the study in 2001 assumes a replacement of 40 %. The charcoal is supposed to be produced in Norway using recognized technology (Lambiotte, VMR, Lurgi). There is no charcoal production in Norway as it is now, but charcoal is imported. The results from the studies are summarized in Table 4. The charcoal production plants have different capacities; 60 000 tons charcoal a year [14], 12 500 or 25 000 tons [15], and 25 000 tons [16]. The CO_2 action costs are calculated in NOK per ton CO_2.

Table 4: Action costs for reducing greenhouse gas emissions in Norway and global.

Ferro-Alloy	Coal/coke cost NOK/ton fix C	**Charcoal cost NOK/ton fix C**	Charcoal Import/ technology	Invest-ment MNOK	Deprec-iation Years	CO_2 costs[2] Norway NOK/tCO_2	CO_2 costs Global NOK/tCO_2	Ref
FeSi75	Coke 1500	2500	Import			220	180	[14]
"	"	3050	Lurgi	150	20	285	220	
Si	Coal 1000	2500	Import			170	160	
"	"	3050	Lurgi	150	20	230	210	
FeSi75	Coke 1500	3350-3540	VMR	36-72[1]	15	470-530	310-370	[15]
FeSi92	"					420-500	280-340	
Si	Coal 1000					400-420	400	
FeSi75	Coke 1600	2900	Lambiotte		15		160-245[2]	[16]
		3300	VMR				235-315[2]	
		2800	Import				210	

[1] Low and high numbers for 12 500 – 25 000 tons charcoal per year
[2] Lowest costs with energy recovery, highest costs without.

The production costs for charcoal is among other factors depending on the possibility to utilize surplus energy from the charcoal process. The lowest cost for producing charcoal was with energy recovery and comparable to the price for imported charcoal. This cost would in 2001 be competitive for the smelting plants at a CO_2 quota price round 300 NOK per ton CO_2.

A recent marked review by Syvertsen and Monsen in 2008 [17] showed that charcoal import to Norway still costs approximately the same in 2007 as assumed in Table 4, while the imported amount had decreased below 20 000 tons. Norwegian charcoal import was around 60 000 tons in 2000 [16].

Both wood chips and charcoal is used in Norway today, mainly for silicon production, wood chips to get a more permeable charge and charcoal because of its high reactivity. Substitution of some coke or coal with charcoal will in most furnaces result in a slightly higher silicon yield. It is necessary to perform tests both on pilot scale and industrial furnaces before charcoal can make up the major part of carbon raw materials. It could probably be technically feasible to replace up to 80 % of the fossil carbon with charcoal, but several assessments are given in the report to SFT [7]:

- Charcoal is becoming a scarcer commodity in the international market, and import to Norway is reduced.
- Charcoal quality is very important; Controls are necessary to have consistent and unchanging properties.
- Charcoal is more expensive than coal and coke. Imposing use of charcoal will result in unprofitable production, closing down and moving production to countries with fewer restrictions.

The FeMn and SiMn industry in Norway uses mainly coke as reductant and has no tradition for the use of charcoal. At the moment we do not believe that there are process benefits by using charcoal [18, 19]. There are major differences between charcoal and the most common metallurgical type of cokes used today. Usually charcoal has a lower fixed carbon content and much higher content of volatile matters. Compared to coke industrial charcoal has higher CO_2-reactivity, lower density, and lower thermal abrasive strength. These properties are supposed to have a negative effect on furnace operation. Carbon consumption will increase with increasing CO_2-reactivity.

Previous papers at INFACON [18, 19] have presented some results from pilot production of SiMn in a 150 kW open furnace at SINTEF/NTNU. Different reductants with a large variation in CO_2 reactivity were tested. The aim was to produce SiMn with 18 % silicon and see if any significant difference was found in the coke-bed by using different reductants. Metal with 18% silicon was produced using metallurgical and reactive coke, but not in the first experiments with charcoal. The last experiment showed that SiMn with 18% Si can be produced when charcoal is substituting 50% of the coke on a fixed carbon basis.

A mixed charge of charcoal and coke has previously been used to a large extent in Brazil with up to 80 % fix C from charcoal. These furnaces are principally smaller than most of the Norwegian furnaces, and usually semi-covered or open, not covered as in Norway. However, it should be possible to use some charcoal in the Norwegian plants too, provided necessary tests are carried out. But the high price of charcoal will increase production costs substantially.

3.3 Energy recovery

The total energy input in the Norwegian ferroalloy industry was 6.9 TWh in 2007 [31] of which 99 % comes from electricity, based on hydro power. In addition energy also comes from the consumption of raw materials and reductants as coal and coke. Possible energy recovery not yet utilized was identified in the McKinsey study [31] to 5.1 TWh, constituting of 3 main categories.

- 3.7 TWh (70 %) can be recovered from cooling water (furnaces, boilers), hot metal and slag. Most of the hot water will be at a relatively low temperature (30-50 ºC) and therefore of low quality. Examples of use are heating of fish farms, rose production, etc. It will be hard to find adequate users. Further processing through heat pumps is needed to be exploited.
- 1,1 TWh (20 %) can be recovered from the off gases as electric power, but demands high investments. Cooling of steam turbines is included in the category above.
- 0.4 TWh should be possible to export to other industry as chemical energy (CO gas from FeMn and SiMn production).

Use of recovered energy in the off gas will reduce the use of energy from other sources. It the energy source is coal, oil or gas the recovered energy will indirectly reduce greenhouse gas emission. No direct credit is given for these reductions in Norway, but public support may be available for investments. Increased price for energy will make energy recovery more profitable.

3.3.1 Ferrosilicon and silicon metal

The off gas from the semi-covered furnaces must be cooled before entering a filter in order to remove silica dust. The off-gas with a temperature of 700 to 800 ˚C may be cooled in a heat exchanger or a

boiler producing hot water or steam, which may be used in a turbine producing electric energy. Electricity is produced from the off gas from two plants, so far.

It may be possible to increase the energy recovery from semi-covered Si and FeSi furnaces if the furnaces were covered. A major challenge in this respect is the need for stoking and the handling of dirty unburned off gas. The possibilities for closing these furnaces should be looked into once more.

3.3.2 Ferromanganese and silicomanganese

The off-gas from sealed furnaces typically contains 60-80 % CO. The energy content in the gas from the Norwegian producers is in total about 2 TWh/year. The gas can be utilized as a synthesis gas, or burnt to produce thermal energy or electricity. If it is used instead of coal, oil or natural gas to produce thermal energy or electricity, the CO_2 emissions are reduced correspondingly.

At present time energy is recovered at 3 of the 4 plants in Norway, but the entire recovery potential is not yet developed [7]:

- One plant produces electricity (88 GWh/year) and energy for district heating (12 GWh/year)
- Another plant sells 66 % of the off gas to neighbour industry
- A third plant sells 90 % of the off gas to neighbour industry for energy purposes.

3.4 Electrolytic production

Traditionally, metals such as magnesium and aluminium that are not suitable for carbothermic reduction are produced by molten salt electrolysis. With reasonable yield and production rates, an electrochemical process might also be a simple and low cost alternative to chemical and metallurgical routes to SoG Si. Both in electrolysis and electro refinement processes, the different degree of nobility (= thermodynamic stability) of the various elements is utilized to design a fairly selective production process.

In Si electrolysis, the raw material such as SiO_2 (s), K_2SiF_6 (s), SiF_4 (g) or $SiCl_4$ (g), is normally dissolved in an electrolyte and decomposed. The Si ions are reduced at the cathode to elemental silicon. The anode reaction product might be O_2, CO, CO_2, F_2 or Cl_2 evolution depending on the raw material and anode material. In an electro refining process (as opposed to electrolysis), the raw material is impure silicon, typical metallurgical grade Si (MG Si).

In the eighties there was a general “boom” in research for new Si electro deposition processes. Rao et al. seemed to come quite far with their electrolysis process of K_2SiF_6 in LiF-KF [20]. Here the main objective was to deposit Si coatings on suitable substrates that could be used directly as wafers. They obtained coherent uniform films with ≥ 99.99% purity. A patent by Grong and Tuset from 1982 describes an electro refining process where pure Si is produced from MG Si in its molten state at T>1400°C [21].

In later years, the renewed interest in developing less costly production methods for SoG Si has resulted in a number of patents and publications. An electrolysis process suggested by Stubergh is based on quartz dissolved in cryolite [22]. Fluoride melts are in many cases preferred over chlorides for electro deposition. However, the corrosiveness of the various fluoride electrolytes and difficulties in separating the metal and bath have led to several investigations in chloride melts. Many chloride melts are easily dissolved in water. Olsen’s patent application describes a process for SiO_2 decomposition in $CaCl_2$-CaO melts [23]. The FFC process is based on a similar electrolyte, with the difference that SiO_2 is not dissolved in the melt [24, 25]. A quartz pellet constitutes the cathode and Si powder is formed “in place” while O^{2-} leaves the cathode and is oxidised at the anode.

A recent Russian patent describes a process where SiO_2 (s) is converted to SiF_4 (g), which is entered into a fluoride electrolyte and decomposed to Si powder and F_2. The F_2 is recycled to react with SiO_2 to SiF_4. Silicon is then discharged from the electrolyser in the form of silicon powder slurry and eutectic melt. Thereafter, silicon is separated from the molten electrolyte by dissolving this melt in anhydrous hydrogen fluoride. The resulting composition HF+ (LiF-KF-NaF) in the form of liquid phase and silicon particles is filtered. The liquid phase is subjected to distillation of hydrogen fluoride, which is reused in the dissolution stage [26].

The purpose of this research has been to develop cost-effective and environmentally good alternatives to the present dominant method with chlorination of MG-Si to chlorosilane, followed by decomposition to SoG-Si. This Siemens process is very energy intensive (around 200 kWh/kg). If this electricity is produced in coal-fired power plants, this will add an emission of about 400 kg CO_2 per kg SoG-Si. The amount of electric power used in an electrolytic process is expected to be of the same order as in the arc furnace. Provided that the primary anode product is CO_2 as it is in the Hall Heroult process[1], the CO_2 emission from the electrolytic process will be half the emission from the arc furnace process, if Carbon anodes are used. If inert anodes are used, the electrolytic process gives no CO_2 emissions.

3.5 Sequestration of CO_2

Research has been carried out on the reaction of blast furnace off-gas with the slag from steel production [27]. Part of the off-gas is sent through a column (tower) filled with crushed slag and water flowing through. The reactions for CaO may be:

$$CaO + H_2O = Ca^{+} + 2\ OH^{-}$$

$$CO_2 + H_2O = 2\ H^{+} + CO_3^{2-}$$

$$Ca^{2+} + CO_3^{2-} = CaCO_3$$

MgO will probably react in the same way.

Calculations have shown a theoretical possibility of binding about 20-30 % of the total CO_2 emissions from SiMn production in the SiMn slag by carbonization. A standard SiMn slag contains typically around 20 % CaO, 9 % MgO, 8 % MnO, and 2 % K_2O, and around 1200 kg slag is produced per ton metal. The off gas from the SiMn production could all the same be used for carbonization of these oxides in the slag since this slag often ends up in land fillings.

Another possibility for sequestration of CO_2 in the SiMn slag is to use the CO_2 containing off gas from the SiMn production for direct carbonization of crushed SiMn slag or slag granules in a fluid bed (or packed bed) [28]. Appropriate temperature for carbonization of CaO in the slag would probably be around 500 °C [29]. Carbonization of MgO should be carried out below 350 °C since $MgCO_3$ decomposes at 350 °C. These possibilities for sequestration of CO_2 in SiMn slag should be studied.

4 CONCLUSIONS

Greenhouse gas emissions (GGE) from ferroalloys production amount to 4 % of Norwegian GGE. Requirements to reduce GGE cannot be met by a single initiative, but by a set of actions. The efficiency and yields are fairly good, but improvements resulting in decreased GGE are still possible. One example is the Boudouard reaction in the FeMn and SiMn furnace which represent a significant amount of CO_2. Charcoal gives no GGE, and becomes a possible alternative to coal at a quota price of 300 NOK/ton CO_2[2]. Energy recovery will not reduce GGE from the plant, but will give reduced emission on national and global level. Electrolytic production is an option especially to produce solar grade silicon directly. Sequestration of CO_2 with process slag can give a small reduction of GGE. It may be a reasonably priced alternative, and ought to be investigated.

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