

The Use of Biocarbon in Norwegian Ferroalloy Production

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1 ABSTRACT

The Norwegian ferroalloy industry is at present responsible for about 3 million tonnes of CO₂-emissions a year. These come from fossil carbon, mainly coal and coke that are used as reductants in the smelting process. If biocarbon is used as a substitute for coal and coke it is possible to reduce these emissions considerably. The term biocarbon means carbon from biological and renewable resources, for example charcoal and wood chips.

This paper gives a brief review of the reasons for using biocarbon and presents the trends in biocarbon use in the Norwegian ferroalloy industry. Results are presented from the pilot scale smelting experiments that have used biocarbon as reductant. As charcoal strength and reactivity are important for process performance, these features have been measured. Although the charcoal is not produced in Norway, the possibilities for increasing the charcoal yield in the carbonization process have been studied and the results from laboratory experiments are included.

2 INTRODUCTION

The Kyoto agreement encouraged increased attention to be paid to emissions of CO₂ due to its role in potential global warming. From a global reference, CO₂-emissions from a process using biological carbon combined with hydropower should be very small if there is a balance between the carbon in the process and the growth of the trees used to produce the biological carbon.

In 1997, the Norwegian Ferroalloy Producers Research Association (FFF) launched a five-year research project aimed at clarifying the consequences of increased use of biocarbon, and the possibilities the Norwegian ferroalloy industry had to reduce their share of Norwegian fossil CO₂-emissions. Central participants in this research are SINTEF and the Østfold Research Foundation,

together with the industry representatives, Elkem ASA and Fesil ASA.

In Norway, biocarbon is expensive compared to coal and coke and cost reductions will be essential for the success of the project. As charcoal is not produced in Norway, large amounts are imported and the possibilities for cutting costs through a more rational collection and transportation of timber are studied /1, 2/. Other ways of cutting costs lie in increasing the yield of charcoal from the carbonization process, by the choice of process technology and by optimum use of surplus process heat. However, the most important possibilities for process improvements are likely when charcoal is used in the production of ferrosilicon and silicon.

The Norwegian production of ferroalloys is well above one million tonnes per year and includes ferromanganese (FeMn), silicomanganese (SiMn), ferrochromium (FeCr), silicon (Si) and ferrosilicon (FeSi). Norwegian producers are Elkem ASA, Fesil ASA, Finn fjord Smelteverk A/S, ERAMET Norway, Tinfos Jernverk and Globe Norge Hafslund Metall.

The method of generating electric power is important when determining the total amount of CO₂ emitted from ferroalloy processes. This is clearly shown in Table 1³.

Table 1 - Specific CO₂-emissions from ferroalloy processes³.

Electric power	Total specific CO ₂ -emissions (kg CO ₂ /kg alloy produced)	
	Si	FeSi 75 %
Hydropower/	4.3	4.4
Gas power	10.6	8.5
Coal power	17.7	13.5

In Norway, the electric power supply is based on hydropower, which has very low CO₂-emissions. Specific CO₂-emissions from the production of FeSi and Si are 3.5-4.7 tonnes⁴ compared to 1.0-1.6 tonnes⁵ from the production of FeCr, FeMn, and SiMn. Because of possibilities of process

improvements, the biocarbon project has so far focused on CO₂-emissions from the production of FeSi and Si. Charcoal and wood chips are currently used to a small extent, but not in the production of manganese and chromium alloys in Norway.

3 INDUSTRIAL MOTIVATION FOR USING BIOCARBON

The most important economic parameter for the choice of reduction materials is the specific price per tonne of Fixed Carbon. If the process performance is unchanged the Fixed Carbon price should determine the choice of carbon material.

Normally, bio-based reduction materials such as charcoal or wood chips have a higher price for Fixed Carbon compared to coal and coke. Nevertheless such materials are used both for silicon production and even for the production of some ferrosilicon qualities. Other reasons than the strict conversion cost in the use of bio-based carbon materials are presented in Table 2.

Table 2 - Main reasons for using biocarbon.

MAIN REASONS	EFFECTS ON THE PROCESS
PROCESS IMPROVEMENT:	Improved furnace gas distribution.
	Possible decreased charge segregation.
	Increased condensation area and mass for SiO-gas.
	Improved electrical resistance.
	Large amount of wood chips will decrease the retention time of quartz.
	Wood chips may change the energy content in the off-gas.
	Better SiO reactivity.
PRODUCT QUALITY:	Bio-based carbon material may have less impurities than fossil carbon. This may improve the silicon and silica quality.
ENVIRONMENTAL EFFECTS:	Reduction in the emission of fossil-based CO ₂ .
	Possible lower content of trace elements than fossil-based carbon materials.
	Reduction in sulphur will lower SO ₂ -emissions.

4 TRENDS OF USING WOOD CHIPS AND CHARCOAL IN NORWAY

At present, wood chips are chopped into pieces that are coarser than matchboxes. All fines are screened to make it possible to handle wood chips through bins and feeders, but there will always be a chance of bridging and stops in the flow. The traditional high volume consumer of wood chips is the silicon process. In Europe, coal is the main source of carbon in this process, but for each tonne of silicon between 800 to 1 200 kg of wet wood chips is normally mixed into the charge to raise the yield and smoothen the furnace operation. This practice has been unchanged for years. There may be a means of agglomerating a mix of biocarbon fines and other materials. Some tests have been run in full-scale operation in Norway, but it is difficult to obtain the required properties at an acceptable price.

The consumption of charcoal has more than doubled during the nineties. Today it is normal to supply more than 10 % of the fixed carbon from charcoal. Though this source of fixed carbon costs much more than carbon from low ash coal, its valuable properties (Table 2) have made this development possible.

South America and Asia have been the main suppliers of metallurgical charcoal to Norway. The processing of charcoal may be simple and the quality can fluctuate considerably in a single cargo. Since it seems difficult to establish economical production of charcoal in Norway, ferroalloy producers in this country are interested in charcoal projects abroad. Important conditions are environmentally sustainable logging in the woods, compatible costs and industrialized production making it possible to optimize the charcoal qualities for metallurgical use.

The above-mentioned effects such as the reduction of trace elements, SO₂ and fossil CO₂ are proportional to the amount of biocarbon. Other effects like silicon yield and general furnace operation are not linearly dependent. Both the silicon and the ferrosilicon processes seem to reach the full benefit of the charcoal before it replaces 50 % of the total carbon. This means that a substantial replacement of coal and coke above this level cannot be compensated for by the metallurgical properties.

The Kyoto agreement commits the industrialized world to reduce the emissions of fossil CO₂. It is most probable that Norwegian industry will be allowed to emit a certain percentage of its normal rate of emissions free of charge while emissions exceeding this level must be bought as emission quotas on the open market. This situation has influenced the ferroalloy industry to start to search for cheaper/better biocarbon and optimize processes to utilize their valuable properties as much as possible. However, at present no one knows what will be the cost if/when you have to buy CO₂ quotas in the future.

Norway introduced a tax on emissions of sulphur dioxide (SO₂) from the use of coal and coke on 1 January 1999. This tax means 4 – 5 USD/t of silicon/ferrosilicon. As the sulphur content of charcoal is only 5-15 % of the level found in coal and coke, charcoal will also save some of these SO₂-taxes.

The Kyoto agreement has substantially influenced the interest in biocarbon in the Norwegian ferroalloy industry. In the future it may become more costly to make a major switch from coal/coke to charcoal compared to buying CO₂ quotas, but the ongoing work will move the economically optimum amount of biocarbon to a higher level.

5 CHARCOAL PRODUCTION

Charcoal is manufactured from biomass by pyrolysis in large kilns or retorts. By-products are pyroligneous liquid and gases (i.e. volatile matter). The yield of the different reaction products varies with the biomass species and heating conditions. Larger particle sizes and slow heating favour the formation of charcoal.

Between 30 and 60 % of the energy content of the feedstock will be accumulated in the charcoal after carbonization (depending on the charcoal yield). Chemicals (methanol, acetic acid, smoke flavours, etc.) can be extracted from the pyroligneous liquid. However, these are only “niche” products and usually the pyroligneous liquid is burnt together with the gases to provide heat for the carbonization. Surplus energy can be used for pre-drying the feedstock or producing hot water or steam in boilers for heating.

There are over a hundred concepts and methods for producing charcoal. Figure 1 shows the three most common principles of heating that are used to initiate carbonization and maintain high temperatures during the processes. For those who are interested in industrial carbonization processes, we refer to a recent survey given in Ref. 6 and a more comprehensive review given in Ref. 7.

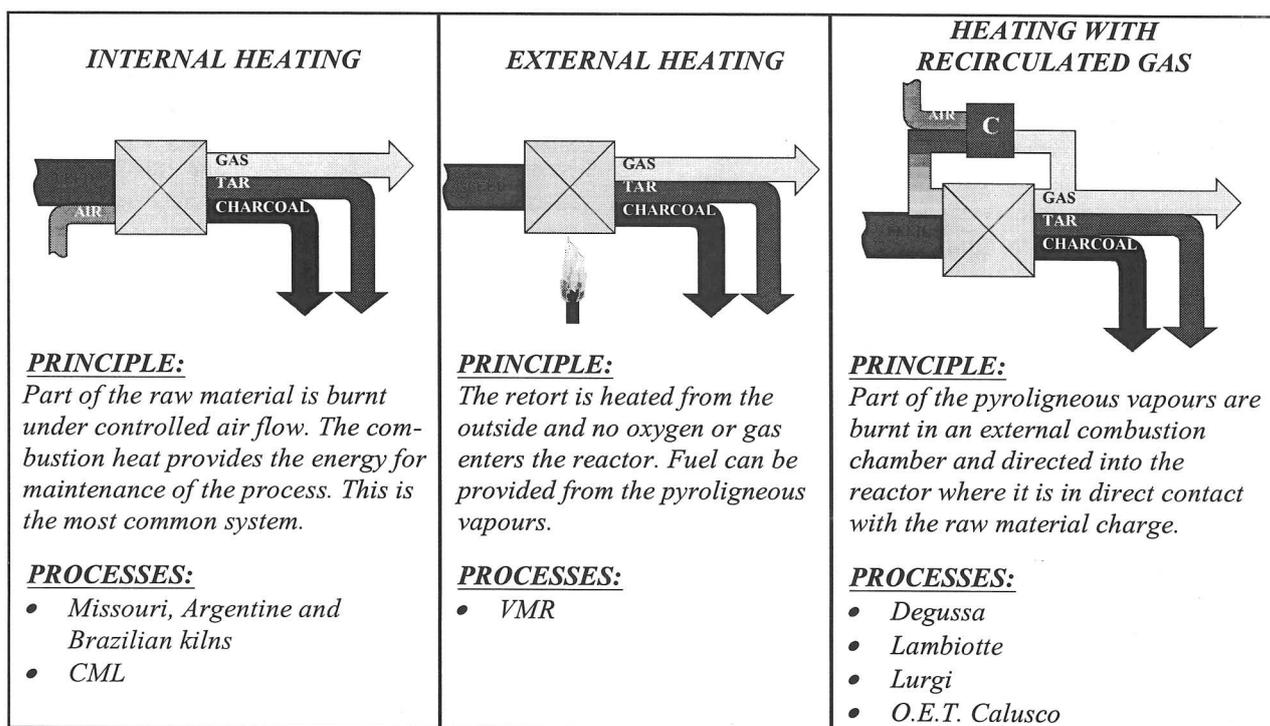


Figure 1 - Heating systems^{6,7}

The charcoal yield (y_{char}) is a measure of the efficiency of the pyrolysis process. We define the charcoal yield as $y_{\text{char}} = m_{\text{char}} / m_{\text{bio}}$, where m_{char} is the dry mass of charcoal produced by the pyrolysis of a biomass feedstock with a dry mass m_{bio} . Unfortunately, this definition is intrinsically vague because charcoal is not a well-defined chemical compound. Some charcoals are nearly pure carbon; others have undergone only partial pyrolysis and retain significant amounts of oxygen and hydrogen.

In order to make meaningful comparisons, a better measure of pyrolysis efficiency is needed. Consequently, we emphasized the efficiency of the conversion of the organic matter in biomass to a nearly pure carbon. To measure this efficiency, the charcoal is subject to a proximate analysis according to the standard ASTM test procedure. In essence, the charcoal is carbonized in an inert environment at a high temperature. Thereafter, its volatile matter content is defined as $\%VM = 100 \cdot (m_{\text{char}} - m_{\text{cc}}) / m_{\text{char}}$, where m_{cc} is the dry mass of the carbonized charcoal that remains after heating. Likewise, its ash content is given by $\% \text{char ash} = 100 \cdot m_{\text{ash}} / m_{\text{char}}$, where m_{ash} is the dry mass of ash that remains following the combustion of the carbonized charcoal. The Fixed Carbon content of the charcoal is defined as $\% \text{Fixed Carbon} = 100 - \% VM - \% \text{char ash}$. We define the Fixed Carbon yield (y_{FC}) of a carbonization process as: $y_{\text{FC}} (\text{wt}\%) = y_{\text{char}} \cdot \% \text{Fixed Carbon}$.

Pyrolysis experiments

In order to study the heating conditions (heating rate, temperature, residence time and pressure) that give the highest charcoal and Fixed Carbon yields, pyrolysis experiments were carried out in three different reactors. In the first of these, at SINTEF,

charcoals were produced at atmospheric pressure in a 3.7 litre stainless steel retort placed in a Nabertherm muffle oven. The retort has a capacity of approximately one kilo of dry wood per batch. In the second reactor, also at SINTEF, pyrolysis experiments were carried out on single particles in a macro thermogravimetric analyzer (TGA). Both the muffle oven and the TGA are equipped with a programmable temperature controller, which provides control of the heating rate, temperature, and reaction time. In the third reactor at the University of Hawaii, charcoals were produced in a pressure vessel with an internal volume of 7.2 litre. At the bottom of this reactor there are two internal 4 kW cartridge heaters that are controlled by a temperature controller. The reactor's outer wall is heated by four band heaters totalling 4.4 kW. The pressure within the reactor is controlled by a back-pressure regulator. Combustible gases released by the back-pressure regulator are burnt in a flare.

Pine (*Pinus silvestrus*), the most abundant wood species suitable for charcoal production in Norway, was chosen as feedstock. The typical dimensions of the feedstock pieces used in the muffle oven and pressure vessel were 3x3x5 cm. For the TGA experiments, dowels (D = 4.5 cm, L = 7 cm) were cut from wood logs with a hole saw.

Table 3 displays some of the results from the experiments in the muffle oven and pressure vessel. More details are available in Ref. 8. The charcoal yields for rapid heating to 450 °C (R#2) and 550 °C (R#4) are 29.1 % and 24.6 %, respectively. Higher pyrolysis temperatures give charcoals with a higher Fixed Carbon content, hence the Fixed Carbon yields for the rapid heating experiments are practically the same (~21 %).

Table 3 - Charcoal and Fixed Carbon yields obtained from pinewood under different heating conditions.

Heating	Temperature T_{peak} (°C)	Heating time (min)	Retention time at T_{peak} (min)	Pressure (MPa)	Proximate Analyses ^a			Average	
					VM (wt%)	Fixed C (wt%)	Ash (wt%)	Charcoal yield (wt%)	Fixed C yield ^b (wt%)
Slow (R#1)	350	180	60	0.1	39.4	60.1	0.50	39.9	24.0
Rapid (R#2)	450	60	60	0.1	26.6	72.4	1.00	29.1	21.1
Slow (R#3)	450	240	60	0.1	25.0	74.6	0.40	32.1	23.9
Rapid (R#4)	550	60	60	0.1	14.4	84.5	1.10	24.6	20.8
Slow (R#5)	550	300	60	0.1	12.6	86.6	0.90	28.8	24.9
Rapid (R#6)	400	60	60	1.0	35.9	63.1	1.00	43.2	27.3

^a % of dried charcoal

^b % of dried feed material. Fixed Carbon yield (wt%) = Charcoal yield · % Fixed Carbon

Less than half of the carbon contained in the feedstock is converted to Fixed Carbon by the rapid pyrolysis at atmospheric pressure (0.1 MPa). The bulk of the carbon leaves the reactor as tars and gases (i.e. volatile matter). An efficient carbonization process must minimize the formation of these volatile byproducts by converting them to solid carbon, water, and carbon dioxide. The scientific literature⁹ offers guidance concerning conditions that improve the yields of carbon from biomass. Larger particle sizes and slow heating favour the formation of carbon by enhancing the contact time of the volatiles with the solid carbon product. It is well known that the volatiles are not stable at elevated temperatures in the presence of charcoal, or decomposing solid biomass. The volatiles adsorb onto the surface of the solid and quickly carbonize, releasing water, carbon dioxide, and methane as byproducts.

The results given in Table 3 show that an increase in heating time raises the Fixed Carbon yield from 21 % to 24 % (R#1 at 350 °C, R#3 at 450 °C). Slow heating to 550 °C results in a further small increase in the Fixed Carbon yield to 25 %.

Fundamental studies of the influence of pressure on biomass pyrolysis have shown that very high yields of charcoal are obtained when pyrolysis is conducted at elevated pressure in a closed vessel where the vapours are held captive and in contact with solid products of pyrolysis^{10, 11}. Corroborating these findings, pyrolysis of pine at elevated pressure (1.0 MPa) increases the Fixed Carbon yield to 27.3 % (R#6). Remarkably, carbonization at elevated pressure produces high yields without a prolonged reaction time, and no significant holding time at the peak temperature is needed.

Both the results from thermogravimetric analyses¹² and from the retort experiments in the muffle oven at SINTEF are included in Figures 2 - 4. The results in Figure 2 show that a slow rate of heating clearly gives the best yields of Fixed Carbon at atmospheric pressure. The large gap in the results from the thermogravimetric analyses for same rate of heating, can be attributed to the other parameters which were investigated: high and low temperature (450-550 °C), short and long retention time at peak temperature (10-60 min), and humid and dry samples. A positive effect of prolonged retention time on the yield of Fixed Carbon was also observed in these TGA experiments.

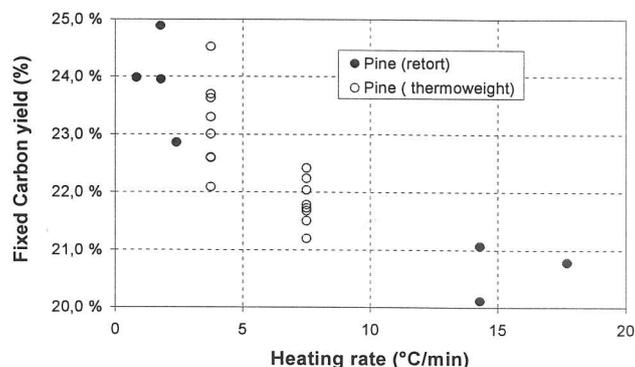


Figure 2 - Effect of heating rate on Fixed Carbon yield.

Compressive strength

The charcoals obtained a compressive strength that was 3-4 times higher in the direction parallel to the wood fibre than radial to the fibre. Most likely charcoals will be crushed at their weakest point. Hence, most attention should be paid to the compressive strength radial to the wood fibre.

A beneficial effect of slow heating on the radial compressive strength can be observed in Figure 3. It should be emphasized that the points marked as pine (retort) represent the mean value of 7 replicates, while only single test pieces were available from the experiments carried out in thermoweight.

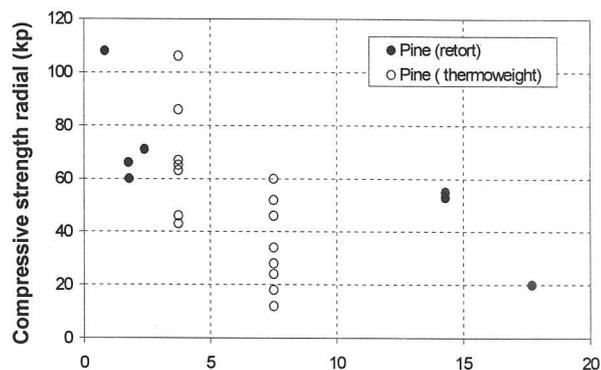


Figure 3 - Effect of heating rate on compressive strength radial to the wood fibre.

Again, the large gap in the results for the same rate of heating can be attributed to the other parameters that are investigated. The other parameters seem to be of less importance than the heating rate for charcoal strength.

SiO reactivity

The reactivity test was carried out at 1650 °C according to a standard procedure that is well described in the literature¹⁵. The reactivity number indicates the amount of SiO gas passing the actual sample without reacting. Hence, a low reactivity number indicates a high reactive material.

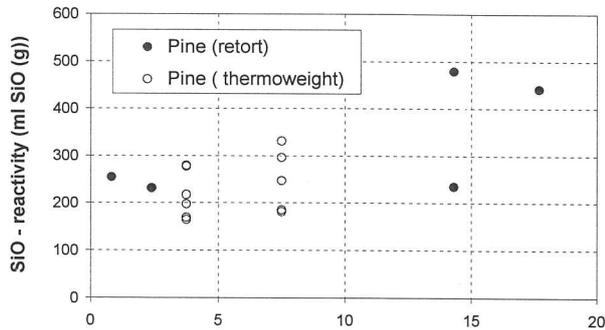


Figure 4 - Effect of heating rate on SiO reactivity.

The results in Figure 4 indicate that a slow rate of heating during the pyrolysis process has a beneficial effect on charcoal reactivity, resulting in low SiO reactivity numbers. These charcoals all obtained low reactivity numbers, in the range 160 - 480 ml SiO (g), and are to be considered as normal to highly reactive charcoals. In comparison a typical highly reactive material has a reactivity number in the range 900 - 1600 ml SiO, while coals can be found in the range 900 - 2000 ml SiO.

In addition to this, the best SiO reactivities were in most cases obtained for a short retention time at peak temperature, while temperature level seems to be of less importance for charcoal reactivity.

The charcoals produced at elevated pressure (1.0 MPa) also obtained low reactivity numbers, in the range 200 - 250, and can be considered as highly reactive charcoals.

6 EXPERIENCE FROM USING CHARCOAL IN A PILOT FURNACE

Smelting experiments have been performed in order to demonstrate that ferrosilicon and silicon metal can be produced without any use of fossil reductants.

During normal production about 800 kg coal and 375 kg coke on a dry basis are consumed to

produce a tonne of FeSi 75 %. In the production of silicon a share of 10 % Fixed Carbon from wood chips is considered typical, but still nearly 1400 kg coal is needed per tonne. The consumption results in fossil CO₂-emissions of 3.9 tonnes per tonne FeSi 75 % and 4.7 tonne per tonne silicon⁴. By replacing these reductants by biocarbon it may be possible to nearly eliminate the fossil CO₂-emissions, as shown in Figures 5 and 6. The remaining CO₂-emissions will be due to the consumption of electrodes.

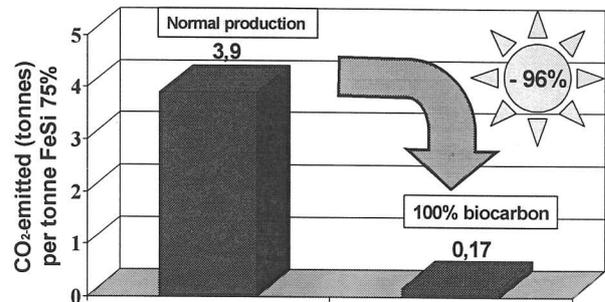


Figure 5 - Potential reduction in fossil CO₂-emissions in the production of FeSi75%.

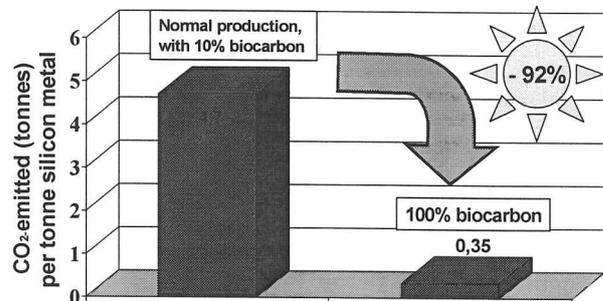


Figure 6 - Potential reduction in fossil CO₂-emissions in the production of silicon.

The smelting of ferrosilicon and silicon with charcoal as a reductant was performed in the single electrode pilot furnace shown in Figure 7. The bottom lining has a cast-in contact and the carbon electrode is operated in order to maintain a constant load. The photo shows the tapping of silicon using charcoal as a reductant. Metal was tapped each hour.

Towards the end of each tapping, the electrode was lowered and the furnace switched off for a short period in order to poke the materials on the furnace top, as shown in Figure 8. Between each tapping, the furnace was operated at a load of 130-160 kW

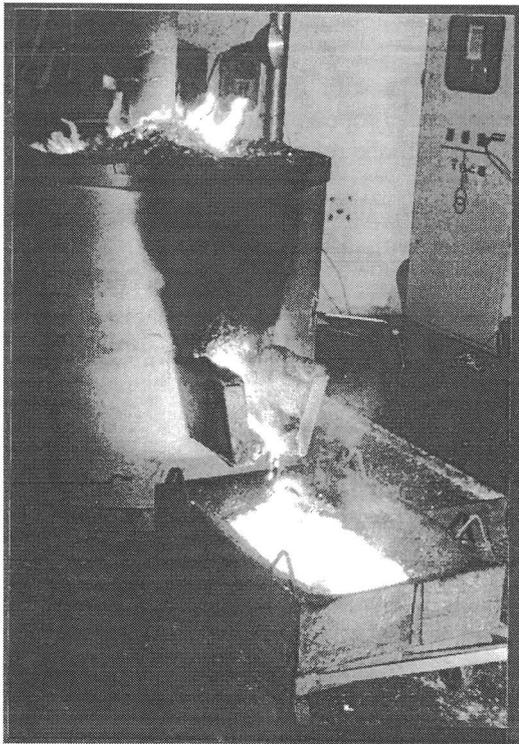


Figure 7 - A single electrode pilot furnace. Tapping of silicon with charcoal as reductant.

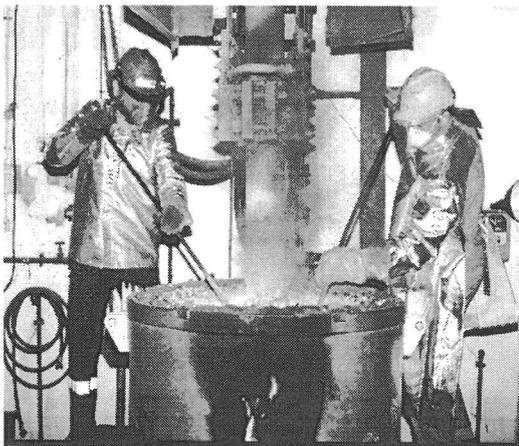


Figure 8 – Poking during ferrosilicon smelting.

(35-38 V, 3.5-4.5 kA) while a new batch of mixed raw materials was charged manually.

Analyses of the reductants that were used are shown in Tables 4 and 5. The charcoal from Brazil was produced industrially from eucalyptus trees, while the charcoal from Europe was produced from European mixed wood in pilot experiments in a VMR plant. Spanish quartz containing 99.7 %

Table 4 - Proximate analyses of reductants.

	Charcoal from		Reference Coke
	Brazil	Europe	
Volatiles (%)	18.6	17.8	3.9
Ashes (%)	1.4	1.7	5.0
Fixed C (%)	80.0	80.5	91.1

Table 5 – Chemical analyses of the ashes, in percentage of dry mass of reductant.

	Charcoal (%)		Coke (%) Reference
	Brazil	Europe	
NiO		0.0004	
Fe ₂ O ₃	0.03	0.09	0.65
Mn ₂ O ₃		0.02	
TiO ₂	0.003	0.001	0.11
CaO	0.61	1.06	0.03
K ₂ O	0.41	0.15	
Na ₂ O	0.02	0.01	
MgO	0.15	0.20	0.07
Al ₂ O ₃	0.06	0.02	1.17
SiO ₂		0.13	1.67
SUM	1.27	1.69	3.69

SiO₂ was used for both silicon and ferrosilicon smelting. This quartz quality has only minor amounts of impurities (0.27 % Al₂O₃, 0.007 % Fe₂O₃, 0.0015 % TiO₂). The hematite pellets used for ferrosilicon smelting contained 65.2 % Fe.

Grain sizes were in the ranges: 5-30 mm (charcoal), 3-30 mm (quartz), 10-20 mm (pellets) and 3-12 mm (coke reference).

The coke reference is to be considered a normal reactive coke, it obtained a reactivity number of 1200 ml SiO (g). However, the charcoals were much more reactive. They obtained low reactivity numbers, in the range 355-515 ml SiO (g) and are to be considered normal to highly reactive charcoals.

The charcoal from Brazil was used as reductant in the smelting experiment to produce FeSi 75%. The furnace performed well and we obtained a metal yield of 89.3 % as an average for the last 10 hours with stable production. It is remarkable that this is close to what is realized industrially, while it is 7 % higher than the yield obtained in a previous experiment¹⁴ using English gas coke as reductant. The electric power consumption was 10.0 kWh/kg FeSi 75 %, which is about 2 kWh/kg lower than with the English gas coke. However, it is about 2

kWh/kg higher than realized by industry, but higher consumption could be expected due to the relatively high heat loss from a small furnace.

Charcoal from Brazil was also used in silicon production. The results from this experiment are shown in Figure 9.

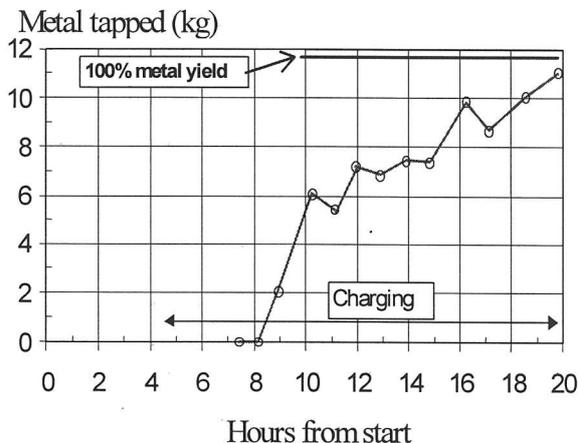


Figure 9 - Production of silicon in a pilot experiment using charcoal from Brazil as reductant.

In this experiment, the furnace performance was satisfactory and we obtained a Si-yield of 84 % as a mean value for the 5 last hours of production while the power consumption was 14.1 kWh/kg metal. The silicon yield is slightly lower than what is normally obtained industrially, although the power consumption is about 2 kWh/kg higher.

The relative high yield obtained with charcoal as reductant can be attributed to the process improvements mentioned in Table 2, especially the better SiO reactivity of charcoals.

To avoid silicon carbide being formed from the start, we initially charged less Fixed Carbon than was needed to reduce all the quartz to silicon. The amount of reductant was increased gradually, and the carbon coverage reached 105 % and 97 % in the ferrosilicon and silicon experiments, respectively.

We also produced silicon metal using charcoal from Europe as reductant. The charcoal was substituted by a standard coke towards the end of the experiment, called the reference coke in Tables 4 and 5. The furnace did not perform satisfactorily in this last experiment due to too high carbon coverage as a result of erroneous Fixed Carbon analyses from the start.

Trace element analyses of the silicon products are shown in Table 6. Silicon contents were 99.0 % and 98.7 % with the respective charcoals from Brazil and Europe as reductants, while it was 95.9 % with the reference coke.

Table 6 – Analyses of trace elements in silicon produced with different reductants.

Element (%)	Charcoal		Coke Reference
	Brazil	Europe	
B	0.003	0.003	0.008
Ni	0.001	0.001	0.004
Fe	0.3	0.5	1.0
Mn	0.09	0.12	0.14
Ti	0.010	0.008	0.025
Ca	0.15	0.31	0.76
K	0.002	0.002	0.018
Na	0.004	0.004	0.032
Mg	0.02	0.02	0.24
Al	0.36	0.33	0.75
P	0.03	0.02	0.02
V	0.001	0.002	0.003

The results indicate a possible low content of trace elements in silicon products using biocarbon as reductant.

Charcoals usually have a low ash content. Major ash constituents are usually CaO, K₂O and MgO, as shown in Table 5. The high content of these oxides in charcoal ashes is not reflected in the product analyses in Table 6. Elements K and Mg may be found in the silica fume, while Ca may have accumulated in the furnace.

7 CONCLUSIONS

The possibilities for the Norwegian ferroalloy industry to reduce their part of the Norwegian fossil CO₂-emissions are studied in a five-year research project. By using biocarbon as a substitute for coal and coke, it is possible to reduce these emissions considerably. As biocarbon is expensive in Norway compared to coal and coke, attention has been focused on lowering costs. One important way of cutting costs is increasing the yield of Fixed Carbon in the carbonization process.

The cost of using biocarbon is one issue. Other aspects are even more important. Thus the

consequences of increased use of biocarbon are being studied, particularly relating to:

- Possibilities of process improvements in the production of Si and some FeSi qualities.
- Possibilities of improving the quality of silicon and silica due to the low ash content in charcoal.
- Reducing emissions of fossil-based CO₂.
- Reducing emissions of SO₂ due to the low sulphur content in charcoal.
- Possibilities of reduced emissions of trace elements due to a low content of trace elements in charcoal.

This research project will be completed in December 2001.

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