Developments of the ULCOS Low CO₂ Blast Furnace Process at the LKAB Experimental BF in Luleå

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Abstract
The ULCOS Blast Furnace process (ULCOS-BF) is one of the developments from the European consortium of steel companies, ULCOS, to minimize CO₂ emissions in blast furnace ironmaking. This process is based on the replacement of hot blast by oxygen, the recycling of hot decarbonated top gas into the lower shaft and normal hearth tuyeres, the capture of CO₂ and its storage in a geological trap (full CCS process). This paper highlights the main features of this ULCOS-BF process, and the expected benefits for CO₂ mitigation. This technology has been demonstrated during two campaigns of 7 weeks by coupling LKAB experimental blast furnace in Lulea to a pilot VPSA unit for CO₂-removal. The concept, preparation and results of the campaigns are described. Following the success of these experiments, studies for the construction of a demonstration unit at ArcelorMittal Florange are currently under progress.

Key Words
ULCOS, Blast Furnace, Top Gas Recycling, CO₂ reduction

Introduction
Since 1896 the concentration of CO₂ in the atmosphere has increased by 25 % and this is widely recognised as a main component influencing global warming through the green house effect [1]. In the traditional processes for producing steel, emission of CO₂ is inevitable, especially for the blast furnace process, which is the main CO₂ producer in an integrated steel works. Since 1950 there has been an enormous research and development effort to make the blast furnace ironmaking technology and operation more efficient. With European collaborative research work (ECSC and RFCS respectively) [2] a coke rate reduction of more than 50% over that period of time was achieved by several improvements like e.g. O₂- enrichment, coal injection, burden distribution, measurement technologies etc.

The carbon consumption at the conventional blast furnace is nowadays approaching the lowest possible thermodynamic values. The usage of reducing agents in an optimised blast furnace is only 5 % above the limit of an “ideal” blast furnace [3]. A further significant reduction of carbon consumption or CO₂ emission will require breakthrough ironmaking technologies. Several technologies have been proposed for further reduction of the fossil carbon usage and the reduction of the CO₂ emission in the blast furnace process itself [4]:
- Recycling of CO from the blast furnace top gas,
- Substitution of CO by H₂ as an reducing agent,
- Usage of Carbon-lean Direct Reduced Iron (DRI),
- Use of Hot Briquetted Iron (HBI) or Low Reduced Iron (LRI),
- Usage of Carbon-lean electrical energy,
- Capturing and Storage of CO₂.

In 2004 the European steel industry launched the ULCOS project: Ultra Low CO₂ Steelmaking [5]. This project is realized by a consortium of the major European steel companies (ArcelorMittal, Tata Steel Europe, ThyssenKrupp Steel Europe, Ilva, Saarstahl - Dillinger Hütte, Rautaruukki Oyj, voestalpine Stahl, SSAB) and LKAB aiming at the research and development of technologies to produce steel with at least 50% lower CO₂ emissions compared to today’s steelmaking benchmark. The cooperative research and development initiative led to numerous ideas to reach this objective. The four most promising solutions have been selected for further exploration: Blast Furnace (ULCOS-BF), Smelting Reduction (HISARNA), Direct Reduction (ULCORED) and Electrolysis (ULCOWIN/ULCOLYSIS).

In the area of the Blast Furnace the technology of applying top gas recycling has been identified as the most promising to operate the blast furnace with very low CO₂ emissions. This is mainly based on the reduction of the usage of fossil carbon (coke) by the re-usage of the reducing agents (CO and H₂) after the
removal of the CO2 from the top gas that leads to lower energy requirements. The usage of pure oxygen instead of hot blast removes the nitrogen from the process, and gives the possibility for underground storage of the removed CO2. In conclusion, the 50% reduction of the CO2 emission from the blast furnace process can be reached by the decrease of fossil carbon consumption and underground storage. Within the ULCOS project the concept of top gas recycling has been experimentally tested at the LKAB’s Experimental Blast Furnace (EBF) in Luleå, Sweden [6,7]. Therefore the EBF was modified and a gas separation plant was built near the EBF. Although the tests at the EBF have been classified as successful, the industrialisation of the ULCOS-BF technology requires an additional scale up step. A demonstration phase at an industrial blast furnace is planned in the 2nd phase of ULCOS (ULCOS II), where the technologies investigated at the EBF will be explored at the industrial scale for demonstration. This paper presents the EBF campaigns results, the scale up to industrial size and the environmental impact.

**History of Recycling of Blast Furnace Top Gas**

The conventional blast furnace is at the moment seen as the thermally most efficient process to produce hot metal with a high productivity. During the last century several new concepts, based on the conventional blast furnace process, have been introduced to reduce the reductant rate and to increase the blast furnace productivity. Already in the 1920’s there was a concept developed to inject hot reducing gas into the blast furnace. Lance [8] calculated that for the metallurgical blast furnace process only 30 % of the coke (at that time 345 kg/thm) would be necessary. The lower gas volume from the tuyeres in the furnace should be compensated by the injection of preheated reducing gas at 1000 °C with 27% CO, 33% H2, and 26% N2 into the lower shaft zone. In the mid 60’s this idea was taken up again in Belgium and in the early 1970’s the first trials were carried out at a 4.6m hearth diameter blast furnace in COCKERILL-Seraing E [9, 10, 11]. A specific amount of 400 Nm³/thm of reformed gas was preheated up to 1000 °C and injected into the lower shaft. A replacement ratio of 0.22 to 0.26 kg of coke per Nm³ of reducing gas was observed. Further investigations were stopped due to economical reasons such as the high price of natural gas. In the late 70’s Fink started to develop a new process which injects cold pure oxygen (Figure 1), fuel and recycled gas at two tuyere levels [12]. Based on the idea of Fink in 1984 Lu developed a process for a conventional blast furnace without a second tuyere row [13]. The main feature of this concept was injection of coal to substitute the coke. Both concepts were never realised and ended as a study. Nearly at the same moment this idea was further used by NKK [14,15] in Japan where a second row of tuyeres was installed in the middle of the shaft. Preheated reducing gas, consisting of recycled top gas without CO2 removal, was injected in these tuyeres. The gas was heated by partial combustion with oxygen. Cold oxygen, coal and cold recycled top gas were injected into the tuyeres of the hearth. NKK tested the process in an experimental blast furnace with 3 tuyeres, an inner volume of 3.9 m³ and a hearth diameter of 0.95 m. The coal injection rate could be increased to 320 kg/tHM, while the coke rate could be reduced to 350 kg/tHM. The lack of energy in the integrated steelworks by recycling the top gas stopped this process, because of the high price of electrical energy and natural gas in Japan.
top gas and recycling of the reducing gas. Three process options were explored in the experimental blast furnace in Luleå. These trials showed a drastically reduced consumption of carbon containing input materials and therefore reduction of CO₂ emissions.

The Design of the Blast Furnace Top Gas Recycling Concept
The ULCOS (top gas recycling) blast furnace project started in 2004. To study the process, a consortium of partners was formed with engineers from the steel companies from the ULCOS consortium, the industrial partners and private research laboratories and a university. Several theoretical studies have been performed to get a better understanding of the new top gas recycling process:
- Mathematical studies with heat and mass balance models and a 3D axi-symmetrical model of the blast furnace [18,19,20] (Figure 2) aimed at calculating the main data and inner condition of the new process in order to select the best operating conditions;
- Internal state of the process was evaluated by metallurgical laboratory tests of the ferrous burden behaviour in terms of reduction, hot degradation, softening and melting and the effect of carbon deposition, under the conditions of the new process regime;
- Mathematical model of the raceway conditions (Figure 3) and gasification tests in both laboratory and pilot scale were accomplished for the design and engineering of the tuyeres under the constraints of simultaneous injection of recycled gas, pure oxygen and pulverized coal;
- A feasibility study at laboratory scale was done to find a proper technology for heating the recycled CO rich gas, taking into account chemical phenomena like C-deposition and metal dusting. Recuperative and regenerative gas heating systems were evaluated and compared with partial combustion of the gas for gas heating;
- A cost evaluation of investment and operational cost of the blast furnace top gas recycle configuration;

The results have been used to set up an experimental programme at the experimental blast furnace (EBF), for which EBF has been modified. Heat and Mass balance calculations and the experience from conventional blast furnace operation have been used to define proper start up and shut down procedures. For the removal of the CO₂ from the top gas a gas separation plant has been designed and erected near the EBF.

The ULCOS Top Gas Recycling Blast Furnace
Heat and mass balance models were combined, with the experience of the conventional blast furnace practice, to calculate several process concepts of the blast furnace with CO₂ free top gas recycling and combustion of coal with pure oxygen at the hearth.

Figure 3: The effect of the position of the shaft injection level on the internal temperature distribution

Figure 2: The temperature distribution in the tuyere and raceway for Version 1, with the oxygen and coal lance
tuyeres. Four flow sheet versions have been defined and examined on the possible reachable C-saving and the feasibility to run the blast furnace under these new concepts. The conclusion was that the so-called Versions 1, 3 and 4 should be able to achieve a fossil carbon saving of 23% or higher with a rather high pulverized coal injection level. Version 2 was rejected because of the low carbon saving and the necessary challenging technology to heat the recycle gas. All versions include CO₂ removal and the injection of CO rich product gas into the hearth, the usage of pure oxygen and the injection of coal together with the reducing gas. In Version 1 and 4, product gas is also injected at a shaft tuyere (Figure 4). The differences are the recycled gas temperature, and the position of the injection points:
- Version 1, the product gas is recycled cold at the hearth tuyeres and hot at the lower shaft;
- Version 3, the product gas is recycled hot at the hearth tuyeres;
- Version 4, the product gas is recycled hot at the hearth tuyeres and hot at the lower shaft;

The temperature of the recycled gas varies from room temperature to 1250 °C. In all cases the gas is heated in a regenerative system. The expected fossil carbon savings are for Version 1 21% at a coal rate of 170 kg/tHM, Version 3 24% at a coal rate of 150 kg/tHM and Version 4 25% at a coal rate of 150 kg/tHM.

The Modification of the Experimental Blast Furnace
In order to bring the theoretical studies together and to get confidence in its feasibility this blast furnace top gas recycling concept was tested at pilot scale during two campaigns of maximum 7 weeks at the LKAB’s experimental blast furnace (EBF), which main characteristics are given in Table 1.

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Working volume</td>
<td>8.2 m³</td>
</tr>
<tr>
<td>Hearth diameter</td>
<td>1.2 m</td>
</tr>
<tr>
<td>Working height</td>
<td>5.9 m</td>
</tr>
<tr>
<td>Hearth tuyeres</td>
<td>3</td>
</tr>
<tr>
<td>Top pressure</td>
<td>1.5 bar</td>
</tr>
<tr>
<td>Pebble bed heaters</td>
<td>2</td>
</tr>
<tr>
<td>Production</td>
<td>35-40 t/day</td>
</tr>
<tr>
<td>Typical tape time</td>
<td>5-10 min</td>
</tr>
</tbody>
</table>

Table 1: Characteristics of the EBF

For implementing the ULCOS-BF process at the pilot facility several modifications on the EBF and its auxiliary facilities were designed, engineered and installed to run the 3 preferred Versions:

- In the vicinity of the EBF a Vacuum Pressure Swing Adsorption (VPSA) gas separation plant for the removal of the CO₂ of the top gas was constructed by Air Liquide. The equipment was tested in classical EBF campaigns before the ULCOS campaigns;
- A safety study was conducted in an extended HAZOP;
- For a safe preheating, transport and injection of the CO rich top gas, improvements were made on the gas heater (pebble bed) and the transport system;
- The hearth tuyeres were redesigned and installed after raceway calculations (Figure 3);

Figure 4: The three different Versions of the ULCOS Blast Furnace process flow sheets
- A shaft gas injection system with 3 shaft tuyeres and a gas distribution system was designed and constructed on the EBF (Figure 2);
- The pulverized coal injection system was adapted to improve the stability of injection at high PC-injection levels.

Table 1: Selected Versions of the ULCOS-BF process could be tested during two campaigns, in 2007 and 2009. The modifications are designed such that both conventional and ULCOS mode is possible. Before each campaign the start up was done in conventional mode and after a sufficient reference period the furnace was switched to the ULCOS mode. In Figure 5 the adaptation of the ULCOS-BF concept at the EBF is given.

The Experimental Blast Furnace Campaigns
To test the whole concept of this new process with blast furnace top gas recycling two campaigns were conducted. The objective of the trials was to demonstrate the operation of the EBF in a complete top gas recycling mode with pure oxygen and PCI at the hearth tuyeres. This has been done under the three defined versions. The ferrous burden consisted of 70% sinter from Rautaruukki Oyj and 30% LKAB pellets. Coke and pulverized coal and flux materials came from SSAB. The hot metal production rate was kept at a constant level of 1.5 t/h and the PCI rate has been varied between 130 and 170 kg/tHM. During the different trial periods the volume of the recycled top gas has been maximized in order to get maximum fossil carbons saving. The EBF results have been compared with model calculations.

Finally after the modification and construction the EBF and its auxiliaries were tested. Thereafter the three selected Versions of the ULCOS-BF process could be tested during two campaigns, in 2007 and 2009. The modifications are designed such that both conventional and ULCOS mode is possible. Before each campaign the start up was done in conventional mode and after a sufficient reference period the furnace was switched to the ULCOS mode. In Figure 5 the adaptation of the ULCOS-BF concept at the EBF is given.

Table 2: Schedule of the first campaign at the EBF, starting on the 24th of September 2007

<table>
<thead>
<tr>
<th>Week 1</th>
<th>Week 2</th>
<th>Week 3</th>
<th>Week 4</th>
<th>Week 5</th>
<th>Week 6</th>
<th>Week 7</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conventional start up</td>
<td>Reference period</td>
<td>Steppage connecting the VPSA</td>
<td>Period of TG recycling Version 3</td>
<td>Steppage Open shaft tuyeres</td>
<td>Period of TG recycling Version 4</td>
<td>Optimization cold rate</td>
</tr>
</tbody>
</table>
Version 3: Recycling of decarbonated top gas into the hearth tuyeres (blast tuyeres) only after heating to 1250°C; Version 4: Recycling of hot decarbonated top gas both at hearth tuyeres 1250°C and in the shaft at 900°C.

Next to regular hot metal and slag sampling, the EBF is equipped with 2 in-burden probes to take in situ measurements of gas composition and temperature and samples from the burden material. For further investigation of the burden material behaviour during this new process, baskets with different sinter and pellet material were charged into the furnace just before the stoppage of the campaign. These baskets were recovered from the EBF during the dissection of the furnace after the quench with nitrogen. Also the cohesive zone region and raceway have been recovered for further examination. The trials were started by one week of normal operation (hot blast) with sinter, first for heating up and stabilization of the furnace, then for establishing a reference for conventional blast furnace operation. After the reference a stop was made for disconnecting the blower and connecting the product gas to the pebble heaters. The start up was made by artificial blast: cold oxygen from the lances and hot nitrogen from the pebble heaters. In progressive steps the nitrogen from the pebble heaters was replaced by decarbonated top gas (product gas) (Figure 7 and 8). This start-up was made according to plan and did not bring any difficulty.

Campaign Results
Under the new process the blast furnace operation was experienced as stable with a smooth burden descent and it was easy to maintain the thermal stability. The gas efficiency in the shaft was stable during the different versions and measurements of the in-burden shaft probes showed a good gas distribution. There have been a few failures of the equipment recorded at which the EBF had to be stopped during the operation under the new process. Some long stoppages required to come back to the coke rate of the “conventional operation” with cold oxygen and hot nitrogen (artificial blast) operation. The small stoppages required only to stop the gas injection and the addition of some extra coke and/or coal. After start up with nitrogen, when there is no top gas available, the product gas could be recycled again within approximately 1 hour. All problems were solved as forecast and the thermal stability of the BF has never been seriously in danger. Along the campaign, the gained experience and increased confidence allowed progressively to make the EBF recovery faster. Each time during change in Version (3 → 4, 3 → 1) the blast furnace was stopped for approximately 8 hours for making the necessary gas connections to the shaft tuyeres and the change of hearth tuyeres. The change over can be clearly seen in Figures 7 and 8.

![Figure 7: K-20 results on reductant rate, recycled gas injection and carbon saving](image1)

![Figure 8: K-23 results on reductant rate, recycled gas injection and carbon saving](image2)

The laboratory experiments showed that the conventional burden material would not be a problem for the new process. The RDI tests showed a similar disintegration for the conventional as well as for the new process. This was confirmed during the campaigns.
No particular process problems were related to burden properties. The results of both probe and excavation samples showed the reduction profile of a centre working furnace (Figure 6): a low reduction level at the wall and a higher level at the centre for both sinter and pellets samples. Tumbling tests on the excavation samples show disintegration behaviour similar to the one of a conventional blast furnace process, which is corresponds to the lab tests. From the burden testing work it could be concluded that the burden properties as used in today’s conventional blast furnace seem no problem for the ULCOS BF process.

The operation of the VPSA has been smooth and nearly without failure. It always provided the required gas quality and the required gas amounts. In general no safety issue has been recorded with the new process. The connection between the EBF and the VPSA worked very well during the trials, but changes in top gas composition from the EBF have an immediate impact on the VPSA operation. Both facilities have to work in a very close relationship.

**Carbon Saving**

In figures 7 and 8 the campaign results are presented. It can be seen that during the trial the coke usage dropped from approximately 530 kg to 400 kg, which represents a considerable carbon saving. In the first campaign the carbon consumption could be reduced by maximum 15% in Version 3 with a top gas recycling ratio of 72%. Version 4 gave a carbon consumption saving of 24% with a top gas recycling ratio of 90%. In terms of coke and coal consumption, up to 123 kg/tHM were saved in the new process (Version 4) compared to the reference operation period.

<table>
<thead>
<tr>
<th></th>
<th>K-20</th>
<th>K-23</th>
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<tbody>
<tr>
<td></td>
<td>Version 3</td>
<td>Version 4</td>
</tr>
<tr>
<td>Carbon saving [%]</td>
<td>15</td>
<td>24</td>
</tr>
<tr>
<td>Top gas recycle ratio [%]</td>
<td>72</td>
<td>90</td>
</tr>
</tbody>
</table>

**Table 3:** Results of EBF campaigns on carbon saving and top gas recycle ratio for the 3 Versions

Due to the relatively short test duration and the fact that this was the first experience with top gas recycling at the EBF, Version 3 was not optimised during the first campaign. It was then tested again during the second 5 weeks campaign to determine the optimum operating conditions and to draw reliable conclusions regarding the maximum carbon savings under this type of operation. The maximum carbon saving reached for Version 3 was 23% and 18% for Version 1, with top gas recycling rates of 87% and 80% respectively. Version 1 could not be fully explored because the trial was stopped due to technical problems at the EBF (table 3). Anyway, the experimental blast furnace campaigns proved that it is possible to run the blast furnace process at a much lower fossil carbon level as today’s blast furnaces do. A carbon saving up to 24% has been proven by the injection of the reducing decarbonated top gas. This would be a significant drop compared to today’s best practice blast furnace process. As a matter of fact, the application of this technology on modern blast furnaces would lead to reduce the carbon consumption from a current level of ~405 kgC/tHM down to ~295 kgC/tHM.

**CO₂ Emission Reduction of the Blast Furnace Process**

The direct carbon savings on the blast furnace process will lead to a direct reduction of the CO₂ emission of 24%. However when these savings are considered on an integrated steelworks, they have to be aligned per tonne of hot rolled coil, as at the moment the blast furnace top gas is fully utilized. Therefore the recycled blast furnace gas needs to be replaced by natural gas. In the ULCOS-project a separate team took into account these considerations. Version 1, 3 and 4 showed a saving of 12%, 14% and 15% CO₂/tHRC respectively. Another technology has to be applied to reach the ultimate objective to save more than 50% CO₂. This can be achieved by application of CO₂ capture and storage technology (CCS). From the first ULCOS campaign there is a CO₂ reduction of 24% in the BF due to gas recycling plus 52% at the VPSA [17] when considering the storage of the removed CO₂.

Maximum 75% reduction of CO₂ emission with respect to the reference period can be identified during the different trial periods of the K-20 campaign (Figure 9).

![Figure 9: Reduction of CO₂ emission in the different periods identified during K-20 (x-axis: trial periods)](image-url)

However at plant level, extra energy (natural gas, electricity) is required to compensate for the decreased amount of blast furnace gas normally used in the plant and for operating the CO₂-removal unit. Anyway, a 60% decrease of the direct CO₂ emission of the integrated steelworks can be achieved. After extraction of the CO₂ from the recycled gas by the (V)PSA, the gas needs to be made storage ready. This will be done by the application of cryogenic techniques.
Scale-up to Industrial
The blast furnace is the core of an integrated steelmaking site, and therefore this new process has to be scaled up to industrial. As the transfer of the EBF ULCOS campaign results is not straightforward, the ULCOS partners decided to move their project into a next phase, ULCOS II, where the new process will be demonstrated on an industrial blast furnace.
In this up-scaling phase specific attention will have to be paid to:
- the tuyere technology for simultaneous injection of product gas (mainly CO and H2) and pulverized coal under full oxygen,
- the technology for shaft gas injection,
- technology for distribution of gas over the radius, especially at the shaft,
- product gas heating and all kind of safety issues.
The corresponding engineering studies are currently under progress at the ArcelorMittal Florange site.

Conclusions
The ULCOS-BF test campaigns performed at the LKAB’s experimental blast furnace that the new top gas recycling blast furnace process is feasible and this under safe conditions as no safety issue was recorded. A considerable step change could be made in the fossil carbon consumption at the blast furnace by the application of the recycling gas injection technology. A CO2-saving of 24% at the blast furnace is possible, and with the application of carbon capture and storage 60% C-saving is feasible regarding emissions per ton of hot rolled coil. The VPSA plant was able to remove the CO2 efficiently from the blast furnace top gas, and up to 90% of the top gas could be treated and recycled. It was possible to operate the blast furnace process and the gas separation plant VPSA in a closed loop. The obtained C-savings were consistent with the predictions from the flow sheet calculations. The tests also indicated that conventional burden material sinter, pellet and coke can be used for the ULCOS blast furnace process.
These EBF campaigns in the ULCOS mode can then be considered as a great success.

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