Utilization of Organic Mixed Biosludge from Pulp and Paper Industries and Green Waste as Carbon Sources in Blast Furnace Hot Metal Production

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Abstract: A six day industrial trial using hydrochar as part of the carbon source for hot metal production was performed in a production blast furnace (BF). The hydrochar came from two types of feedstocks, namely an organic mixed biosludge generated from pulp and paper production and an organic green waste residue. These sludges and residues were upgraded to hydrochar in the form of pellets by using a hydrothermal carbonization (HTC) technology. Then, the hydrochar pellets were pressed into briquettes together with commonly used briquetting material (in-plant fines such as fines from pellets and scraps, dust, etc. generated from the steel plant) and the briquettes were top charged into the blast furnace. In total, 418 tons of hydrochar briquettes were produced. The aim of the trials was to investigate the stability and productivity of the blast furnace during charging of these experimental briquettes. The results show that briquettes containing hydrochar from pulp and paper industries waste and green waste can partially be used for charging in blast furnaces together with conventional briquettes. Most of the technological parameters of the BF process, such as the production rate of hot metal (<1.5% difference between reference days and trial days), amount of dust, fuel rate and amount of injected coal, amount of slag, as well as contents of FeO in slag and %C, %S and %P in the hot metal in the experimental trials were very similar compared to those in the reference periods (two days before and two days after the trials) without using these experimental charge materials. Thus, it was proven that hydrochar derived from various types of organic residues could be used for metallurgical applications. While in this trial campaign only small amounts of hydrochar were used, nevertheless, these positive results support our efforts to perform more in-depth investigations in this direction in the future.

Keywords: hydrochar; pulp and paper industries; blast furnace; organic mixed biosludge; green waste; utilization of wastes; reduce landfill; industrial symbiosis

1. Introduction

Steel production is resource intensive and generates CO2 emissions. In blast furnace (BF) operations, carbon is charged in the form of coke and pulverized coal to provide heat to melt the iron ore and to reduce the iron oxides to iron by a reduction of iron from oxides. The largest part of the CO2 emissions (85%) of the total emissions from the steel industry originates from the coal used to reduce iron ore [1]. Despite the fact that SSAB’s BF’s in Sweden are among the most efficient in the world in terms of minimizing emissions, they account for 10% of the total CO2 emissions in Sweden [2].

The use of renewable biomass resources is increasingly important to reduce greenhouse gas (GHG) emissions and to meet the increasing energy demand. In general, biomass is a biological or organic material, which can act as a source of renewable energy...
through thermal or biochemical transformation processes [3]. Utilization of biomass as a substitute for fossil fuels combustion plays an important role in minimizing methane from landfills and carbon dioxide emissions, which aid in tackling the global warming issue. Sustainable managed biomass resources are considered green, as they do not contribute to net CO₂ emissions or global warming [4]. The release of carbon from biomass combustion is assumed to be balanced by the uptake of carbon when the feedstock is grown and it results in zero net emissions over some period [5,6].

Previous studies have investigated the possibilities to treat maize straw by using the hydrothermal carbonization (HTC) technology for hydrochar production to produce bio coal for injection in a BF [7] and to use torrified sawdust in a BF to reduce the fossil carbon consumption and CO₂ emissions [8]. The main target of using biomass is, obviously, to reduce the formation of CO₂ from fossil fuels (coal, petroleum, gas, etc.) and at the same time upgrade and valorize biomass wastes in an industrial symbiosis instead of keeping them in landfills. For the utilization of biomass as a sustainable renewable energy source, some steps that need to be fulfilled were compiled by Hari Bhakta Sharma et al. (2020); (i) sourcing locally to minimize CO₂ emissions during transportation and extraction of biomass; (ii) making use of local waste such as waste wood, yard trimmings and other organic fraction of municipal solid waste that would have otherwise been sent to landfills; and (iii) utilizing the best energy efficient technologies. Also, Miedema et al. developed Equation (1) to calculate the net renewable power production which compares the total power supply chain energy consumption compared to a conventional reference supply chain based on coal for energy production [9]:

\[ E_{\text{Renewable power}} = \frac{E_{\text{Biomass}} - (E_{\text{Supply chain}} - E_{\text{Reference chain}})}{E_{\text{Supply chain}}} \times 100\% \]  

where \( E_{\text{Biomass}} \) is the energy contained in the biomass, \( E_{\text{Supply chain}} \) is the energy consumption in the whole supply chain, and \( E_{\text{Reference chain}} \) is the energy consumption in the coal reference chain. It is clear that the efficiency of using biomass depends on the energy content of biomass and the energy consumption within the whole supply chain system.

Hydrothermal carbonization (HTC) is a thermochemical process for converting wet biomass into a carbon-enriched solid, commonly referred to as hydrochar [10]. The HTC process is carried out in aqueous conditions at 180–250 °C. The process water can be reduced to 50% by mechanical dewatering and thermal drying can reduce the water content to values below 5%. Mechanical dewatering is much less energy-consuming than thermal drying, since the water does not have to be evaporated [11]. A comparative study of the fuel quality of pellets from raw biomass from woody biomass and agro-residues and corresponding hydrochar was done by Liu et al. [12]. The study concluded that hydrochar pellets were more suitable to be used for power generation than raw biomass pellets. Most of the hydrochar pellets had increased fixed carbon contents, higher heating values, improved combustion characteristics, decreased ash contents and higher mechanical strengths compared to the pellets made of raw biomass.

The pulp and paper industries produce 1.4 million tons organic and inorganic wastes in 2019 in Sweden [13]. Despite the great efforts from the pulp and paper industries to reduce their environmental impact some of the wastes end up in landfill areas, where chemical leaching and GHG emissions can cause significant environmental issues [14]. Therefore, the disposal of industrial paper mill sludge is of a large concern and it has a great importance all over the world [15]. For instance, Europe hosts more than 500,000 landfills of which 80% essentially contain urban solid waste and 20% contain more specific industrial wastes and residues [16]. It should be noted that it is getting more difficult and more expensive to deposit wastes in landfills, due to stronger regulations and requirements with respect to the environment impact. According to the European council directive 1999/31/EC on the landfill of waste, the prevention, recycling and recovery of waste should be encouraged as should the use of recovered materials and energy, so as to safeguard natural resources and to obviate a wasteful use of land [17].
Up to now, the most common methods of sludge disposals are incineration and landfilling [18]. However, since year 2001 Sweden has had a ban on landfills of untreated organic waste [19]. The two dominating strategies for sludge handling at pulp and paper mills in Sweden are: (i) a mechanical dewatering followed by a composting to make a landfill covering material or a soil amendment and (ii) a mechanical dewatering and incineration with landfill application of the ashes [20]. Moreover, since April 2020 Sweden has introduced a tax on incineration of wastes. It is 100 SEK/ton in year 2021 during a transitional period, with an increase in price over the coming years. However, tax exemptions have been introduced for biofuels [21]. The tax is expected to contribute to a more resource efficient waste handling to increase the recycling degree as well as to encourage a fossil-free society [22]. This is an important step to reach Sweden’s goal of net zero GHG emissions in year 2045 [23] and to thereafter reach negative emissions. Negative emissions means that GHG emissions from activities are less than for example the CO₂ absorbed by nature. Negative emissions can also be reached if Sweden helps to reduce the emissions abroad by participating in climate projects [24].

The efforts to reuse, remanufacture and recycle materials are important for the development towards a sustainable future. In previous studies it was found that CaO-rich wastes from pulp and paper mills can successfully be used as slag formers to partly replace primary lime in electric arc furnace during the different stages of steel production [25] and argon oxygen decarburization converter operations [26]. Biosludge is a by-product of a wastewater treatment processes and it can be used as a raw feedstock for production of organic fertilizers and building materials, due to its high contents of organic carbon and plant nutrients [27]. Moreover, the biomass char obtained from biosludge and green waste can partially be used in a BF process as a C-containing reagent instead of using conventional coke. According to previous studies [28,29], a mixture of fine powder coke and iron ore has a much higher reduction reactivity due to a close contact with each other compared to that of conventional layer-by-layer burden in the blast furnace. Also, an increased gasification reaction in the carbon-iron ore mixture can significantly improve the reduction process in the BF and decrease the temperature of the thermal reserve zone [30–32]. Furthermore, it was reported [32] that the reactivity of biomass char obtained from biomass by the carbonization process is much higher than that of coke.

The aims of this full scale production blast furnace trials were to test the possibilities: (i) to utilize the organic mixed biosludge from pulp and paper industries and green wastes as hydrochar pellets after a hydrothermal carbonization in BF process instead of incineration and landfilling; (ii) to use these hydrochar pellets as a part of conventional briquettes containing in-plant fines (such as fines from pellets and scrap, dust, etc.) used for the BF process; (iii) to use briquettes containing hydrochar pellets as a small part of the carbon source in a BF; and iv) to control and compare the technological parameters and conditions of the BF process to a conventional reference charging and experimental charging by using different amounts of various briquettes.

2. Experimental

2.1. Materials

Hydrochar from two types of feedstock were used to replace part of the carbon source in a blast furnace, namely mixed biosludge and green waste. A mixed biosludge is a mixture consisting of biosludge (sedimented bacteria) and pre-sedimented sludge (fiber and sand rejects) in a pulp and paper mill during production processes. Furthermore, green wastes consist of garden residues.

The HTC treated hydrochars were pressed into pellets. Pellets from green waste is a standard material produced by the Ingelia HTC plant. The chemical compositions given in Tables 1 and 2 are typical values from chemical determinations, while the values for mixed biosludge are taken from the specific batch of material used in the given trials. The ash content was analyzed according to the UNE 32004:1984 standard [33], where the
A sample was heated up to 815 °C and kept for one hour. The ash composition was analyzed by using an inductively couple plasma (ICP) method. Also, the volatiles were determined according to the UNE 32019:1984 standard [34], where the samples were heated up to 900 °C for 7 min without a presence of air. The elemental analysis was made by using a Fisons Element Analyzer 1108 CHNS-O. The tests were made by Ingelia on their industrial plant in Nàquera and analysis undertaken by the Institute of Chemical Technology (UPV) and CENER.

Table 1. Main elements of hydrochar pellets, dry and ash free base (daf) in wt%.

| Feed Stock       | C * | H * | N * | S * | Ash Content (%dry) ** | Volatiles (%dry) ** | Fixed C (%dry) ** |
|------------------|-----|-----|-----|-----|-----------------------|---------------------|------------------|
| Mixed biosludge  | 62.0| N/A | N/A | 0.3 | 16.0                  | 65.0                | 20.0             |
| Green waste      | 61.4 ± 1.6 | 6.3 ± 0.3 | 1.3 ± 0.09 | 0.1 ± 0.1 | 10.2 ± 2.3 | 64.7 ± 5.6 | 25.0 ± 5.1 |

* Values referring to dry basis and without ashes. ** values referring to dry basis. N/A: Not Available.

Table 2. Ash composition of hydrochar pellets in wt% of ashes.

| Feed Stock       | Na2O | K2O | MgO | CaO | Al2O3 | Fe2O3 | SiO2 | P2O5 |
|------------------|------|-----|-----|-----|-------|-------|------|------|
| Mixed biosludge  | 1.3  | 5.8 | 1.3 | 12.9 | 10.7  | 2.5   | 52.4 | 4.5  |
| Green waste      | 0.9 ± 0.4 | 4.3 ± 1.7 | 3.9 ± 0.5 | 42.7 ± 4.9 | 6.3 ± 1.0 | 4.6 ± 0.7 | 31.5 ± 1.9 | 4.6 ± 0.7 |

Approximately 4.5 tons of hydrochar from mixed biosludge and 4 tons of hydrochar from green waste in the form of small pellets were produced by the Ingelia HTC plant. Since the hydrochar pellets have a relative low strength, they could not be directly charged into a blast furnace as a separate charging material. Therefore, the hydrochar pellets were briquetted together with the commonly used briquetting materials by SSAB Merox in the following proportions: 2% hydrochar pellets, 12% cement, and 86% in-plant fines. The normally used briquettes contain 8% cement and 92% in-plant fines such as fines from pellets and scraps. In the hydrochar containing briquettes, the amount of cement was increased due to difficulties to get the briquettes to stick together and due to their low mechanical properties. The briquettes containing hydrochar from mixed biosludge from pulp and paper mills are referred to as a paper sludge hydrochar (PSH) and the briquettes containing hydrochar from green waste are referred to as a green waste hydrochar (GrH) in this study. In addition, the produced briquettes were cured for five weeks and stored outside according to the conditions of transportation and storage used in the steel plants. Typical photographs of samples of organic sludge, green waste, hydrochar pellets and bio-briquettes used for industrial trials are shown in Figure 1.

Figure 1. (a) mixed biosludge; (b) green waste; (c) hydrochar pellets produced at Ingelia; and (d) bio-briquettes containing hydrochar produced by SSAB Merox.
2.2. Method

The bio-briquettes were top charged into the blast furnace (BF) together with conventional briquettes. The first three days of the experimental trial campaign, the briquette charge contained 10% of PSH briquettes of the total amount of charged conventional briquettes (PSH-10 trial) during the first day, 20% (PSH-20 trial) during the second day, and 40% (PSH-40 trial) during the third day. On the fourth day, 10% of green waste hydrochar briquettes were charged (GrH-10 trial), 20% on the fifth day (GrH-20 trial), and 22% on the sixth day (GrH-22 trial). Moreover, two days before (Ref-1 and Ref-2) and two days after the hydrochar campaign (Ref-3 and Ref-4) were selected as the reference periods to represent a conventional production to enable a comparison of technological parameters. The total charge of briquettes in BF (containing low manganese briquettes, high manganese briquettes and briquettes containing hydrochar) was 301.35 ton/day. The hydrochar briquettes were added instead of a part of the briquettes having low Mn contents.

Samples of raw material, dust and sludge were collected during the reference and experimental trials. The charging rate and chemical composition determinations of all charged burden and injected materials were collected to evaluate and compare the results from the different trials. Moreover, the production rate of hot metal, slag, dust and sludge, as well as the chemical composition of top gas and heat losses were recorded and collected during the experiments.

3. Results and Discussion

It is well known that the strength of the briquettes used in BF is important, both at room temperature and at high temperatures. The main quality index in the iron and steel making industry is the cold strength of the briquette, the tumbler handling strength (TTH) [35]. After the curing period, a determination of the TTH values were made according to the ISO 3271 standard [36]. The results showed that the values were 45% for PSH briquettes and 63.5% for GrH briquettes. These TTH values were considered to be too low, as they should be at least 75% for this specific BF (the requirements for TTH strength may differ for different BF). However, the values were deemed to be acceptable for short term industrial trials. Moreover, it was noted that some fine particles were generated from the hydrochar briquettes due to a low strength. Therefore, the hydrochar briquettes were sieved to separate the fines before charging the material into the BF. The results showed that PSH and GrH generated 5% and 20% fines, respectively. It is apparent that a long storage time outdoor caused the TTH values to become worse compared to when using a short storage time. These fines influence a stable BF operation, since it leads to problems with the gas cleaning system and a higher counter pressure when the fines clog the porous bed in the furnace. Therefore, the strength of the briquette needs to be improved by an optimization of the recipe and briquetting process in order to carry out successful longer trials in the future.

The average values obtained for the production rate, fuel rate, amount of injected pulverized coal (PCI), amount of dust, hot metal temperature, amount of slag, basicity (CaO/SiO$_2$) and ratio of sulphur contents in slag/hot metal from the trial campaign and reference periods are compiled in Table 3.
Table 3. A compilation of the main production parameters during the trial campaign and the reference periods.

| Test Day | BF Productivity (ton HM/day) | Fuel Rate (kg/tHM) | PCI (kg/tHM) | Dust (kg/tHM) | Temp.(HM) (°C) | Slag (kg/tHM) | CaO/SiO₂ (%S)/[%S] |
|----------|-----------------------------|-------------------|--------------|--------------|----------------|---------------|------------------|
| Ref-1    | 2845.0                      | 477.2             | 103.6        | 20           | 1463           | 168.4         | 0.89             | 15.1           |
| Ref-2    | 2909.2                      | 470.6             | 99.2         | 13           | 1474           | 168.4         | 0.92             | 17.8           |
| PSH-10   | 2913.6                      | 472.8             | 96.4         | 18           | 1472           | 169.2         | 0.91             | 17.6           |
| PSH-20   | 2842.2                      | 472.5             | 101.3        | 16           | 1469           | 168.4         | 0.88             | 15.2           |
| PSH-40   | 2894.9                      | 473.2             | 100.0        | 14           | 1469           | 168.4         | 0.89             | 14.6           |
| GrH-10   | 2860.7                      | 475.3             | 101.0        | 15           | 1454           | 169.9         | 0.88             | 13.2           |
| GrH-20   | 2855.9                      | 480.2             | 99.6         | 12           | 1457           | 171.2         | 0.89             | 13.3           |
| GrH-22   | 2811.9                      | 488.0             | 104.8        | 15           | 1458           | 173.6         | 0.91             | 12.6           |
| Ref-3    | 2835.7                      | 483.2             | 100.1        | 12           | 1473           | 173.5         | 0.88             | 16.6           |
| Ref-4    | 2787.0                      | 488.7             | 102.4        | 20           | 1474           | 173.5         | 0.94             | 20.3           |

In Figure 2a, it can be seen that the production rate of hot metal (in ton of hot metal per day) was larger in the PSH trials (2883.6 tHM/day) and almost the same during the GrH trials (2842.8 tHM/day) compared to that in the reference trials (2844.2 tHM/day on average). However, it should be pointed out that the production rate can differ by about 200 tHM/day without changing any process parameters during a conventional production. The average amount of produced dust was similar during the PSH trials (16.0 kg/tHM) and lower in the GrH trials (14.0 kg/tHM) in comparison to that in the reference trials (16.25 kg/tHM), as can be seen in Figure 2b.

The fuel rates (Figure 3a) in the PSH trials (on average 472.8 kg/tHM) were very similar as those obtained in the Ref-1 and Ref-2 trials, while they were higher in the GrH trials (up to 488.0 kg/tHM in GrH-22) and in the Ref-3 and Ref-4 trials. However, the highest fuel consumption (488.7 kg/tHM) was obtained during the last reference day (Ref-4). Also, the amount of PCI (Figure 3b) was lower in the PSH trials (99.2 kg/tHM) and almost the same as in the GrH trials (101.8 kg/tHM) in comparison to the amounts in the reference trials (101.3 kg/tHM on average).
The average carbon contents in the hot metal and the average temperatures of hot metal obtained during reference and experimental trials are presented in Figure 4. It can be seen that the carbon contents in the hot metal was slightly lower in the PSH and GrH trials (on average ~4.48% and 4.44%, respectively) compared to the reference trials (4.51%), but it fluctuated significantly during the reference periods as well. The temperatures when using PSH briquettes were almost the same (1470 °C) as in the reference periods (1471 °C on average). However, when using hydrochar from green waste (GrH trials), the temperatures were significantly lower (~1456 °C). Therefore, it was necessary to add coke to increase the hot metal temperature.
The slag amount was increased due to the addition of coke to increase the hot metal temperature. It can also be seen that the basicities of the slags in all trials fluctuated significantly during the studied period, including during the reference trials. This could have been due to a high moisture content of the top charged limestone during the trials, due to the high humidity in the atmosphere. The higher moisture content in the limestone affected the charging system, so that less limestone was actually added to the BF. This, in turn, resulted in a lower CaO content in the slag and large fluctuations of the slag basicity. Despite that, the average values of the slag basicities during the experimental trials (~0.89 in the PSH and GrH trials) were smaller than the values in the Ref trials (~0.91). Moreover, the lower basicity of the obtained slags in the experimental trials might also be explained by the relatively high concentrations of SiO$_2$ in the hydrochar pellets, which were used for preparation of the experimental PSH and GrH briquettes, as given in Table 2.

![Figure 5](image.png)

**Figure 5.** (a) Average calculated slag amount; and (b) basicity of slag defined as the CaO/SiO$_2$ ratio obtained during reference and experimental trials.

The average contents of sulfur in hot metal, slag and dust during all trials are given in Figure 6. Moreover, the coefficients of the S distribution between the slag and hot metal ($K_s = (%S)/(%S)$) for all trials are given in Figure 6d. It was found (Figure 6a) that the S content in the hot metal increased with an increased amount of added PSH and GrH briquettes. More specifically, the average S contents in the hot metal increased from 0.072% in the reference trials up to 0.076% and 0.088% in the PSH and GrH trials, respectively. The largest concentration of S (0.094%) was obtained in the GrH-22 trial. This may be explained by the lower basicities in most of the experimental trials, as can be seen in Figure 5b. At the same time, the average S contents in the slag (Figure 6b) decreased from 1.24% in the reference trials to 1.20 and 1.15% in the PSH and GrH trials, respectively. As a result, the $K_s$ value decreased significantly from ~17.4 in the Ref trials to 15.8 and 13.0 in the PSH and GrH trials, respectively, as shown in Figure 6d. This can possibly be explained by the lower basicities of slags in the experimental trials compared to the reference trials, which was caused by relative high concentrations of SiO$_2$ in both types of hydrochar pellets (PSH and GrH) used for preparation of experimental briquettes. Another explanation could be the increased amount of charged coke (containing Si), due to the use of wet charge materials. In addition, the charged lime was also wet which lead to a decreased sulphur refining. It should also be pointed out that the S contents in dust varied in all trials (from 0.32% to 0.42%), as can be seen in Figure 6c. However, the average value of the S contents in the dust were very similar: 0.36% in the Ref trials 0.35%, and 0.37% in the PSH and GrH trials, respectively.
Figure 6. Average contents of sulphur (%) in (a) hot metal; (b) in slag; (c) in dust; and (d) ratio of sulphur in slag/hot metal obtained during reference and experimental trials.

According to the results shown in Figure 7a, the average P contents in the hot metal are quite stable and almost the same (~0.029) in all trials. Furthermore, they were found to be independent on the type and amount of experimental briquettes added during the blast furnace operation. Also, though the FeO content in slags showed a tendency to decrease during the trial period (Figure 7b), the average values of the FeO content in the slags were also very similar for all trials, namely 0.51% in the reference periods and 0.52% and 0.49% in the PSH and GrH trials, respectively.
The economic and environmental effects of using hydrochar briquettes in BF operations can be determined in a reliable manner if the blast furnace operation is stable. As was mentioned above, an advantage of using bio-briquettes is the higher reactivity of bio carbon compared to conventional coke. It can be explained by the larger gasification rate of a biomass char and the larger reduction rate of the iron oxides when using bio-briquettes. It can be expected that an enhancement of the reactivity of the burden in a BF can decrease the reducing agent by decreasing the ratio of CO and CO₂ in the top gas as well as the thermal reserve zone temperature [32]. However, it was difficult to evaluate the economical and environmental effects of using different bio-briquettes in the BF during these short-term industrial trials, due to the low strength of the tested bio-briquettes. Therefore, more investigations and industrial trials are needed in the future to optimize the properties of bio-briquettes containing hydrochar pellets as well as the blast furnace process, when charging bio-briquettes containing hydrochar pellets at the top of the blast furnace. Moreover, one thing that has to be pointed out is the unpleasant smell of the hydrochar during the briquette making, which could be an issue that needs to be considered and solved when working with these types of material.

4. Conclusions

Two types of hydrochar, obtained from a mixed biosludge from a pulp and paper mill and from green waste, were used to replace a small part of the carbon source in bio-briquettes used in a blast furnace (BF) during a 6-day industrial trial campaign. The main aim with these initial industrial trials was to study if there were any negative effects on the production rate or hot metal quality by using the different amount of bio-briquettes containing various biomass materials. The main results can be summarized as follows:

- Briquettes containing wastes from pulp and paper industries (such as a mixed paper sludge hydrochar (PSH) and a green waste hydrochar (GrH)) can partially be charged in a BF together with conventional briquettes during short term industrial trials.
- Most of technological parameters of the BF process in the experimental PSH and GrH trials were very similar compared to the values in the reference trials. For example, the differences between the average values of the reference days and the average values for the trial days were the following: (i) <1.5% with respect to the production rate of hot metal, fuel rate and amount of slag, (ii) 1.5–14% for the amount of dust, (iii) ~2% for the amount of injected pulverized coal, (iv) <3.5% for the FeO content in the slag, (v) 1.5% for the C content in the hot metal, (vi) 6–23% for the S content in the hot metal.
metal and (vii) <1.5% for the P content in the hot metal. Moreover, no noticeable influence on the blast furnace performance could be seen during the experimental trials in comparison to the reference trials.

- Overall, the addition of PSH briquettes show better operational results compared to the GrH briquettes.
- The strength of the briquettes needs to be improved before future long–time trials are carried out.

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