

Sponge Iron with the New Black: Production and Use of Bio-coal in a Direct Reduced Iron (DRI) Plant

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ABSTRACT

Höganäs AB produces sponge iron using a packed bed DRI process where coke and coal act as a reducing agent. This study summarizes the R&D efforts to replace fossil-based reducing agents with biomass-based “new black” (bio-coal). To build both practical and fundamental knowledge in evaluating the feasibility of fossil coal replacement with bio-coal, a holistic investigation of the entire bio-coal value chain have been carried out. It starts from the material performance needed for practical utilization of bio-coal in the existing DRI plant, on to assessing strategies for the conversion of biomass into bio-coal that give required qualities, and consequently to the evaluation of available biomass feedstock resources based on defined techno-economic criteria for bio-coal production and use.

KEYWORDS

Bio-coal, direct reduced iron, slag formation, Höganäs process, slow pyrolysis, resource analyses

INTRODUCTION

Steel industry contribute ca.7% of the global greenhouse gas (GHG) emission, which is around 2.8 billion tons [1]. Various industrial initiatives around the globe are trying to find a way to break away from the industry’s dependence to fossil-based fuels and raw materials, namely coal. One example is the use of H₂ as reducing agent, e.g., H₂-based direct reduced iron (DRI) and H₂ injection in blast furnaces [2]. Another example is the use of biomass-based fuels and reducing agents. Mandova et al. showed [3] that 42% of GHG emission from fossil fuels and reducing agents used in steel plants within EU can be reduced by replacing with biomass-based fuels and reducing agents. Among biomass-based raw materials, bio-coal is the most promising candidate to replace fossil coal in steelmaking. Norgate et al. [4] showed that 57% of on-site carbon footprint of integrated steel plants can be reduced by substituting fossil coal with bio-coal. Besides the primary advantage to lower GHG emission, bio-coal produced from stem wood assortments also contain lower ash, sulfur, and phosphorus compared with pulverized coal, which would reduce slag ratio up to 50% in the process [5]. Nevertheless, the lack of mechanical strength in bio-coal limits its application to replace top-feed coke in large-capacity blast furnaces [6]. Consequently, it has been concluded by

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Suopajarvi et al. [7] that bio-coal injection to the blast furnaces through tuyere is the largest potential to replace fossil coal.

Höganäs AB produces sponge iron for a niche metal powder market using a packed bed DRI process where coke and coal act as a reducing agent (the so-called Höganäs process) [8]. In the Höganäs process, reduction of iron progresses in a packed bed of solid reducing agent and iron ore without intermixing with each other. The replacement of solid reducing agent with other forms (i.e., gas and/or liquid) would demand an overhaul of this established industrial process and the development efforts may become too costly. Therefore, one of the most promising ways to reduce GHG emissions from this process is to replace fossil-based reducing agents with bio-coal. During the Höganäs process, sponge iron can be obtained from series of chemical reactions:



This paper summarizes the effort to build both practical and fundamental knowledge in evaluating the feasibility of fossil coal replacement with bio-coal. First, several bio-coal samples, produced from various raw materials in several industrial pyrolysis reactors, were evaluated to assess the consequence of their use in the Höganäs process. Furthermore, the in-process behavior of the ash forming matter in bio-coal was evaluated by thermochemical equilibrium calculation. Then, a series of laboratory-scale experiments was carried out to find an optimal way to produce bio-coal with desired properties at high conversion yields. Finally, the availability of promising suitable biomasses was screened, with focus on forest residues.

EVALUATION OF BIOCOAL CHARACTERISTICS

In this section, physical and chemical properties of several bio-coal samples from industrial pyrolysis reactors were analysed and compared with typical coal samples used in the steel industry.

Bio-coal Samples

Four bio-coal samples were collected from several industrial-scale pyrolysis plants. Bio-coal A represents bio-coal from fast pyrolysis. Bio-coal B is from a modern converter-type pyrolysis reactor at intermediate heating rate. Bio-coal C is from a conventional slow pyrolysis process. Bio-coal D is from an innovative two-step pyrolysis process. Some bio-coals were also densified by an extrusion process currently under development.

Bio-coal A is a bottom-ash residue from a cyclone-type biomass gasifier [9], operated with spruce powder. The temperature inside the reactor had a variation from 700 °C at the bottom to 1200 °C near the fuel injection point. Due to a short particle residence time of ca. 10 s, bottom-ash residues contain relatively high carbon content. An estimated heating rate of the biomass powder during pyrolysis is in the order of 10^3 – 10^4 °C/s. Bio-coal B was obtained from an indirectly-heated auger type pyrolysis reactor, where birch chips (10-30 mm in size) were pyrolyzed at maximum temperature of 600 °C. The reactor is non-isothermal and it had a temperature profile similar to a counter-current heat exchanger. The residence time of the particles was ca. 60 minutes, and the estimated heating rate 10–20 °C/min. Bio-coal C is a commercial charcoal produced for barbeque purposes. There is no information available for the production conditions. However, typical conditions for industrial production of charcoal is

referred here [10]: carbonization in retorts at a temperature of around 500 °C and a residence time of 8 to 40 hours. Bio-coal D was produced from two-step pyrolysis methods: pyrolysis at 350-400 °C in a rotary kiln, followed by a heat treatment at 1100 °C in a retort [11].

Characterization Methods

Sample preparation. Each sample was prepared according to DIN 51701-3. Initially, each sample was homogenized, and a portion of the sample (± 300 g) representing the original sample was taken by quart-sampling. Sample was pre-dried in an oven at 40 °C for at least 12 hours. Bulk density was measured after pre-drying. Each pre-dried sample was then divided evenly into eight sub-samples using Retsch PT100 rotary sample divider repeatedly until the amount of representative samples reached the requirement for analysis methods.

Densities. Bulk density was measured according to analogue VDLUFA-Method A 13.2.1. Approximately 300 ml of homogenized sample was filled into a graduated cylinder (500:5.0 ml). The mass of the sample was then weighed, and the volume of the sample was measured after 10 times of compression by means of falling (tapping). To measure envelope density, the mass and volume of around 15 ml of Al₂O₃ were first measured in a measurement cylinder (25:0.5 ml) after tapping. Approximately 3-5 ml of sample was then added into Al₂O₃, and the final mass and volume were measured. Envelope density was later calculated as the ratio of mass difference to volume increase. True density was measured using Micromeritics AccuPyc II 1340 gas displacement pycnometry system with helium as the medium.

Reactivity of bio-coal with CO₂. The reaction rate of bio-coal sample with CO₂ was measured with a thermogravimetric analyser, TGA8000 coupled with gas mixing device GMD8000 from PerkinElmer Inc. Bio-coal was ground to particle size below 75 μm and ca. 0.5-1.2 mg was spread at the bottom of an alumina crucible to minimize the influence of mass diffusion. Gas flow rate was 50 mL min⁻¹ in the standard state. Temperature was raised from 30 °C to reaction temperatures at 10 °C min⁻¹ under N₂. After it stabilized at the reaction temperature, the gas composition was changed to 20% of CO₂ in N₂ (volume basis). The sample was held until the mass was stabilized. The experimental data was normalized to calculate the conversion (X) in the following equation.

$$X = \frac{m_0 - m}{m_0 - m_{ash}} \quad (4)$$

Here, m_0 is the initial mass, m_{ash} is the final mass, and m is the sample mass during gasification. Finally, the reactivity of bio-coal was evaluated by initial value of the conversion rate, which is defined by the time derivative of conversion.

Results and Discussion

Density is one of the most important properties because it directly determines the mass of carbon that can be packed into a sagger. The density can be defined by three different ways, namely, bulk density (mass divided by the volume of packed bed), particle/envelope density (mass divided by the volume of particle envelope), and true/skeleton density (mass divided by the volume of solid excluding internal pores). Figure 1 compares the densities of different bio-coals with those of fossil-based materials. Bulk densities of bio-coals were significantly lower than the ranges of coal. It showed a significant improvement upon densification, but the values remained lower than the ranges for coal samples. The difference can be explained by the envelope velocity, implying that the bio-coal has much higher internal porosity inside

each particle even after the densification. There was relatively smaller difference in skeleton density, albeit there seem to be a weak correlation with pyrolysis temperature. It is reasonable as high pyrolysis temperature results in the orderly aligned carbon structure (so-called annealing) [12].

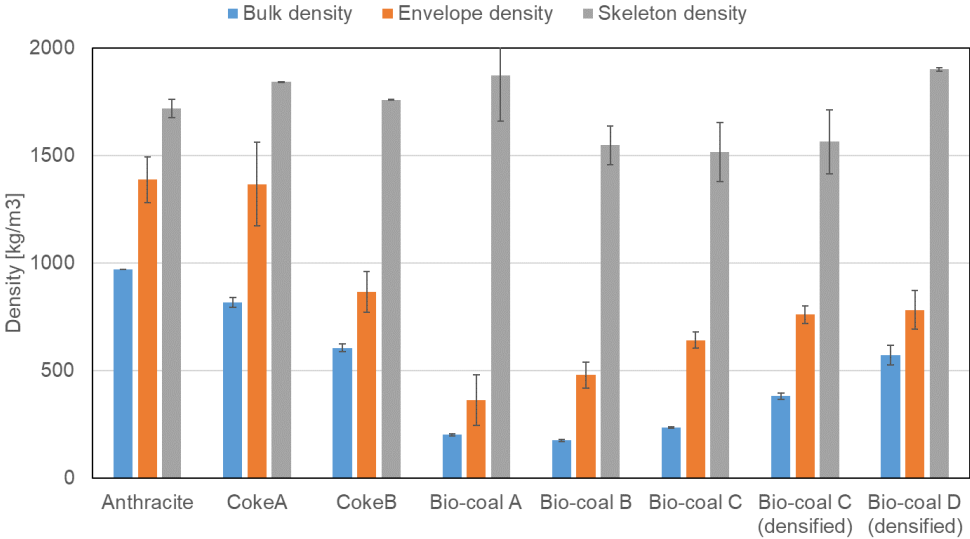


Figure 1. Comparisons of the densities

The Boudouard reaction, reaction (1), is a critical reaction for the iron ore reduction as it provides the reducing gas, CO. Therefore, the reactivity of bio-coal is one of the most important properties. Figure 2 shows the comparison of the reactivity in the form of Arrhenius plot. The results show that the reactivity of bio-coal slightly differs from each other. Bio-coal that has been pyrolyzed at lower temperature and shorter residence time had relatively high reactivity. However, the difference is notably smaller than the comparison with fossil-coal. Hence, it would be a major challenge to control the reaction rate of the Boudouard reaction when replacing fossil material with bio-coal. Together with the density of bio-coal, this result highlights the challenges in replacing fossil-coal with bio-coal.

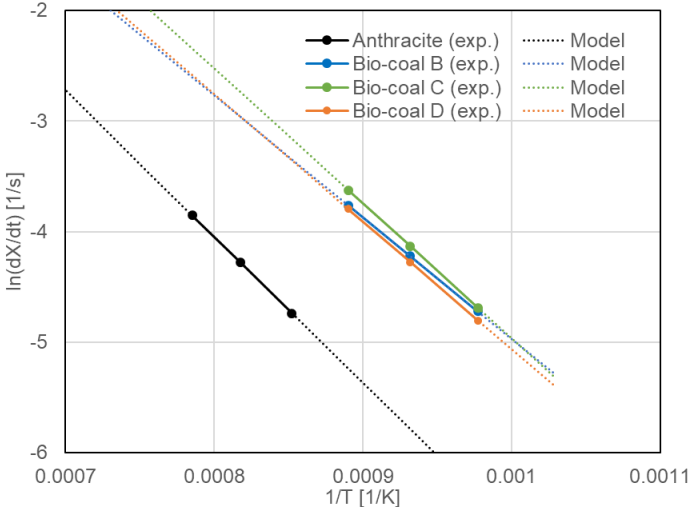


Figure 2. Comparisons of the reactivity of charcoal

BEHAVIOR OF ASH FORMING ELEMENTS IN THE DRI PROCESS

The amount and composition of the ash forming matter in the reducing agents play a significant role in the iron ore reducing process. The ash content and presence of critical inorganic elements such as K and P can strongly affect the melting behavior and ash-iron interactions, i.e., the purity of produced metallic iron. As bio-coal potentially has high K and P contents, investigating the behavior of inorganic elements in reduction mixture and possible interaction between ash matters and iron/iron ore are of immense importance. This section describes ash criteria for replacing fossil-based reducing agent through thermodynamic equilibrium calculations (TECs).

Methods

Thermodynamic Equilibrium Calculations (TECs) were used to predict the behavior of inorganic elements in the reduction mixture (melt formation, chemical composition and viscosity of melt, condensed phases, release of alkali). TECs were also applied to predict the interaction between the ash from bio-coal and iron/iron ore. TECs were performed using the Equilib module of FactSage 8. The databases used were a combination of FactPS (for gas and stoichiometric solids) and FToxid together with FTsalt databases. Viscosity of melt was calculated by viscosity module in FactSage 8. It should be pointed out that this module does not consider the effect of P on the viscosity value.

The following raw biomass were selected to represent a wide range of bio-coal with various fuel ash composition; 1) stemwood rich in Ca-K-Si, 2) softwood bark rich in Ca-K, 3) tops and branches from softwood, i.e., forest residues, rich in Ca-Si-K, and 4) wheat straw rich in Si-K. To estimate elemental analysis of bio-coal from typical elemental compositions of raw biomass [13], the following assumptions were considered [7,14–16]: 1) the yield of bio-coal after pyrolysis is 35%, 2) 90% of Cl and S are released from the raw biomass, 3) 5% of K, Na, and P are released from the raw biomass, and 4) no release of refractory elements, i.e., Ca, Si, Mg, Al and Fe from the raw biomass.

TEC was carried out at 1000 and 1200 °C. The following five reduction mixtures were investigated: (1) fossil-based reference reduction mixture, (2) bio-coal from stemwood, (3) bio-coal from bark, (4) bio-coal from forest residues (tops and branches), and (5) bio-coal from wheat straw. All the reduction mixtures are mixed with lime.

Results and Discussion

Alkali release. Figure 3 shows the predicted release of alkali to the gas phase from each reduction mixture at 1200 °C and 1000 °C. Compared to fossil coal, bio-coal usually contained very low amount of Na compared to K, therefore in Figure 3 the main release of alkali (g) is related to release of mainly elemental K and to some extent K compounds or to the gas phase. Regardless of type of reducing agents, at higher temperature, higher alkali release was predicted. The highest release of K was predicted for wheat straw-based reduction mixtures since the concentration of K is very high in the bio-coal. Reduction mixtures containing char from bark and forest residues showed higher release of K than fossil-based reduction reference mixtures where Si content was high and K captured by silicate phases or silicate melt.

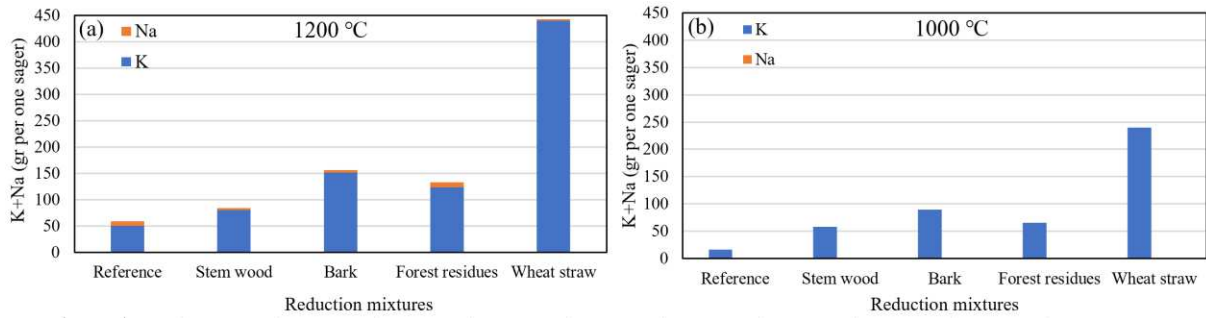


Figure 3. Predicted K+Na release (gas phase) from the sagger at (a) 1200 °C, (b) 1000 °C.

Condensed phases. Results from TECs showed that ashes from fossil-based reduction mixtures and wheat straw-based reduction mixture were dominated by silicate phases due to the high Si content. Oxides dominated ashes from stemwood, bark and forest residue-based reduction mixtures, mainly CaO due to the higher Ca concentrations. Figures 4 and 5 show the predicted amount of melt in one sagger and viscosity of the melt, respectively. The lowest amount of melt at 1000 °C and the highest amount of melt at 1200 °C were predicted for fossil-based reduction mixtures. At 1200 °C, viscosity of predicted melt for fossil-based reduction mixtures is approximately five times higher than bio-coal based reduction mixtures. This is due to the higher content of Si and Al and lower content of K. Reduction mixtures based on forest and agricultural residues (i.e., bark, forest residues, and wheat straw) showed higher amounts of melt at 1000 °C but lower at 1200 °C, compared to the fossil-based reduction mixture references. Bio-coal forms a substantial amount of low-viscosity melt for a wide range of temperatures. Hence, risks related to melt flows should be assessed upon 100% replacement of reduction mixture with bio-coal.

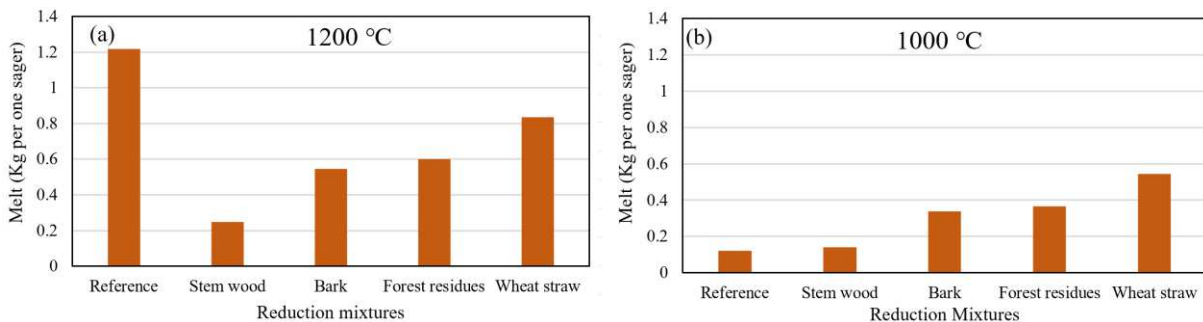


Figure 4 Predicted melt formation (a) 1200 °C, (b) 1000 °C.

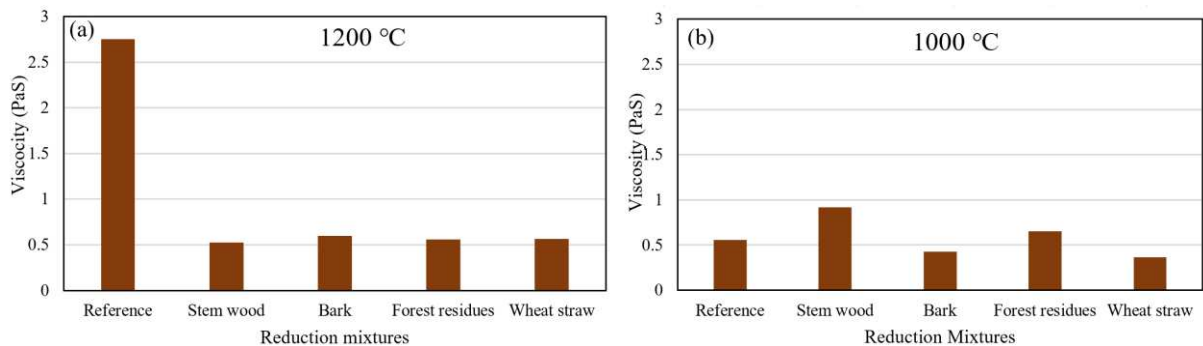


Figure 5. Viscosity of the melt predicted to form (a) 1200 °C, (b) 1000 °C.

BIO-COAL PRODUCTION METHODS

The previous sections showed some critical differences in bio-coal as a substitute of fossil coal, namely, density, reactivity, and ash chemistry. In order to improve the properties of bio-coal, laboratory experiments were carried out to improve bio-coal production methods with ash leaching.

Methods

Three different types of biomass were used, namely pine bark, pine forest residue, and corncob. They are abundant forest by-products and agricultural wastes in many countries with high content of Ca, K, and Si. Sample size was above 4 mm and the sample was dried at 105 °C prior to the pyrolysis experiments. A part of sample was leached by acetic acid to reduce inorganic content.

Bio-coal samples were produced by using an externally heated cylindrical reactor. A wire mesh basket was used to hold the sample. The reactor was purged with N₂, supplied from the bottom of the reactor. After preheating the reactor to 700 °C, ca. 5-10 g of biomass was introduced to the reactor. The biomass was pyrolyzed for 5 minutes to complete the reactions while reducing the effects of thermal annealing. The bio-coal was then cooled down under inert atmosphere.

Results and Discussion

Ash content. Alkali metals, especially K, are prone to be released into gas phase during the DRI process as previously shown in Figure 3. In addition, Ca and K are known to have catalytic activities for the Boudouard reactions. Figure 6 shows the contents of these ash forming elements in both biomass and bio-coal samples. Acid leaching showed a significant effect to reduce K content in biomass while the effects on Ca content remained relatively moderate. Meanwhile, both Ca and K contents increased significantly upon pyrolysis as seen from high values of bio-coals. The results agreed with the previous work from Hedayati et al. [15] that showed that more than 85% of K and P remained in bio-coal. As the bio-coal yields from pyrolysis were between 15-30%, the relative concentration increased significantly. Hence, there is a need to leach ash forming elements from ash-rich biomass if the pyrolysis temperature is relatively low (500-900 °C).

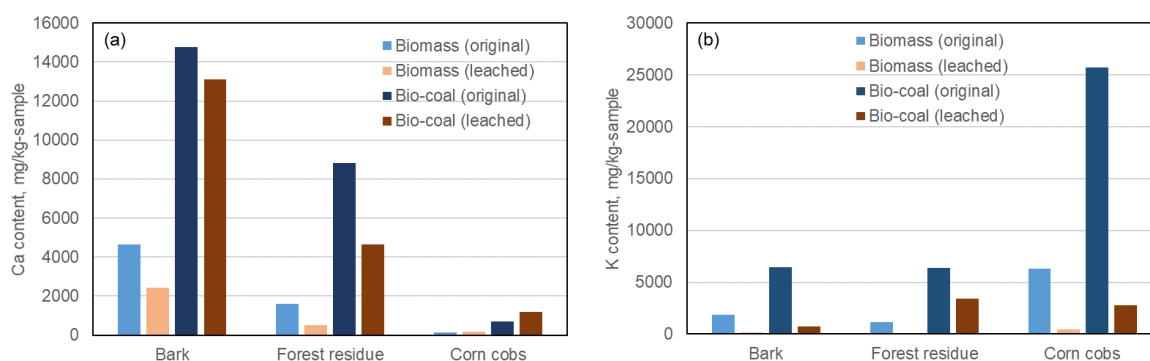


Figure 6. Ash contents of raw biomass and bio-coal produced at 700 °C in N₂; (a) Ca (b) K.

Reactivity of bio-coal. As previously shown in Figure 2, the reactivity of bio-coal is much higher than that of coal while the difference among bio-coals (from stemwood and bark) was shown to be relatively small. However, the reactivity can vary significantly among bio-coals from ash-rich biomasses due to the catalytic activity of alkali metals [17]. The change in ash contents upon acid leaching inevitably affects the reactivity of bio-coal as well. Figure 7 shows the reactivity of bio-

coal from three different biomasses with and without acid leaching. The reactivity decreased significantly upon leaching for all three types of biomass. A loose correlation with K contents in Figure 6(b) can be discerned, although other ash forming elements may also affect the results. However, this correlation seems to be valid only for bio-coal from biomass without leaching, and the correlation diminished after the leaching.

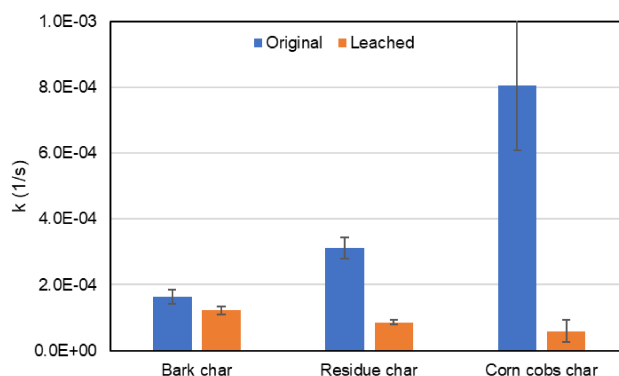


Figure 7. Reactivity of bio-coal produced at 700 °C in N₂. Measured at 800 °C in 20% of CO₂ and 80% of N₂.

AVAILABILITY OF BIOMASS RESOURCES

The previous sections have shown that the type of biomass used as bio-coal raw material has an impact on the ultimate bio-coal properties when used in the reduction mixture. This section provides a short summary of availability of suitable domestic biomass resources, based on the current state of knowledge regarding use of forest biomass for energy purposes.

Biomass Supply Potentials

Several previous studies have been conducted to estimate biomass potentials for different time perspectives. Table 1 summarises the potential increase in the supply of different lignocellulosic biomass types until 2030 and 2050, respectively, as provided by Börjesson [18]. Börjesson concludes that overall, the supply of domestic biomass in the Swedish energy system could increase by ~35% to 2030, and by ~50% by 2050, compared to current levels.

Table 1. Potential for increased supply of biomass suitable as bio-coal raw material in Sweden compared to the use in 2020 [18], considering technical, economic and ecological restrictions.

Biomass raw material	Increased supply potential (TWh/y)	
	2030	2050
<i>Forest biomass assortments</i>		
Logging residues (branches and tops) ^a	16-18	18-21
Roundwood (damaged and thin trees)	5-7	6-8
Forest industry by-products	6-12	10-16
<i>Agricultural assortments</i>		
Straw ^a	2-4	2-3
Brushwood	5-8	8-10
Fast-growing deciduous trees ^b	–	2-4

^a Ecological and techno-economic limitations.

^b On downgraded arable land.

Logging residues from forestry operations, mainly in the form of branches and tops (“*grot*”), constitute the single biggest source for increased biomass supply. There are no publicly

available statistics on actual extraction and utilisation of logging residues. Nevertheless, an estimate can be made from the share of all harvested areas where extraction of residues (any volumes) has been reported [19], as shown in Figure 8. In northern Sweden, logging residue extraction is reported for very low shares of the total harvested area, while in southern Sweden the harvesting potential can be assumed to be close to fully utilised.

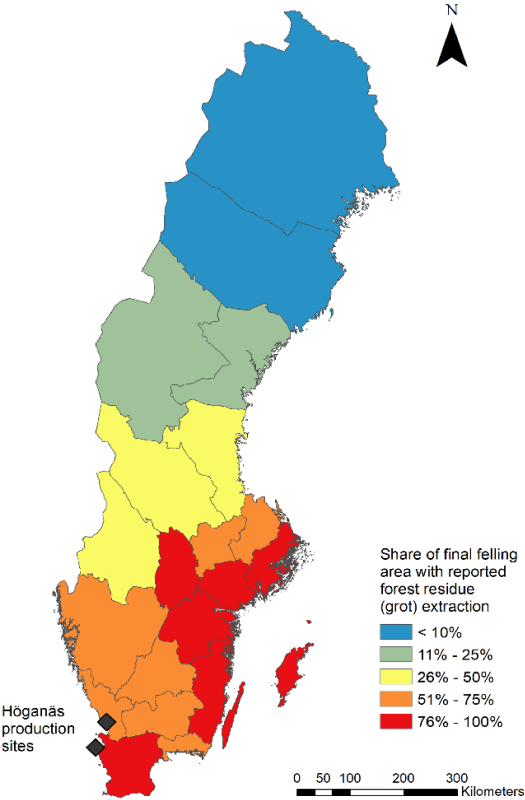


Figure 8. Share of final felling area where residue extraction has been reported, on county level for Sweden. Average 2016-2020. Source: Swedish Forest Agency statistics [19].

Sustainability Aspects

In order to ensure the sustainability of biomass used to replace fossil energy and materials, several policies are in place, e.g. the EU Renewable Energy Directive (RED II), which includes criteria to ensure that solid biomass is produced sustainably, irrespective of its geographical origin. National legislation govern how logging residues are allowed to be extracted in Sweden, e.g. from which types of land, which quantities, and whether ash must be recirculated [20]. Recent research has assessed that extraction of logging residues from approximately 50% of the clear-cut areas can be done without unacceptable impacts on critical environmental objectives regarding, e.g., biodiversity and acidification, in particular if ash recirculation is also increased [21–23].

Cost and Competition Aspects

The actual supply potential of biomass, the market potential, is strongly dependent on current and future demand from different sectors and willingness to pay depending on, e.g., different policies. The Swedish biomass demand is expected to more than double until 2045, based on published roadmaps for a fossil-free Sweden [24]. With an increased demand for biomass, the prices increase, which creates incentives to produce more raw material in existing production systems. The price of relevant biomass types today is comparable to that of fossil gas, but higher than that of fossil coal. In order to be cost competitive, a price on CO₂ emissions (e.g.

through a tax or emission allowances) is thus required. In absolute terms, the cost of wood chips for the user ranges from ca 18 to 20 EUR/MWh [25] when sourced from forest. The cost can be lower for industrial by-products and higher for new assortments (such as brushwood or logging residues from thinning operations), due to the need for technology development.

CONCLUSION

This study highlights the key technical and economic issues to replace fossil-based reducing agents with bio-coal as summarized below.

- Bio-coal and fossil coal have similar true densities, but the particle and bulk densities of bio-coal are significantly lower than fossil coal. The development of effective densification is a key development area.
- Intrinsic reactivity of bio-coal produced from different reactors (and reaction conditions) is relatively similar to each other. However, the intrinsic reactivity is significantly higher than that of fossil coal, and the difference needs to be mitigated by increased mass diffusion resistance.
- Thermodynamic equilibrium calculation predicted that forest-based bio-coal reduction mixtures have much lower melt formation with lower viscosity than coal-based reduction mixture at 1200 °C. Higher release of alkali was also predicted both at 1000 °C and 1200 °C.
- For the reduction mixture with bio-coal from wheat straw, considerable amount of melt, which was in the same range as the reference mixture, was predicted, but with lower viscosity at 1200 °C. Higher release of alkali was also predicted at 1000 °C and 1200 °C.
- Acid leaching resulted in profound reduction of K content in both biomass and bio-coal while the majority of Ca remained after leaching. Virtually, there was no release of ash forming element during pyrolysis, which resulted in significantly higher ash content.
- Reactivity of bio-coal decreased significantly by acid leaching. The difference in the reactivity of bio-coal between the different raw biomass types seems to be related to the K content before leaching. However, this is not the case after acid leaching.
- From a market point of view, flexibility in feedstock sourcing (both regarding quality and geographic origin) and suitable supply chain options will be important in order to be able to use residual biomasses as bio-coal feedstock.
- Incentives (e.g. policies) needed to mobilise necessary biomass resources, in particular for biomass sources that require both new technical and new market structures.

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