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Increasing the sustainability of steel production in the electric arc furnace by substituting fossil coal with biochar agglomerates

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Abstract

Biochar fines from a wood gasification plant and from pyrolysis of agricultural residues were investigated as substitutes for fossil coal used in the steel production in the electric arc furnace. During previous tests biochar fines with high specific surface showed problematic burn-off behaviour. Therefore the agglomeration behaviour of the biochar fines was investigated. Different binary and ternary mixtures of biochar with water and binders were tested in a hydraulic stamp press and evaluated with regard to green strength and fatigue strength of the briquettes after three days. One selected mixture was used to produce pillow briquettes in a double roll press. The abrasion behaviour of the produced briquettes was tested and compared to an anthracite reference coal. Melting tests in a pilot electric arc furnace showed that the agglomerated biochar reacts similar to the reference coal. The briquetting leads to reduced reactivity and slower burn-off compared to the biochar fines.

Keywords

biochar, agglomerates, steelmaking, electric arc furnace

List of symbols

BC	biochar

- BCB biochar briquette
- EAF electric arc furnace
- PVA polyvinyl alcohol
- RC reference coal

1 Introduction

In addition to the blast furnace – converter route, which is using the primary raw materials iron ore and coke, the electric steelmaking in the electric arc furnace (EAF) mainly based on scrap is the second most important steel production process. The global share of electric steelmaking in terms of crude steel production in 2013 was about 27.5 %.¹ Steelmaking in the modern EAF covers its energy demand to up to 40 % by using chemical energy from fossil fuels. Apart from natural gas used in burners, coal is used as chemical energy source. The coal is charged into the furnace as lump coal, mainly anthracite, in the basket or pulverized by lance or injector as a slag foaming agent. The average specific consumption of coal in electric arc furnaces is around 12 kg per ton of produced steel.² Using a coal with high carbon content as an example, this generates approximately 43 kg of carbon dioxide emissions per ton of steel. These emissions from the use of fossil carbon make up 40 to 70 percent of the total direct emissions of the EAF of 60-100 kg CO₂/t_{steel} (Ref. 3). The different carbon sources and related direct emissions can be seen in Fig. 1. Based on the 2013 amounts of EAF crude steel production of 66.3 million tons in the EU27¹ the use of fossil coal in the EAF accounts for about 2.9 million tons of direct CO₂ emissions.



Fig. 1: Direct CO₂ emissions of a typical EAF

Therefore the partial introduction of biomass in a form of biochar in the steel production process is considered as a viable option to reduce the greenhouse gas emissions and hence the environmental impact of the process.^{4,5} Moreover in times of increasing prices for fossil energy sources like coal and natural gas as well as increasing prices for electricity and CO₂ emission certificates, it can be an economical option, to utilize charcoal from biogenic residues, which are classified as CO₂-neutral.

Therefore, especially since the last decade, there have been several research efforts in this area. Results of this research were, among others, that biochars are suitable for partial substitution of fossil fuels in the silicon manganese (SiMn) production⁶ and in the general manganese alloy production.⁷ In the production of SiMn it has been observed that with the use of charcoal a higher metal yield can be obtained than using fossil coke. The metal produced with charcoal shows significant lower level of silicon content. Furthermore, the use of charcoal increased the slag formation. This fact was also observed by Yunos et al.⁸ They substituted fossil coke in laboratory scale with degassed palm kernel shells and examined the differences in the reactions with the slag. It was found that the palm kernel shells have a more intensive and longer lasting gas generation. Based on their results they draw very positive conclusions for the partial substitution of coke with palm kernel shells on an industrial scale. Griessacher and Antrekowitsch⁹ showed the possibility of targeted production of biochar from biomass with specific reaction behaviour. They used pyrolytic processes at various temperatures and analysed the reactivity of the carbonized product. They came to the expected result that with increasing carbonization temperature, the yield of the carbonized product decreases and its carbon content increases. This differs depending on the starting material by a factor of up to 3.5 in the percentage mass difference. Thus, it is in principle possible to selectively prepare biochars with a predetermined reaction behaviour. A known reaction behaviour is crucial for the use of biochar in metallurgical processes. Therefore, the results of Griessacher and Antrekowitsch point to an untapped potential of various biogenic

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carbonisates in the metallurgical industry. In addition, they pointed out the potential of using the by-products of the pyrolysis itself. These provide even more ecological benefits of using biogenic carbonisates. Regarding the agglomeration of biogenic carbonisates Quicker et al.¹⁰ could demonstrate the possibility of agglomeration of various biomass carbonisates in laboratory scale. The combination of molasses as a binder and alumina cement as a support grain achieved the highest strength and is comparable to metallurgical coke.

Previous investigations by the authors^{11,12} using the dusty, carbonaceous residue of biomass gasification demonstrated that the use of biochar in the EAF is in general feasible. The results of these first investigations on the use of biochar instead of fossil coal in the EAF steelmaking process showed that there are no significant negative impacts on the steel or slag composition. Problems occurred regarding the handling of fine-grained carbonaceous materials with a high specific surface in the presence of liquid steel and oxygen. Previous tests showed the direct combustion of the highly reactive biochar fines right after arc ignition in the pilot EAF operated by the Department for Industrial Furnaces and Heat Engineering.

On the basis of these results, the use of biochar fines charged with the scrap in the bucket of an industrial scale EAF as a substitute for lumpy coal seemed not to be feasible without further processing of the biochar fines. The chosen approach to solve the problem was to carry out trials to estimate the briquetting behaviour of the biochar. The aim was to produce compact agglomerates with a lower reactivity than the biochar fines, more similar to the reaction behaviour of commonly used lumpy coal. That way the replacement of fossil coal with biochar fines would be possible. The use of such biomass based carbon sources can decisively increase the sustainability of EAF steelmaking. In addition, the use of carbonaceous residues from biomass gasification could result in economic advantages in terms of raw material prices and the reduction of necessary CO₂ certificates.

2 Material and methods

Two different types of biochar have been used for these investigations. On the one hand a residual from a wood gasification plant (BC1) and on the other hand a product from pyrolysis of agricultural residuals (BC2), which are shown in Fig. 1. Previous tests of the raw material have demonstrated an unwanted fast burning in the pilot EAF and also a high risk of uncontrolled burn-off in industrial EAF trials.^{11,12}



Fig. 1: Residual from wood gasification BC1 (left) and biochar fines from pyrolysis process BC2 (right)

To evaluate the properties of the biochar briquettes it is important to compare them with conventional fossil coals used in the electric arc furnace. An anthracite coal, which is commonly used in German steel plants, was chosen as a benchmark for the compacts. The reference coal (RC) has an average grain size of 20 mm, which is similar to the average grain size of the agglomerates that can be achieved by the briquetting. Therefore, it can be expected that due to the grain size no major differences occur during the use in the EAF. The proximate and the elemental analysis of the biochars and the reference coal are given in Table 1. As binders water, polyvinyl alcohol (PVA) and molasses have been tested.

Table 1: Proximate and elemental analysis of the input materials

Content	Unit	RC	BC1	BC2
moisture	wt-%	3,3	2,3	11,9
ash	wt-%	5,35	29,6	22,2

volatile matter	wt-%	1,8	9,8	11,9
C _{fix}	wt-%	89,55	58,3	54,0
С	wt-%	88,4	64,7	57,8
S	wt-%	1,12	0,38	0,09
Р	g/kg	0,13	3,86	2,65

2.1 Briquetting Tests

The briquetting tests have been conducted at the Unit of Technology of Fuels (TEER) at RWTH Aachen University. The mixtures of biochar, water and binders have been prepared using a plow mixer. The mixing time was five minutes. In a first step agglomeration tests with a sample quantity of 50 g were performed using a hydraulic stamp press with a pressure of 100 MPa, a pressing time of 20 seconds and a cylindrical press mould and appropriate extrusion die (diameter 50 mm). All mixtures were tested using the stamp press to produce single briquettes. If a stable briquette could be produced from the mixture, three briquettes per mixture were produced and tested on a hydraulic testing press to determine green strength. Additionally their fatigue strength was tested after three days and preliminary drop tests from 2 m on a flat surface have been conducted.

The mixtures investigated include binary mixtures of biochar with water, biochar with PVA and biochar with molasses as well as ternary mixtures of biochar, water and PVA and biochar, water and molasses. The ternary mixtures investigated are shown in Table 2.

Water [%]	Binder [%]	10	15	17	20	24	30
5		$BC1^+$	-	$BC1^+$	-	$BC1^+$	$BC1^+$
10		$BC1^+$	-	$BC1^+$	-	$BC1^+$	BC1 ⁺ / BC2 ⁺
15		$BC1^+$	-	$BC1^+$	-	$BC1^+$	BC1 ⁺ */ BC2 ⁺ *
20		BC1 ⁺	BC1 ⁺ */ BC2 ^{+*}	BC1 ⁺	BC1 ⁺ / BC2 ⁺	BC1 ⁺	BC1 ⁺ / BC2 ⁺

Table 2: Ternary mixtures of biochar, water and binder

+ molasses, * PVA

In a second step a selected mixture was continuously briquetted in a double roll press to produce 30 kg of biochar pillow briquettes for further investigations. Using the double roll

press the water content of the mixture was varied again to verify the best mixture for briquetting the biochar and to determine the influence of this parameter on the continuous briquetting. The briquettes were stored for one week after manufacturing to increase the strength of the single compacts.

Since first pilot tests in the EAF showed that especially the small grain size of the original material can lead to major problems, tests regarding the abrasion resistance of the briquettes in comparison to the abrasion resistance of the reference coal were a main aim of the conducted investigations. For this purpose 1 kg of the produced biochar briquettes and the reference coal were subject to drop tests. The drop tests were performed according to ASTM D440-07 "Standard Test Method of Drop Shatter Test for Coal".¹³ These drop tests comprise the samples being overthrown in a 2 m long tube with a diameter of 180 mm. This procedure is repeated four times. After that the samples are screened at 6.3, 10 and 16 mm. The share screened smaller than 10 mm was classified as fine material.

Drop resistance and friability could be determined by the drop tests. Besides these points it is important to know the transportability of the compacts to secure long-distance transports by truck or train. Therefore the biochar briquettes were placed on a box sieve to simulate their resistance against vibrations. In order to compare the briquette's physical behaviour with the reference coal's behaviour, 1 kg of each material was treated with a box sieve. The samples were strained through vibration with 1400 rpm for 60 sec to determine the materials' abrasion strength (fraction of fines).

2.2 Pilot trials using biochar briquettes in the pilot scale EAF

In addition to the tests of the physical properties of the briquettes, melting trials in a pilot electric arc furnace have been conducted with the briquetted biochar as well as the reference coal. For these trials the respective coal was charged together with scrap and slag formers into the EAFs crucible. For each trial 50 kg of scrap and slag former were charged. The amount of

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briquetted biochar (BC1) and reference coal were normalized with regard to 1 kg of elemental carbon content, leading to 1.79 kg of biochar briquettes (55.9 wt-% C) and 1.13 kg of reference coal (88.4 wt-% C) being charged into the crucible. The trial includes meltdown of the scrap in the EAF till tapping temperature of about 1650°C is reached. During the heat especially the off-gas composition was analysed and recorded. After tapping and cooling down, samples of steel and slag have been taken for further analysis.

3 Results and Discussion

3.1 Results of the Briquetting Tests of BC1

First binary mixtures of BC1 with water, PVA and molasses have been investigated in the hydraulic stamp press. The use of water as binder leads to partially agglomerated briquettes at best. So water alone is no adequate binder for the biochar studied. The tests with molasses as binder showed that an addition of 30 % of molasses lead to the production of complete briquettes. The preliminary drop tests showed that the green briquette can withstand the first drop with strong deformation and breaks up on the second drop. Measurement of the green strength showed insufficient results. With 30 % PVA as binder again a complete briquette could be produced in the stamp press, but the compacts left the pressing tool with major deformations. Based on this, further tests with binary mixtures were not considered to be expedient.

So the briquetting tests in the hydraulic stamp press showed that binary mixtures of biochar with water, PVA or molasses did not lead to sufficient results. Even with binder additions of up to 30 % no satisfactory briquette production was possible. In all tests the briquettes did not have an adequate green strength.

Therefore in a subsequent step, ternary mixtures of biochar, water and a binder were tested. The mixtures showed improved agglomeration behaviour. To compare both binders,

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briquettes were prepared with 30 % binder content and 15 % water content. The green strength of the molasses mixture was 0.87 MPa and in comparison to this value, the green strength of the PVA mixture was 0.69 MPa. A further advantage of the molasses mixture is the better press out behaviour. The briquettes left the pressing tool without any major damage. Fig. 2 shows the compacts of both mixtures in comparison.



Fig. 2: Briquettes consisting of biochar, water and molasses (left) and PVA (right) After molasses has been identified in combination with water as a good binder, various molasses-water-mixtures were tested to determine an optimal mixture for technical tests. The water content of the mixtures was varied between 5 % and 20 % and the molasses content was adjusted to values between 10 % and 30 %. The green strength of each of the briquettes was measured directly and is given in Table 3. The table shows that at lower water concentrations no stable briquettes could be produced. For the other mixtures, the strengths are in the range of 0.69 MPa and 1.73 MPa. The highest green strength could be achieved with a composition of 24 % molasses, 15 % water and 61 % biochar.

Table 3: Green	strength of	of biochar.	water and	molasses	mixtures
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in MDa		Molasses				
in MPa		10%	17%	24%	30%	
Watar	15%	0.78	1.27	1.73	0.93	
water	20%	0.69	1.17	1.30	n.b.p.	

n.b.p.: no briquetting possible

The pressure resistance of three mixtures with high green strength was also determined after three days again to define the shelf life of the compacts. The chosen blends had following compositions: 17 % molasses and 15 % water; 17 % molasses and 20 % water; 24 % molasses and 20 % water. Because of the high binder content the mixture with 24 % molasses and 20 % water was not selected. Fig. 3 illustrates for all three mixtures the green strength and the fatigue strength after three days. It can be observed that the strength increased for all three mixtures after three days of storage. This fact can be explained by the hardening of molasses. The best fatigue resistance could be achieved with a value of 6.13 MPa by the addition of 24 % molasses and 15 % water to the biochar. Based on the results of the green strength and the fatigue resistance this ratio of the binder and water was chosen for the continuous production of biochar pillow briquettes.



■ Green strength ■ Fatigue resistance

Fig. 3: Green strength and fatigue resistance of selected mixtures

Using the double roll press high quality compacts could be produced at a maximum briquetting force of 10 kN. At higher forces the material began to extrude laterally through the roll nip. Similar effects were observed by an increase in the addition of the water content. At about 17.5 % water content in the mixture the material was flowing back into the screw instead of entering the roll nip. At lower water contents a briquetting on the double roll press

was no longer possible. Therefore, these experiments confirmed that the composition which was already determined in the test in the hydraulic stamp press is well suited for continuous briquetting.

3.2 Results of the Briquetting Tests of BC2

Similar to the briquetting tests of BC1, first mixtures of BC2 with water, molasses and PVA have been investigated in the hydraulic stamp press. For the binary mixtures BC2 with molasses and even BC2 and PVA it was only possible to produce a stable briquette with a minimum share of 30 % of binder. Further results from the briquetting tests of ternary mixtures (see table 3) were also not satisfactory regarding the press-out behaviour, green strength and fatigue resistance after drying.

Because of the required amount of more than 30 % of binder to produce stable briquettes out of binary mixtures and the results from ternary tests, BC1 was chosen to produce pillow briquettes for the pilot EAF trials. The use of binary mixtures of BC2 and more than 30 % of binder is economically not feasible. The ternary mixtures showed no acceptable press out behaviour and did not reach the minimum of required green strength

3.2.1 Properties of BC1 pillow briquettes compared to reference coal

The biochar briquettes have a similar grain size as the reference coal as can be seen on Fig. 3.



Fig. 4: Biochar briquette (BCB) and piece of reference coal

The carbon content of the dry biochar briquettes has been reduced to 55.9 % compared to the 64.7 % of the original biochar. This is due to the mixing of the biochar with the molasses which dilutes the carbon content of the mixture. After storage the biochar briquettes showed significantly lower dust emissions, improved transportability and better dosability than the original biogenic feedstock.

The comparison of the drop test results for the two samples is shown in Fig. 5. The share of fine material is even lower in comparison to the reference coal.



Fig. 5: Grain size distribution after drop test

The results of the vibrating sieve tests show also that the BC1 briquettes behave comparable to the reference material (Fig. 6). So by briquetting the problems experienced with the fine original material can be solved.



Fig. 6: Grain size distribution after 60 s on vibration sieve

3.3 Results of the EAF pilot trials using biochar briquettes

Regarding steel and slag composition the use of the biochar briquettes showed similar results as the tests with biochar fines previously described.^{11,12} The biochar briquettes have no negative impact on steel and slag chemistry. Like the previous trials using the biochar fines the steel analysis regarding the carbon content shows that the carburization of the melt by the reference coal is higher than using the agglomerates. Also phosphor and sulphur show the same behaviour as in the previous trials and there is no discernible negative influence on the steel composition from the higher phosphorus content of the agglomerates in comparison to the phosphorus content of the used anthracite coal (cf. Table 1). The results of the slag analysis in general show that there is no negative impact detectable by using the biochar briquettes.

Main differences in the use of biochar briquettes compared to biochar fines and reference coal can be seen when examining the off-gas composition measured over the course of the heat in the pilot EAF.

Fig. 7 shows the off-gas composition when the reference coal is used. Due to the lower reactivity of the reference coal the reaction of oxygen with the carbon carrier starts at a later point of time (~ 600 s after power on) and extends over a time period of about 2100 s with a constant CO formation level of about 4 to 5 vol.-%. In difference to this reference trial Fig. 8

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shows that the CO formation in the previously reported trial using the biochar fines^{11,12} starts directly after power on. With hardly any time delay after arc ignition it comes to a rapid combustion of the biochar fines that have been charged with the scrap. The majority of the CO gas is generated in a short time period with a very high formation rate. These first results showed that unprocessed biocoal fines are not favourable to charge with the scrap in an electric arc furnace. Due to the large specific surface area of the fines the reactivity is too high for charging the fine material without a prior processing step.

In the evaluation of the off-gas composition of trials with biochar briquettes (Fig. 9) it was found that the use of briquetted biochar has a positive influence on the combustion behaviour. In comparison to Fig. 8 the direct CO peak is missing and the reaction of oxygen and biochar starts at a later point in time. Generally it can be determined that the combustion behaviour of the briquettes is closer to the combustion behaviour of the reference coal (Fig. 7), except the higher CO formation rate.



Fig. 7: Off-gas composition using reference coal







Fig. 9: Off-gas composition using BC1 briquettes

4 Conclusions

In order to solve the problems encountered, when trying to substitute fossil coal with finegrained biochar, the agglomeration of this biochar has been investigated. After testing a number of binary and ternary mixtures of biochar with water and binder in a hydraulic stamp press, a selected mixture was used to produce biochar pillow briquettes in technical scale on a double roll press. It was possible to create stable briquettes from a fully biogenic blend with characteristics which are comparable to the characteristics of an anthracite reference coal that is presently used in electric steel mills.

These biogenic briquettes have been used as charge carbon in pilot EAF melting trials. The evaluation of the results of these trials regarding steel, slag and off-gas composition showed no negative impact on the slag and steel chemistry. Concerning the off-gas composition it could be shown that the combustion behaviour of the briquettes is close to the combustion behaviour of the anthracite reference coal.

On the basis of the results presented in this paper the problems encountered trying to use biochar fines can be solved by briquetting of the biochar with water and a biogenic binder. Therefore biochar shows a great potential for the use in industrial scale EAF steelmaking. However, the use of the biochar briquettes should be tested in an industrial steel mill to prove

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the suitability in industrial scale. Moreover, the briquetting of more types of biochar fines from different production processes should be investigated to diversify the possible input materials for the substitution of fossil carbon sources in electric steelmaking.

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