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BIOMASS USE IN IRONMAKING: OPTIONS AND LIMITS[1]

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Abstract

In short terms, only few options to mitigate the CO_2 emissions can be introduced in the steel industry both from economical and technical points of view. Use of biomass is one of such options. In this contribution, firstly biomass sources, resources, availability, processing steps and products as well as quality requirements are discussed controversially. Afterwards three options for the use of biomass in ironmaking are presented: injection into shaft furnaces, incorporation into the burden materials or coal blends or reducing gas generation. Two first ways (direct injection and charge of biomass containing burden into the blast furnace) are discussed more detailed primarily based on recently completed and running at the IEHK, RWTH Aachen University projects. Injection behaviour and relevant characteristics of charcoals produced from oak, olive and eucalyptus at carbonisation temperature in the range of 360-560°C were compared with fossil coals typically used as PC. Then torrefied materials produced from pine and beech wood chips at varying temperatures in the range of 200-350°C were examined. Next, an approach for lowering the thermal reserve zone temperature by using high reactive carbonaceous materials and operation of large blast furnaces vs. mini blast furnaces are discussed. Composite pellets with cold embedded charcoal were investigated with this background. Reduction and volume change behaviour as well as and strength of these materials were examined. Pellets without reductant and with embedded coal were investigated as reference materials.

Key words: Charcoal; Torrefied biomass; Injection; Self-reducing pellets and composites; Mini blast furnace .

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1 INTRODUCTION

Mitigation of the CO_2 emissions affecting global warming is the main reason why 300 years after the introduction of coke instead of charcoal, the steel industry looks for the biomass use. Theoretically there are many possibilities to reduce or even to avoid the CO_2 emissions in iron- and steelmaking but only few of them can be realised in short and medium terms both from technical and economical points of view. Biomass use is one of these possibilities although its efficient and sustainable application depends on the world region (e.g. Brazil and Australia are more suitable than Europe) and further factors discussed in the next chapter.

Biomass can be mainly used in three ways in iron- and steelmaking [1]:

- by gasification to generate gas for reduction or heating
- by injection into blast furnace, Corex or electric arc furnace
- by incorporation into coal blend for cokemakig, composites, self-reducing pellets, fuel for sintering etc.

Many studies on biomass gasification for syngas generation in fluidised bed using pilot High-Temperature-Winkler (HTW) gasifier were performed at the IEHK, RWTH Aachen University under supervision of Prof. Gudenau; grass, straw, wheat, miscanthus, corn and further biomass materials were investigated along with coals, lignite and organic waste [2,3]. Recently the HTW gasifier was modernised and equipped with a new control system for gas flow and temperature measurement. From the collected data, a prediction for the behaviour of carbonaceous materials (coals, biomass etc.) for industrial gasification processes is possible [4].

This contribution deals with two further ways: biomass injection into the blast furnace (BF) via tuyeres and use of self-reducing pellets with embedded biomass materials.

In the past, injection of charcoal fines was studied [5] and then applied [6] in small charcoal blast furnaces in Brazil. Recent studies based on experiments and modelling were focused of charcoal injection into modern coke blast furnaces [7-12] Mixtures of coal with charcoal [7,8] or charcoal with sugarcane bagasse [13] were studied as well. Current research program, partly presented in this contribution, aims at injection of torrefied biomass (TB). These materials are produced by lower temperatures compared to charcoal (below 350°C).

Use of self-reducing pellets (SRP) and composites with cold embedded carbonaceous materials can avoid or hinder swelling of these burden materials in DR processes and in BF ironmaking and improve their reduction behaviour [14,15]. Further target of usage of agglomerates and composites with embedded high reactive carbonaceous materials is the possibility for decrease of carbon consumption in the BF by means of transition of FeO-Fe reduction equilibrium to lower temperature affecting decrease of thermal reserve zone (TRZ) temperature or the starting temperature of solution loss reaction ($CO_2 + C = CO$), see **Fig. 1**. This shift would improve the CO-gas utilisation efficiency, resulting in lower reducing agent consumption [16,17]. It has to be stressed that carbon saving at lower reserve zone temperature while using highly reactive materials can only be realised at certain conditions [18] that consider e.g. strength of such materials. Current study, partly presented in this contribution, aims at use of SRP in the mini blast furnace. Furthermore, carburisation behaviour of iron ore-carbon composites with different biomass types has recently been investigated in the scope of cooperation between the IEHK, RWTH Aachen University and the Kyushu University. The focus of this research was on effects of carbon crystallinity and ash content in charcoal on carbon dissolution in molten iron and carburisation reaction in the composites [19].

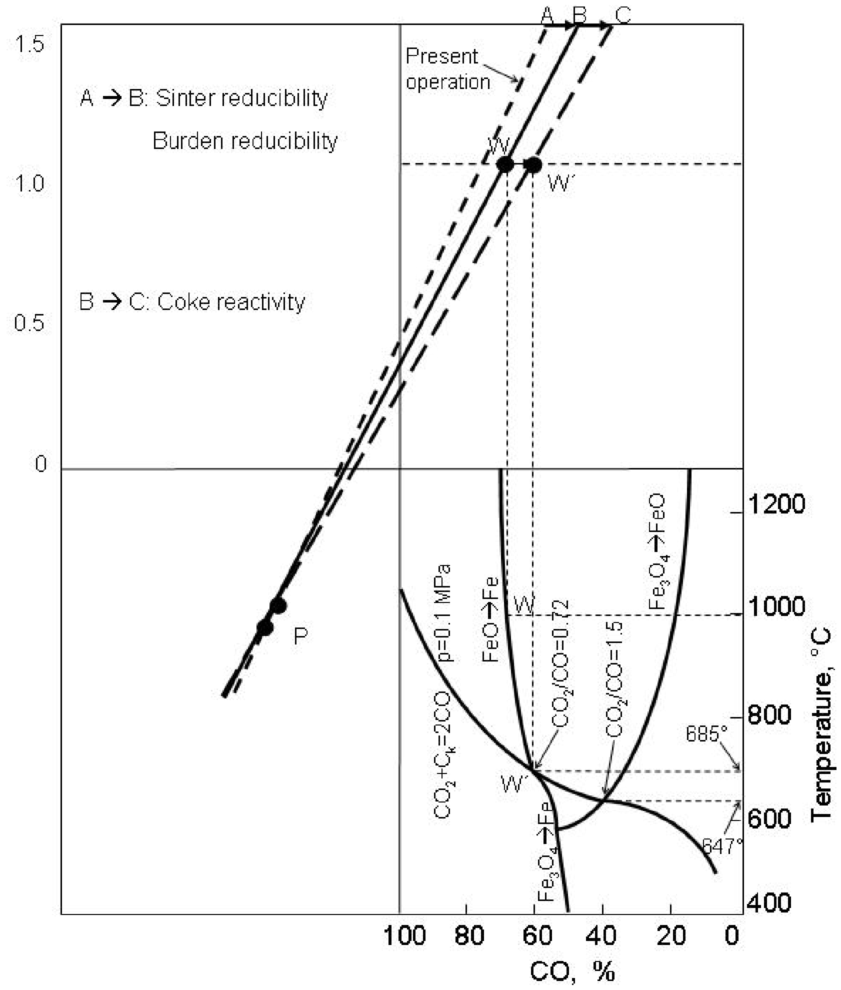


Fig. 1. Rist operating diagram combined with Baur-Glaessner equilibrium diagram and Boudouard reaction at P=0.1 MPa [18]

2 BIOMASS SOURCES, AVAILABILITY, PROCESSING AND QUALITY

The background for favourable CO_2 balance while using renewable biomass is the photosynthesis effect: $6CO_2 + 6H_2O = C_6H_{12}O_6 + 6O_2$, $\Delta H = 2826$ kJ; all the carbon in biomass is generated from CO_2 of the atmosphere and only part of it is released again as CO_2 in the ironmaking process. Beside the photosynthesis effect, various factors such as feedstock type and production, harvesting, processing, transport have to be considered for estimating the total environmental effect of biomass use. Furthermore, economic and technological factors have to be analysed carefully. For example, unique biomass properties like very low sulphur content, high ratio of carbon to ash and high specific surface may improve hot metal quality and blast furnace productivity. Presence of harmful elements in some types of biomass must be controlled; they may affect the metal quality at high injection rate.

Biomass sources can be categorised e.g. as follows: agricultural residues, forest biomass, energy crops, bio-waste streams (municipal solid waste, packaging and household waste wood, sewage sludge, food processing wastes etc.) and algae [20]. Presently woody and rarely herbaceous biomasses are being taken into account while discussing the biomass use in the steel industry. The worldwide biomass use can be very roughly estimated, because reliably statistics like for the fossil energy sources is missing. Renewables provide about 13% of global energy supply and biomass provides more than 3/4 of all renewable energy, mainly from woody biomass [21]. Biomass production and availability of its free resources differs significantly by world regions. In Germany and in Europe, the woody biomass production and use is balanced [21]. It has to be mentioned that share of energetic use of biomass (for heat and power generation) is steady increasing [22]. In the case of biomass import, costs and emissions by its transport have to be considered.

Chemical, physical and mechanical properties of raw biomass do not enable their high efficiency use in the steel industry. It is beneficial to use pyrolysed biomass - charcoals, semi-charcoals or torrefied materials - which are characterised by improved quality [23]:

- limited moisture absorption potential
- lower oxygen and higher carbon content
- higher calorific value
- higher apparent density
- better grindability (important for injection)
- better crushability (important for embedding).

By biomass trading, the processing on the export site is preferable from costs point of view because yield of charcoal makes up less than 1/3 and torrefied materials less than 2/3 (**Fig. 2**). Available processing technologies in exporting and importing countries have also to be considered both from economic and environmental points of view. The global warming potential of the emissions during production might be greater than the global warming potential of the emissions during combustion. Therefore advanced industrial technologies for charcoal production which enable recovery of chemicals and heat energy from the smoke are strongly recommended. Emissions of further GHG such as CO, CH_4, NMHC as well as TSP (Total Suspended Particulates) have to be takes into account.

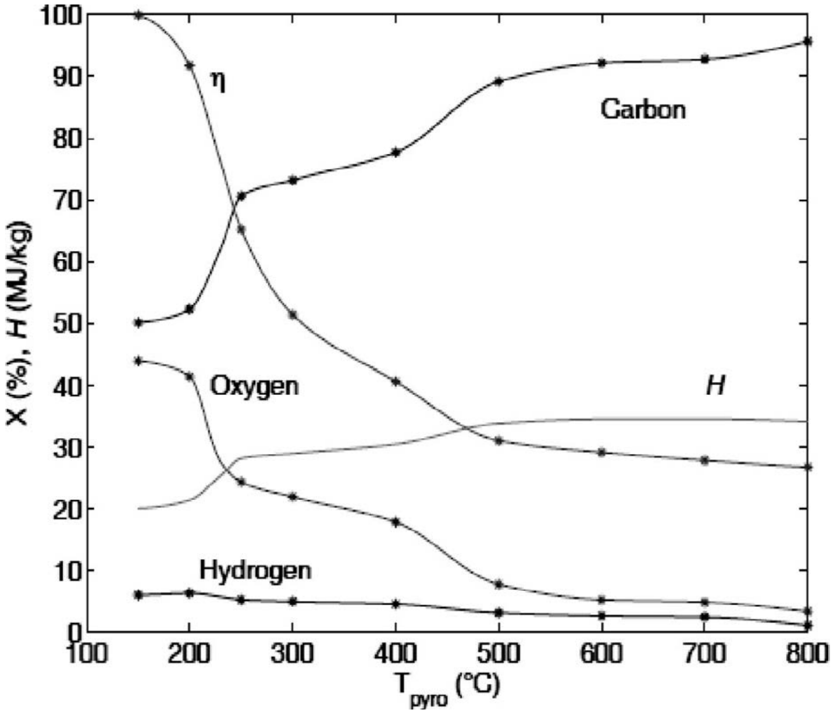


Fig. 2. Dependence of composition, yield (η) and heating value (H) on biomass pyrolysis temperature (example of Norway spruce) [24]

3 INJECTION OF PYROLSED BIOMASS VIA TUYERES INTO THE BF

Table 1 shows chemistry of two types of charcoals from eucalyptus (CC1 and CC 2), two types of torrefied biomass from pine (TB1) and beech (TB2) pyrolysed at three different temperatures and reference PC.

Table 1. Chemical analysis and calorific value of injectants

Injectants	**PC**	**CC 1**	**CC 2**	**TB1l**	**TB1m**	**TB1h**	**TB2l**	**TB2m**	**TB2h**
Ultimate analysis, %:									
Carbon	80.6	82.31	78.8	50.4	50.4	53.4	52.0	52.2	56.1
Hydrogen	4.35	3.14	2.8	6.29	6.29	6.04	5.57	5.54	5.33
Oxygen	5.35	8.24	11.15	42.8	42.5	39.8	41.8	41.6	37.7
Nitrogen	1.65	0.21	0.4	0.13	0.12	0.14	0.20	0.21	0.23
Sulphur	0.45	0.005	0.1	0.006	0.010	0.009	0.007	0.007	0.007
Proximate analysis, %:									
V M	24.7	30.5	ND	84.7	80.2	76.2	82.0	80.0	68.6
Ash	10.89	0.42	6.75	0.41	0.70	0.66	0.47	0.48	0.6
CaO in ash	1.49	24.5	19.11	39.7	39.7	39.7	41.10	41.10	41.10
SiO_2 in ash	57.15	14.0	32.15	5.31	5.31	5.31	2.56	2.56	2.56
LCV, MJ/kg	30.93	29,82	28.93	18.93	19.23	19.94	17.29	18.16	19.92

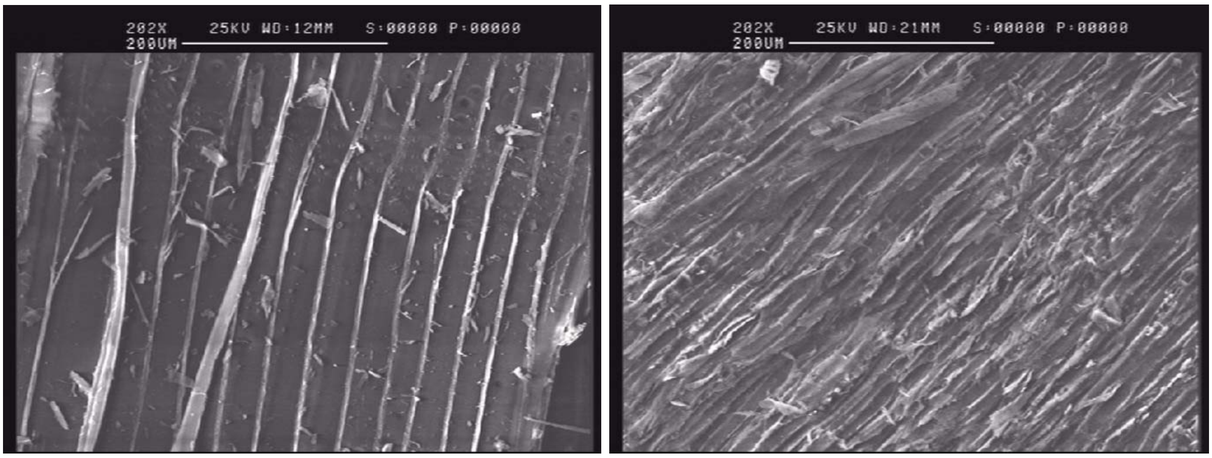
Comprehensive study on injection of charcoals from oak, olive and eucalyptus including CC1 and CC2 showed that combustion behaviour of the tested charcoals is comparable or even better than that for fossil coals for injection [7,25]. Solution loss reaction under shaft simulation conditions for charcoal goes on faster than for PC [7,8].
Present study is focused on torrefied materials. Pine wood chips (TB1) and beech wood chips (TB2), both without bark, were torrefied under same conditions (heating rate and pyrolysis time) at three different pyrolysis temperatures in the range between 200 and 350°C (TB-l, TB-m and TB-h in **Table 1** correspond to ‘low’, ‘medium’ and ‘high’ pyrolysis temperatures respectively). Compared to fossil coal used for injection (PC), torrefied biomass has negligible amounts of ash, but significantly higher amounts of oxygen and volatile matter. At lower and medium pyrolysis temperatures changes of the elementary composition cannot be detected. At higher pyrolysis temperatures the carbon content increases and the oxygen content decreases; the atomic ratio of H/C decreases from 1.50 to 1.36 (pine wood) and from 1.28 to 1.14 (beech wood). The atomic ratio of O/C decreases from 0.63 to 0.56 (pine wood) and from 0.60 to 0.50 (beech wood). The increasing carbon content in combination with the decreasing oxygen and hydrogen content results from the devolatilisation of oxygen-containing carboxylic acids especially formic acid and acetic acid [26].

The porosity of the torrefied pine wood examined using Mercury Inclusion Porosimetry (MIP) for three pyrolysis temperatures is shown in **Table 2**. The pores of the center fluid passage grow due to degradation during torrefaction process. A criterion for the level of decomposition during pyrolysis may be the apparent skeletal density, which is defined as the mass per skeletal volume, measured during MIP. The apparent skeletal density seems to remain on the same level during low and medium pyrolysis temperatures, but increases at higher pyrolysis temperatures.

Table 2. Porosity and apparent density of torrefied pine wood [26]

Pyrolysis temperature	Apparent density, g/ml	Porosity, vol. %
Low	1,085	63,5
Medium	1,021	67,1
High	1,219	73,3

SEM photographs in **Fig. 3** show torrefied biomass from pine wood (TB1) pyrolysed at two different temperatures. It can be recognised that the biomass torrefied at higher temperature has more porous microstructure.



“high” pyrolysis temperature “low” pyrolysis temperature

Fig. 3. REM pictures of torrefied biomass from pine wood (TB1), x200

First injection trials performed using the laboratory injection rig that simulates conditions in the tuyère, injection lance and in the raceway showed that conversion degree of the tested TB is comparable with reference coal.

Table 3 shows BF operation parameters when injecting charcoals and torrefied biomass (TB2) calculated using a mathematical model. Injection rate was kept at 200 kg/tHM. The flame temperature was controlled by oxygen enrichment of blast. Sinter/pellets ratio was adjusted to keep slag basicity on a constant level.

Table 3. BF operation results (selected calculated parameters)*

	PC (base)	CC 1	CC 2	PC-TB2 l	PC-TB2 h
Blast:					
Blast temperature, °C	1200	1200	1200	1200	1200
Oxygen in blast, %	26.78	26.0	25,4	30.0	30.0
PC / CC / TBM, kg/tHM	200	200	200	200	200
Burden, kg/tHM:					
Sinter	992	604	735	820	820
Pellets	435	785	660	590	590
Lump ore	150	150	150	150	150
Slag basicity	1.17	1.17	1.17	1.17	1.17
Calculated parameters					
Coke rate, kg/tHM	283	272	294	360	349
Slag volume, kg/tHM	260	182	214	230	229
Top gas:					
volume (dry), m^3/tHM	1406	1364	1425	1422	1406
CO_2, %	24.53	25.74	24.67	24.27	24.55
CO, %	23.60	22.97	23.63	28.38	28.03
H_2, %	4.68	3.66	3.44	5.66	5.53
calorific value, kJ/m^3	3489.1	3299.4	3358.9	4198.8	4141.8
temperature, °C	150.3	152.7	156.2	127.0	126.9
Heat balance, MJ/tHM					
total heat generated	9780.6	9618.3	9709.5	9661.6	9653.4
heat of top gas	314.1	307.6	327.0	269.8	266.3
external heat loss	700.7	672.6	693.7	676.5	675.2
Flame temperature, °C	2116.8	2118.9	2117.1	2108.5	2119.0
Productivity	100.0	103.0	98.8	99.0	100.0

*Burden and coke compositions are given in the reference [25]

It can be seen that injection of charcoal with low ash and high basicity (CC 1) decreases slag volume and coke rate. Furthermore heat loss decreases and furnace productivity rises. Top gas volume and its calorific value decrease as well. When injecting charcoal with high ash content and relatively low basicity (CC 2), main operation parameters including coke rate and productivity can worsen (depending on the reference PC characteristics) [7]. Injection of high amount of TB characterised by very high VM, very low carbon content and correspondingly low calorific value, would affect strongly the BF operation; therefore PC was replaced with TB by the calculations partly (50% of total 200 kg/tHM). Coke rate was increased by 27.2 and 23.3% by using TB carbonised at low and high temperatures respectively. Decreasing in top gas temperature and keeping the productivity on the initial level is the consequence of additional enrichment of blast with oxygen. Rise of CO content in top gas and its calorific value indicates lowering of gas utilisation rate. Slag volume and heat loss decreases while injecting TB.

The presented in **Table 3** effect of charcoals and torrefied biomass on coke consumption is in agreement with calculations made by Mathieson *et al* [12] showed that the coke replacement ratio for charcoal and torrefied biomass produced from the same wood type are 1.06 and 0.39 kg/kg respectively.

4 PELLETS AND COMPOSITES WITH EMBEDDED CHARCOAL

In the past, reduction and swelling behaviour of hematite and magnetite pellets with cold embedded low and high volatile matter coals and waste plastics were investigated at the IEHK in neutral atmosphere (Argon) [14,15]. Here, real atmosphere in the BF TRZ is being simulated. Furthermore, carburisation behaviour of iron ore-carbon composites with biomass was studied as well.

4.1. Reason for the use of SRP in a mini BF

Despite the common trend for increasing the BF dimensions, modern mini BFs (about 500 m^3) have certain advantages compared to the large ones, e.g. the lower strength requirements of raw materials and may operate with quite low reducing agent rate. Recently the mini BF with working volume of 400 m^3 and capacity of 500,000 TPA was commissioned by Tata Steel Thailand [27]. In the scope of current study, behaviour of new iron burden materials under TRZ simulating conditions is being investigated from in the context of their possible use in the mini BF. These materials – SRP with various cold embedded carbonaceous materials characterised by different reactivity – are being examined.

4.2. Ingredients, composition and characteristics of SRP

Pellets were produced from hematite iron ore used by Tata Steel Thailand mini BF in Thailand (92.56% Fe_2O_3, 4.54% SiO_2) with embedding charcoal from eucalyptus, coal (**Table 4**) and further reducing agents not presented in this contribution; grain size of all ingredients was 45-90 µm. Bentonite was used as binder. Here only SRP with 18 wt.% of reducing agent (every charcoal and coal) are presented; it is a maximal rate of embedded reducing agent that might affect the reduction degree [28]. Relevant chemical and physical characteristics of pellets are shown in **Table 5**.

Table 4. Reducing agent composition, wt. %

	C	H	N	S	O	C_{fix}	Ash	S	VM
Coal	73.2	3.31	1.47	0.4	3.2	68.9	11.9	0.4	17.7
Charcoal	91.6	2.68	0.38	0.02	5.3	80.3	0.57	0.02	19.1

Table 5. Properties of SRP

Pellet type	Reducing agent	C/O atomic ratio	C_{fix}/O atomic ratio	VM, %	Density, g/cm^3	Specific volume, cm^3/g
1	no	0	0	0	3.5	0.29
2	coal	0.67	0.63	3.2	3.0	0.33
3	charcoal	0.82	0.72	3.4	1.8	0.56

4.3. Test results so far

Experimental set ups, test procedure and scenarios are described in reference [29]. **Table 6** shows an example of the test result for pellet reduction after 30 min. It can be seen that embedded carbon promotes iron oxide reduction. Charcoal effect is stronger compared to coal due to its higher reactivity.

Table 6. Weight loss and reduction rate of pellets during isothermal reduction at 900°C in 20%CO-20%CO_2-60%N_2 atmosphere

Pellet type	Total wt.-loss, %	C wt.- loss, %	Reduction degree, %	FeO/(FeO+Fe_2O_3), %
1	7.6	-	15.7	36
2	11.1	30.0	18.3	52
3	16.0	49.4	21.9	63

Fig. 4 shows test scenario for non-isothermal reduction simulating the moving of pellets through the TRZ: gas composition and temperature development follow the black arrows; heating rate was 5K/min; cross (x) marks isothermal plateau at 1000°C for 30 min. Selected results obtained from the Tammann furnace tests are presented in **Fig. 5** and **Table 7**. The results confirm that the highest weight loss is dedicated to SRP with charcoal. Shrinkage behaviour of SRP during reduction was observed for both reducing agents. The maximal shrinking (52%) was observed for SRP with charcoal (Table 7).

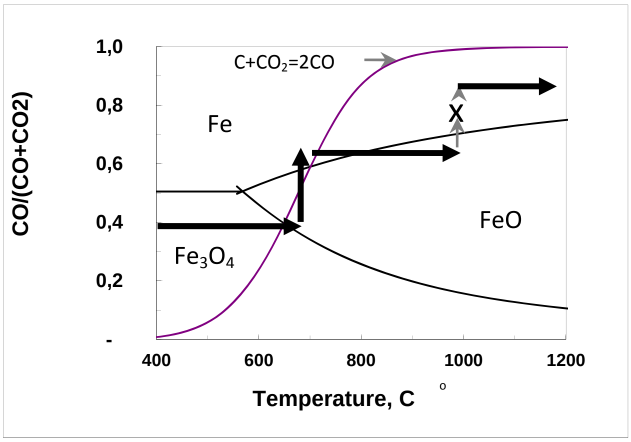


Fig. 4. Test conditions on Baur-Glaessner diagram

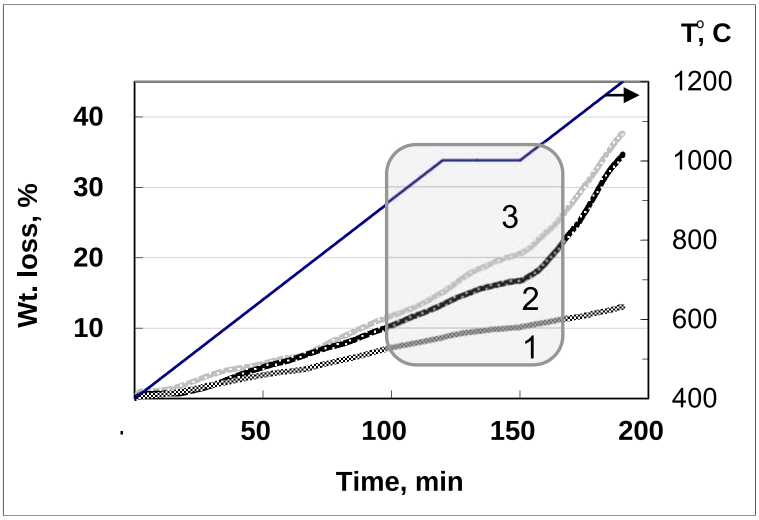


Fig. 5 Weight loss of pellets during non-isothermal reduction: 1,2,3 – pellet types (see Table 6), grey area – TRZ conditions

Table 7. Volume change of pellets underwent non-isothermal reduction tests

Pellet type	Volume change, %
1	0
2	-23
3	-52

Pellet strength depends on firing temperature, type and rate of embedded carbonaceous material (**Fig. 6**). For the normal large BF, pellet strength nearly 2000 N/pellet or higher is required [30].For the mini BF its lower value should be sufficient. Therefore it might be possible to use SRP with embedded charcoal or other biomass in the mini BF by firing at higher temperatures and by limiting reducing agent rate. Investigation of SRP with biomass char targeting at lower amount of embedded carbonaceous material and higher strength of pellets are running now.

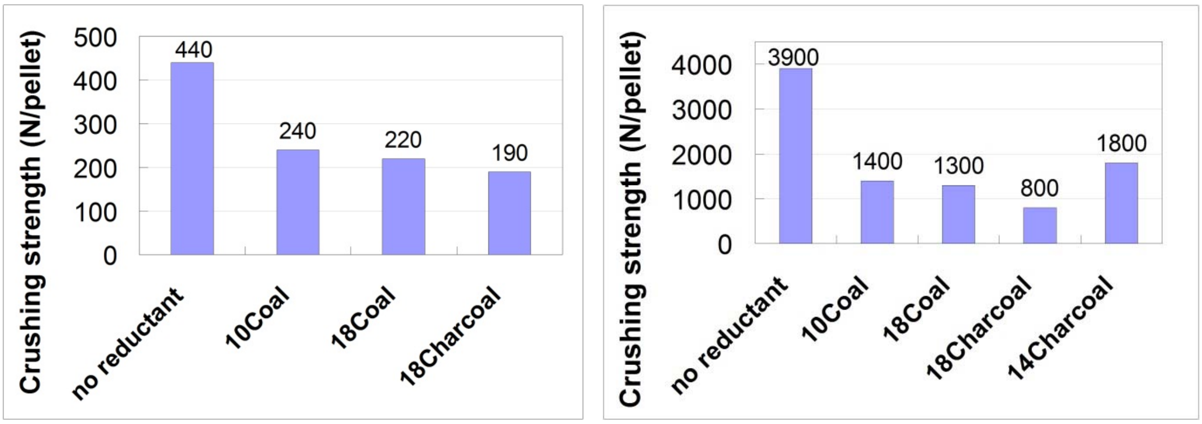


Fig. 6. Crushing strength of SRP after firing at 1000°C (left) and1250°C (right)

4.4. Carburisation behaviour of iron ore - carbon composites with biomass

Carburisation behaviour of iron ore-carbon composites with different charcoals was investigated by Dr. K. Ohno using various analytical and laboratory facilities at the IEHK, RWTH Aachen University in Germany and Kyushu University in Japan. Particularly, effect of carbon crystallinity and ash content in charcoal on carbon dissolution in molten iron and carburisation reaction in the composite were examined

[19]. It was found that charcoal ash strongly prevents the reaction between iron and carbon.

5 CONCLUSIONS

Biomass can be used in iron- and steelmaking by gasification, injection and incorporation into coal blends and burden materials. Its efficient and sustainable application depends on the world region.

Pyrolysed biomass (charcoals, semi-charcoals or torrefied materials) are more suitable for usage in the steel industry.

Charcoal conversion in the raceway might be better or at least comparable with coals for injection. Injection behaviour of torrefied biomass materials is being investigated. When injecting charcoals or torrefied biomass, BF operation results depend mainly on ash content and composition, and pyrolysis temperature of injectants. In case of constant injection rate, torrefied biomass materials require higher oxygen enrichment of blast and causes higher coke rate.

Self-reducing pellets and iron ore - carbon composite with embedded charcoal are characterised by higher reducibility and ability to decrease carbon consumption in the BF but also by lower crushing strength. An introduction of such new burden materials in the mini BF with lower strength requirements might be easily. Amount and type of embedded carbonaceous material as well as firing temperature should be adjusted to rich suitable strength of pellets. Physical and chemical properties of charcoal in composites affect carburisation reaction.

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