



Production of charcoal as an alternative reducing agent from agricultural residues using a semi-continuous semi-pilot scale pyrolysis screw reactor

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ABSTRACT

Nowadays there is widespread concern regarding the CO₂ emissions derived from fossil fuels due to the contribution of this gas to the greenhouse effect. Many metallurgical processes use fossil carbonaceous materials like e.g. petroleum coke as a reducing agent involving the emission of substantial amounts of carbon dioxide. One interesting alternative is the substitution of the mentioned fossil-based reducing agents with biomass derived charcoal. Typical carbonization processes are operated between 623 and 773 K, but this charcoal does not fulfill the required quality for metallurgical processes. In the present paper a (semi-) continuous carbonization process was developed at high temperatures in order to produce charcoal to be used as a reducing agent. The effect of the temperature, residence time, moisture content and grain size were studied in an auger pyrolysis reactor, and the charcoal quality was tested carrying out reactivity tests, CHN + S as well as calculating the fixed carbon percentage.

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1. Introduction

Nowadays there is widespread concern regarding the CO₂ emissions derived from fossil fuels due to the contribution of this gas to the greenhouse effect and therefore to climate change. Moreover, environmental legislation is becoming stricter and companies have to pay additional fees depending on the amount of CO₂ they emit. Many metallurgical processes use fossil carbonaceous materials such as petroleum coke as a reducing agent involving the emission of great amounts of carbon dioxide that contribute to the greenhouse effect. The utilization of CO₂-neutral reducing agents is one of the technological goals in the above-mentioned field. In literature, hardly any direct use of biomass as a reducing agent is reported. The main problem is the very fast reaction of fresh biomass and therefore a relatively low overall yield [1–4]. Fick et al. [5] studied several biomass sources as reducing agents in the ironmaking industry like charcoal, bio-oil, syngas, torrefied biomass and biogas. They concluded that the utilization of charcoal or carbonized biomass was the most promising alternative.

The thermal treatment of biomass is known as pyrolysis and it consists of a heating process in an oxygen-free environment. In this thermal process, three different products are produced: a solid fraction (charcoal), a liquid fraction (bio-oils or tars) and non-condensable gases. Depending on the pyrolysis (temperature and residence time) conditions, one of

these fractions can be maximized [6]. Lower process temperatures and longer vapor residence times favor the production of charcoal (673 K). High temperatures and longer residence times increase biomass conversion to gas (1023–1173 K), and moderate temperatures (773 K) and short vapor residence times are the optimum conditions to produce liquids (bio-oil). The problem of typical carbonization processes comes from the high reactivity of the charcoal produced. Moreover, carbonization processes in typical industrial carbonization reactors at temperatures between 673 and 773 K are often very simple and inefficient processes concerning their charcoal yield and the usage of by-products like pyrolysis gas [7].

In blast furnaces, the main problems of a coke substitution using charcoal are the missing compressive strength and the too high reactivity, which means replacement is only possible in mini blast furnaces of a reduced height for iron ores which can just be found in Brazil and China. These problems are not so important in the nonferrous metal industry, making the target of the work presented in this paper the production of charcoals which can be used in the said field. The requirements for charcoals as chemical reductants in metal processing and recycling are a fixed carbon content of more than 85–90% as well as a volatiles content of less than 10%. As an example, Table 1 shows the chemical analysis, surface area and reactivity values of conventional petroleum coke used in metal recycling processes. In order to fulfill these requirements, high carbonization/pyrolysis temperatures up to 1173 K are necessary [8]. Apart from high temperatures, low heating rates are important in order to maximize the charcoal yield. Thus, the target of the present work is to find the most suitable process parameters (temperature, heating rate, biomass residence time) for the production of charcoal as well as the best biomass conditions (grain size, moisture content).

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Table 1
Chemical analysis, surface area and reactivity CRI and R-values of a typical commercial petroleum coke.

Volatiles %	4.8
Fixed carbon %	90.3
Ash %	4.9
Moisture %	12.4
^a Sulphur %	6.1
Surface area (m ² /g)	2.34
Reactivity	
CRI	13.7
R-value	16.5

^a Sulfur content is given in dry basis.

2. Materials and methods

2.1. Materials

Fruit cuttings from the Spanish region of Extremadura were used as feedstock in the present research activities. This kind of residue from agriculture is generated in vast quantities and has been used neither for the production of energy nor in a substantial way to date. This fact leads to an acceptable price for such feedstock material, which could be quite important in a possible implementation of the process studied on an industrial scale in the near future. Table 2 shows an overview of the chemical analysis of the investigated fruit cuttings. All the experiments were performed using the aforementioned fruit cuttings but some differences were observed in the chemical composition of the two different deliveries from our supplier. However, each effect was studied with exactly the same fresh biomass.

2.2. Pyrolysis unit, experimental procedure

The pyrolysis reactor used for the experiments is an externally heated and gas-tight double-screw reactor (auger reactor), where the temperature and the rotation speed of the screw can be set individually. The schematic drawing of the experimental unit with its main components is shown in Fig. 1. The external and electrical heating are split into three individual heating zones where the temperature is adjusted separately. This guarantees a continuous increase in temperature over the whole length, which can be controlled and observed at five temperature measurement points (T1, T3 and T5 are controllers while T2 and T4 are simple temperature sensors) distributed evenly along the length of the reactor. To prevent heat losses, the heated part of the reactor is thermally isolated with ceramic fiber mats. The length of the screw reactor is 2.3 m and the height of the installation is more than 2 m, allowing the treatment of 1 to 10 kg/h of biomass with a maximum carbonization temperature of more than 900 °C.

The experiments themselves were carried out in a semi-continuous way, which means that the feedstock material was first put into the feeding bin, which was then sealed gas-tight. At the same time, the heating elements were switched on to bring the reactor to the desired

Table 2
Chemical analysis results of the fruit cuttings studied.

	Biomass 1	Biomass 2
	wt.%	wt.%
Volatiles %	74.3	73.6
Fixed carbon %	21.1	23.6
Ash %	4.60	2.78
Moisture %	11.00	6.80
^a Carbon %	46.5	47.3
^a Hydrogen %	6.23	6.5
^a Nitrogen %	0.59	0.47
^a Sulphur %	0.08	<0.03

^a The elemental analysis (CHN + S) is given in dry basis.

temperature. When the reactor reached the desired temperature, the motor was switched on at a certain adjusted rotation speed, feeding the biomass to the reactor continuously. In this way, the biomass temperature increases continuously and as a result, this leads to the release of the volatile matter which then forms the emitted pyrolysis gas. This gas forms the atmosphere in the reactor and leaves the reactor at the top of the charcoal bin where it is analyzed and post-combusted. The atmosphere of the unit is not purged with N₂ beforehand. The initial pyrolysis vapors remove all the oxygen in the very first minutes of the process, as it was observed in the continuous gas measurement.

The carbonized charcoal exits the reactor on the same side as the gas, where it falls into the charcoal bin, also in the absence of oxygen. When the complete biomass is processed, the heating is switched off and the reactor with the charcoal inside cools down to room temperature. When the whole facility is at room temperature, the charcoal bin is opened, the charcoal is weighed and a representative sample is taken in order to be analyzed concerning its chemical composition and carry out some further charcoal reactivity tests, as shown in Section 2.4. After cleaning the charcoal bin, all parts are sealed again and the reactor is ready for new experiments.

2.3. Gas analysis

Beside the produced bio-char, the gases emitted during the carbonization tests were also analyzed using a continuous infrared gas analyzer (ABB-EL3020). A small amount of pyrolysis gases was sucked up and measured by the gas analyzer. The measurable gas components are CO, CO₂, CH₄, SO₂, H₂ and O₂. In order to prevent contamination and problems with the gas analyzer, the tar inside the gas, which can reach up to 25% in total, has to be collected beforehand. For that reason, a two-step tar trap was installed between the pyrolysis unit and the gas analyzer. The first part of the gas cleaning set-up was a common gas washing bottle. The second part consisted of two jars filled with Raschig Rings, where the remaining tar is condensed. The gas analyzer consists of a heated filter so as to ensure that no tar particles go into the analyzer. Apart from the main gas composition, the continuous gas measurement and the online plotting of the composition curves provide relevant information with regard to the process behavior, like the transient state and the steady state time periods.

2.4. Charcoal analysis

A central fact concerning the utilization of reducing agents in metallurgy is their reactivity. In most metallurgical processes it is very important how intensive or at which position inside the process unit the carbon carrier reacts. Here, some differences and changes can have very strong effects on the process and especially on the product quality. So, if charcoal is used as an alternative reducing agent in metallurgy, it is very important to investigate the reactivity of charcoals and their influence on the processes. Therefore, standardized reactivity measurements like the ASTM D 5341–93 [9] method or the Koppers test can be applied. Both tests take the Boudouard reaction (Eq. (1)) as a basis. The carbon reacts with the CO₂ provided and forms CO. Depending on the reactivity of the material, the quantity of reacting carbon is variable. With the Koppers test a so called R-factor (Eq. (2)) can be calculated, whereas the ASTM D 5341–93 method determines the coke reactivity index CRI (Eq. (3)). The R-factor represents the amount of CO in the gas stream after reaction related to the originally existing CO₂, while the coke reactivity index is the mass loss after the reactivity test. Both values are higher at higher reactivity of the investigated material [9,10,11].



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