



Recycling tyre wastes as additives in industrial coal blends for cokemaking



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ABSTRACT

Two industrial coal blends (B1 and B2) used in cokemaking were selected for this study. Two wastes from scrap tyres (TC, F) were added to these coal blends at different ratios (98:2 and 95:5). The investigation was focused on assessing the influence of the additives on thermoplastic properties of coal by means of the Gieseler fluidity test and thermogravimetric analysis. In addition, the blends were carbonized in a 17 kg electrically heated movable wall oven in order to examine the quality of the resultant cokes. Quality of the cokes produced was evaluated by measuring their cold mechanical strength, reactivity and post-reaction strength. It was found that ash composition of the additives contribute to a deterioration in coke quality. Moreover, F causes a greater decrease in coke strength after reaction with CO₂ due to the diminution of bulk density in the coking process and to the higher basicity index. Trace elements were also studied in relation to waste addition.

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1. Introduction

The international coal market has changed considerably in recent years, giving rise to a notable variability in prices. Because of the short supply of high-quality coking coals, industrial coal blends are normally used in cokemaking. The quality of the blend depends on the quality of individual coals and their interaction within the blend. The coal blends are usually made up of coals with different characteristics and from different origin. The co-carbonization of coal blends with additives has been observed to modify the coking properties of coals and the quality of the resulting cokes significantly [1,2]. In the present work, effects of adding carbon-rich materials from the recycling tyre industry to industrial coal blends were studied. Only a small proportion of additives was used because of their high sulphur content that could negatively affect coke quality. Carbonization was studied as a possible route for recycling waste tyres to avoid the need for dumping, thereby contributing to the protection of the environment, and reducing the costs related to waste disposal. Previous investigations studied that tyres and their char could be used as additives to coke-oven blends without causing any apparent deterioration in coke quality, although the particle size of the waste tyre added had an impact on the quality of the coke produced [3,4]. Moreover, the heaviest fraction of tyre-derived liquid can be used as a raw material for producing coke of good quality with low sulphur, ash and metal contents [5]. There are other ways to recycle these kinds of wastes. Waste tyres can be co-injected with metallurgical coke in the electric arc furnace steelmaking process to provide additional energy

for combustion [6]. Previous works study the combustion of different rank coals mixed with end-of-life tyres in an atmospheric fluidized bed combustion (AFBC) pilot plant [7]. This combustion system shows several advantages over conventional systems, such as fuel flexibility and better emission controls. It offers the advantages of making tyres a proper fuel to generate energy when mixed with high ash coals. Addition of tyre to coal hydrogenation processes improves conversions due to a cheap hydrogen donation or to a simple reduction of heat and mass transport phenomena, and thus to lower costs [8].

The resultant coke will be used in a blast furnace for pig iron production, a route that will remain the basis of steel production in the foreseeable future. Coke cannot be fully replaced as a raw material in the blast furnace for physical reasons [9,10]. Its quality plays a significant role in controlling the performance of the furnace. Coke quality mainly depends on the characteristics of the coal or blend, although coking parameters and precarbonization techniques, such as compaction of the blend, also have an influence on its bulk density [11]. A high-quality coke should be able to withstand the gradual descent of the charge in the blast furnace with as little degradation as possible while providing the lowest possible amount of impurities, a high level of thermal energy, a high degree of metal reduction, and optimum permeability for the flow of gaseous and molten products. The use of high-quality coke in the blast furnace will result in a lower coke rate, greater productivity and a lower pig iron cost.

Fluidity is another factor that has a significant influence on coke quality. When a bituminous coal is heated, the individual coal particles fuse and form a porous coherent mass that expands and finally solidifies to form a semicoke that with further heating is converted into coke [1,2,12].

Ash is undesirable firstly because it reduces the carbon content, and also because it has to be melted which will require the addition of more

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limestone in the ironmaking process. This in turn will result in extra slag, leading to higher coke consumption and the restriction of output [13]. In simple terms the coke ash content should not exceed 8–12 wt.% on a dry basis [14].

The sulphur content of the coke directly affects the quality of the iron and the steel produced becomes brittle because of its presence. According to blast-furnace coke specifications, sulphur content should not exceed 0.6% [15]. Ash sulphur is also regarded as an undesirable component in coke, as it impairs its quality and increases its consumption in pig iron production. Furthermore a large amount of thermostable sulphur compounds in the coal increases the thermostability of the sulphur compounds in the coke, the sulphur concentration in the gas phase within the tuyere zone of the blast furnace, and hence the sulphur content of the pig iron [16].

In a previous paper [17], variations in the mineralogy of a coke obtained from blends of a medium rank coal and additives and their effect on coke reactivity was studied. The presence of new Zn-bearing phases in the tyre wastes was found to be the main cause of the increase in coke reactivity along with magnetite levels and pore size. The amount of Zn that remains in the coke depends on the coal, and it has been observed that the higher the rank of the coal, the lower the amount of zinc that remains in the coke [18]. The tyre wastes produced the most reactive chars and consequently only small amounts of tyre wastes should be added to industrial coal blends to avoid any severe impact on the CSR index. In light of these problems, the aim of the present research work was to investigate the possibility of recycling tyre wastes by means of co-carbonization with industrial coal blends and assess the impact of these wastes on coke quality. Two kinds of tyre waste were added to two industrial coal blends. An exhaustive analysis of the properties of the raw materials as well as of the resultant blends was carried out at laboratory scale in order to evaluate the effect of the amount of wastes to be added. The coking of coal blends and their blends with tyre wastes was conducted at semi-pilot scale.

2. Experimental

2.1. Materials

To avoid weathering, two industrial coal blends prepared in the same coking plant and composed of 10 coals of different origin were chosen for the study (B1, B2). Two wastes from the tyre recycling industry were selected as additives: the tyre crumbs (TC) and fluff/fibres obtained as a waste from the grinding and shredding of scrap tyres (F) during the processing of car and truck tyres. The fluff, a mixture of thermoplastic polymers (e.g., polyester and nylon) and tyre crumbs, contains around 60 wt.% of rubber.

2.2. Proximate and elemental analyses

Proximate analyses were performed following the ISO562 and ISO1171 standard procedures for volatile matter and ash contents, respectively. The elemental analysis was carried out on a LECO CHN-2000 for C, H and N (ASTM D-5773), and a LECO S-144 DR (ASTM-ASTM D-5016) for sulphur.

2.3. Gieseler plastometry

Thermoplastic properties of the industrial coal blends and the laboratory-prepared blends formed by adding 2, 5 and 10 wt.% of each additive were assessed by means of the Gieseler method in a R.B. Automazione Gieseler plastometer PL2000, following the ASTM D2639-08 standard procedure which has been explained in detail in a previous paper. The optimum Gieseler maximum fluidity as established by the MOF diagram, ranges from 200 to 1000 ddpn [19].

2.4. Thermogravimetric analysis (TG/DTG)

The TG/DTG analysis of the coal blends was carried out using a TA Instruments SDT 2960 thermoanalyser. 10–15 mg samples with a particle size of <0.212 mm were heated to 1000 °C at a rate of 10 °C/min under a nitrogen flow of 100 ml/min. From the data obtained by thermogravimetric analysis the volatile matter evolved up to a specific temperature (VM_T) and the derivative of the weight loss curve (DTG curve) were calculated. The volatile matter evolved over a specific temperature range was calculated as the difference between the volatile matter evolved up to two specified temperatures ($VM_{T_1 - T_2}$). In addition, T_{max} , the temperature of maximum volatile matter evolution was derived from the TG/DTG curves [20,21].

2.5. Carbonization test

Carbonization tests were carried out in a movable wall oven of approximately 17 kg capacity (MWO17). The dimensions of the oven are 270 mm L × 165 mm W × 790 mm H. A load cell was mounted on the movable wall to measure the force exerted on the wall during carbonization. A programmable controller was used to control the oven temperature. The temperature at the centre of the coal charge was monitored by means of a thermocouple connected to a computer. The coal was charged when the oven reached 1100 °C. The temperature of the wall was kept constant throughout the test. The coke was pushed out of the oven 15 min after the centre of the charge had reached 950 °C. The coking time was around 3.5 h. The moisture of the charge was fixed at 5 wt.%. The bulk density expressed on a dry basis was 771 and 778 kg/m³ for B1 and B2, respectively. More than 80 wt.% of the grain size of the coal blends was smaller than 3 mm. 43 wt.% of the tyre crumbs had a particle size in the 2–3 mm range, while in the case of the fluff/fibres, 57 wt.% it was smaller than 2 mm.

2.6. Ash analysis

The total ash content of the coke samples was obtained by combustion of the organic matter at 815 °C until constant mass, following the ISO 1171 (2010) standard. The concentrations of oxides (Al, Ca, Fe, K, Mg, Na, Si, P and Ti) were determined using X-ray fluorescence spectroscopy (XRF). The XRF apparatus was a sequential wavelength-dispersive Siemens SRS 3000 X-ray spectrometer equipped with a Rh X-ray tube, a 58 position sample turntable, and a flow counter detector. Prior to the XRF analysis the ashes obtained from the coke samples were subjected to a fusion step using lithium tetraborate in order to obtain sample beads for analysis. The device used to prepare the sample was a Philips Per! X3 automatic fused bead machine.

In order to determine trace elements (Zn, Mn, V, Ni, Pb, Co, Cu and Sb), analyses were carried out using inductively coupled plasma spectrometry (ICP-MS) in an ICP-MS 7700x Agilent device. The ashes were digested with HNO₃:HCl at a ratio of 3:1 in a microwave oven.

2.7. Textural characterization

The coke densities were measured using the displacement method in gas and liquid media (helium, water and mercury). The true density (ρ_{He}) of the cokes was measured by means of helium pycnometry in a Micromeritics AccuPyc 1330 Pycnometer. The apparent mercury density (ρ_{Hg}) was determined with mercury at a pressure of 0.1 MPa in a Micromeritics autopore IV 9500 mercury porosimeter. The particle size used for both methods was between 1–3 mm. The apparent water density (ρ_{H_2O}) was determined by water displacement using 300 g of coke sample with particle sizes between 19–22.4 mm.

Total coke porosity was calculated from the apparent water and true helium densities, while the true helium and apparent mercury densities

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