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Research article

Effect of briquette composition and size on the quality of the resulting coke

M.G. Montiano, E. Díaz-Faes, C. Barriocanal *

Instituto Nacional del Carbón (INCAR-CSIC), Apartado 73, 33080 Oviedo. Spain

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ABSTRACT

Five briquettes were prepared using sawdust, a non-coking coal and a binder. Industrial coal blends were used to study the influence of the type of sawdust (pine and chestnut), the binder (coal tar and coal-tar sludge) and the size of the briquettes on the quality of the cokes produced from mixtures containing up to 15 wt.% of the five briquettes. The effect of the briquettes and briquette components on the fluidity of the industrial coal blends was investigated. It was found that biomass and non-coking coal produced a decrease in fluidity, whereas the binders increased it. The combined effect of both types of additive had the global effect of decreasing fluidity. Mixtures of the briquettes with the industrial coal blends were carbonized in a 17 kg movable wall oven in order to assess their influence on the quality of the cokes produced. Their cold mechanical strength (JIS DI150/15 index), reactivity to CO_2 (CRI index) and post-reaction strength (CSR index) were also tested. The composition of the ash of the sawdusts and the reactivity of the briquette components were used as an indication of the effect on coke reactivity. The effects on cold mechanical strength and post-reaction strength were different in some cases.

1. Introduction

The steel industry is an energy and carbon-based intensive process and therefore a major contributor to global anthropogenic CO_2 emissions [1–4]. At the same time, cokemaking is a process where the recycling of wastes is possible, especially high carbon and low ash wastes like sawdust and charcoal [5–9], plastics [10–12], and bituminous wastes [13–15]. Thus recycling in the cokemaking industry could provide a way to reduce the environmental impact of CO_2 emissions, reduce costs and widen the raw materials spectrum to include non-fossil fuels.

The inclusion of sawdust in coal blends for cokemaking has clear advantages such as its low sulphur and ash content and its zero contribution to CO_2 emissions but it also has a number of disadvantages including its low char yield, deleterious effect on coal fluidity [16,17] and low bulk density [18]. A possible way to increase the bulk density of the biomass is to prepare briquettes. Partial briquetting of coal charges was introduced by the Japanese industry in the 70s, as this technique enabled the amount of expensive prime coking coal in the blend to be reduced and a cheap non-coking coal to be used instead without any deterioration of the quality of the resultant coke [19]. In a previous paper [7] a comparison of the direct addition of sawdust and addition via briquetting was carried out. However, other factors such as size and composition still need to be evaluated in order to know whether it is possible to apply sawdust briquetting to cokemaking.

Various binders can be used for the preparation of briquettes. However both sawdust and non-coking coal have a deleterious effect on the development of coal fluidity [16,17] making pitch and coal tar preferable binders considering that both of these produce an increase in coal fluidity [20,21]. Coal-tar pitch has already been successfully used as a binder [22]. The role of pitch in briquettes comprising high-rank and coking coals is to interact with them and modify their carbonization behavior so that the system is sufficiently fluid to wet the surface of non-fusing coals. In addition it needs to be able to form a binder coke with a mosaic optical texture that connects coal-derived coke with inerts. The drawback with coal-tar pitch is its high carcinogenic compound content [23]. An alternative option is to use coal tar which does not cause as great an increase in fluidity as coal tar pitch but is nevertheless liquid and has fewer carcinogenic polyaromatics.

The aim of the present research work is to determine the influence of the type of sawdust and the size of the briquettes and binder used for their preparation on the quality of the coke produced from the co-carbonization of the briquettes with industrial coal blends.

2. Experimental

2.1. Materials characterization

Three industrial coal blends (CB2, CB3, CB4) were used together with the briquettes in the carbonization tests. The briquettes were prepared using a low volatile non-coking coal (C) and two sawdusts, one from chestnut (SC) and the other from pine (SP). As binder for the preparation of the briquettes coal tar (T) and coal tar sludge (M) from the

* Corresponding author.
E-mail address: carmenbr@incar.csic.es (C. Barriocanal).

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coking plant were used. The raw gas evolving from the coke ovens is treated in order to separate the permanent gases, ammonia, benzol, and tar. The coal tar sludge (M) collected at the bottom part of the tar decanter, apart from tar also contains some coal and coke that is drawn away and deposited on the bottom of the decanter. Five briquettes with different compositions were prepared using a roll press briquetting machine (Table 1). The briquettes were ellipsoid shaped with axes 46 and 42 mm in length and weighting around 23 g. On the basis of their different compositions it is possible to study: 1. the influence of including SC (B1 vs. B4 and BM1 vs. BM4); 2. the influence the type of binder (B4 vs. BM4 and B1 vs. BM1); 3. the effect of the two sawdusts, chestnut vs. pine (BM1 vs. $BM1_{sp}$). To study the effect of the size, samples of briquette of weight between 4 and 6 g, with the same composition as $BM1_{sp}$ and BM4 were used for the co-carbonization tests and labelled $BM1_{SP\text{-}F}$ and $BM4_{F}$.

Elemental analysis was carried out following the standard ISO 562 and ISO 1171 procedures for humidity, ash and volatile matter respectively. For the elemental analysis the following standard procedures were used: ASTM D 5016-98 and ASTM D 5373-02 for C, H and N using a LECO CHN-2000 and a LECO S-144 DR instrument for the analysis of S.

The apparent density of the briquettes was determined by means of water displacement by immersing 6 briquettes in a 500 ml container. The density of briquettes $BM1_{SP\text{-}F}$ and $BM4_{F}$ could not be measured because they disintegrate in water.

2.2. Textural characterization

The particle size used to determine the porous structure of the materials was between 1.18 and 0.8 mm. The true density (ρ_{He}) of the sawdusts was measured by means of helium picnometry on a Micromeritics Accupyc 1330 Pycnometer. Their apparent density (ρ_{Hg}) was determined using mercury at 0.1 MPa on a Micromeritics autopore IV 9500 mercury porosimeter. From the true and apparent densities the open porosity corresponding to pore sizes of <12 μm was calculated by means of the following equation:

The total pore volume (V_T) was obtained from the equation:

$$\varepsilon\,(\%) = \left(1 - \frac{\rho_{Hg}}{\rho_{He}}\right) \cdot 100 \tag{1}$$

$$V_T\,(cm^3/g) = \left(\frac{1}{\rho_{Hg}\left(g/cm^3\right)} - \frac{1}{\rho_{He}\left(g/cm^3\right)}\right). \tag{2}$$

The pore size distribution was calculated by applying increasing pressure to the sample from 0.1 to 227 MPa. This resulted in pore sizes in a range of 12 μm to 5.5 nm according to the Washburn equation.

Pore size was classified into two categories: macropores (12 μm > dp > 50 nm) and mesopores (50 nm > dp > 5.5 nm).

2.3. Variation of coal blend fluidity due to briquette addition

The thermoplastic properties of mixtures of the coal blends with 5, 10 and 15 wt.% of briquettes B1, BM1, BM1sp, B4 and BM4 were measured. Also the effect of the two binders, the sawdusts and the non-coking coal on the fluidity of the coal blends was assessed by means of the Gieseler test (ASTM D2639-74), this test having been successfully used previously to determine the modification of coal fluidity due to the use of additives [17,20,24]. The sample was heated at 3 °C/min up to a final temperature of 550 °C, while a constant torque was applied to the stirrer inside the crucible containing the sample. The spin rate of the stirrer was measured continuously until it stopped. The parameters derived from this test were: (i) softening temperature, Ts; (ii) the temperature of maximum fluidity, Tf; (iii) resolidification temperature, Tr; (iv) plastic range, Tr–Ts, which is defined as the difference between the resolidification and softening temperatures; and (v) maximum fluidity, MF, expressed as dial divisions per minute (ddpm).

Table 1
Briquettes composition.

	B1	B4	BM1	BM4	$BM1_{SP}$
T	15	15	–	–	–
M	–	–	15	15	15
SC	15	–	15	–	–
SP	–		–	–	15
C	70	85	70	85	70

Table 2
Main characteristics of the raw materials.

	CB2	CB3	CB4	C	SC	SP	T	M
Ash (% db)[b]	8.8	8.5	7.8	10.2	1.3	0.3	0.8[a]	2.2[a]
Volatile matter (% db)	23.9	23.0	26.2	14.5	78.5	85.3	–	–
Gieseler MF[c] (ddpm)	682	–	1016	n/a	n/a	n/a	n/a	n/a
C (% db)	80.3	82.1	82.4	80.8	50.2	50.7	90.3	89.1
H (% db)	4.6	4.7	4.9	4.0	5.7	6.1	4.7	4.2
N (% db)	1.8	1.9	1.9	1.7	0.5	0.5	0.8	1.1
S (% db)	0.58	0.51	0.62	0.45	0.01	0.00	0.38	0.52
O (% db)[c]	3.9	2.1	2.4	2.9	42.3	42.4	3.0	5.1
Particle size (wt.%)[d]								
>3 mm	12.0	20.4	26.6	14.9	1.0	0.2	n/a	n/a
2–3 mm	7.0	9.8	10.6	7.7	4.5	1.3	n/a	n/a
1–2 mm	14.8	16.6	17.0	15.3	24.6	19.8	n/a	n/a
0.5–1 mm	14.8	16.6	15.3	19.4	65.8	59.6	n/a	n/a
<0.5 mm	51.4	36.6	30.5	42.7	4.1	19.1	n/a	n/a

n/a: not applicable.
[a] Obtained in a thermobalance.
[b] Dry basis.
[c] Maximum fluidity.
[d] Calculated by difference.

2.4. Thermogravimetric analysis (TG)

Gasification was studied on a TA Instruments SDT 2960 thermobalance. Samples of weight 3–5 mg with a particle size of <0.212 mm were heated in N_2 up to 1100 °C and once the temperature was stabilized they were treated with CO_2 using a flow of 100 ml/min until a conversion degree higher than 50% was reached. The cokes/chars employed for the gasification in the thermobalance were prepared in a horizontal oven using a heating rate of 5 °C/min. The carbon conversion (x) and gasification rate or reactivity (r) were calculated be means of the following equations:

$$x = \frac{m_0 - m_t}{m_0 - m_{ash}} \times 100 \tag{3}$$

$$r = \frac{dx}{dt} \tag{4}$$

where m_0 represents the initial mass of char and m_t, the mass at time t.

2.5. Carbonization tests and coke quality determination

Carbonization tests were carried out in a movable wall oven of approximately 17 kg capacity (MWO17) [25]. The dimensions of the oven are 250 mm L × 165 mm W × 790 mm H. The samples were charged when the oven had reached 1100 °C. The temperature of the wall was kept constant throughout the test. The duration of the coking

Table 3
Apparent density of the briquettes.

	B1	BM1	$BM1_{SP}$	B4	BM4
ρ_{H2O} (kg/m^3)	1312	1179	873	1415	1149