Characterization of coal briquettes using tar as a binding material for use in a coke oven

http://dx.doi.org/10.1590/0370-44672017720151

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Abstract

In order to increase competitiveness in the steel industry through reduction of the cost of coal mixtures and increase in the quality productivity of coke in the oven, this research was developed with the aim of increasing the bulk density through briquetting of low power coking coal using tar as a binding material. Initially an industrial scale double roller press was used to manufacture briquettes in the ellipsoid format. The physical properties of the briquettes were analyzed in relation to variation of the amount of tar and the curing time of briquettes. Through the process of coal briquetting using low amounts of tar, it is possible to obtain a fuel of greater density, greater use of soft coals, uniform granulometry and easy handling. The increase in the apparent density of coal through briquetting may reach 42.5%. The success achieved in these tests, through these briquettes, with good chemical and physical characteristics represents ease of production as well as economic gain and low investment when compared to other processes of charge densification in coke ovens.

Keywords: low power coking coal, tar, briquetting, charge densification, physical properties.

1. Introduction

Charge densification processes have been used in large companies mainly in China, India and Japan. Briquetting of low power coking coal can bring benefits to coking plants. The briquettes can be used as a part of the charge of coal, increasing the bulk density and generating an increase in productivity and in the physical-chemical-metallurgical quality of the coke (Jon and Ida, 1960; Zubkova *et al.,* 2014; Montiano *et al.,* 2014; Lima, 2016).

There are several parameters and factors that influence the physical and chemical quality of the briquettes, such as the granulometry of the raw material, the briquetting temperature, the type and the quantity of the binder material, porosity, size and shape of the briquette (Rahman *et al.,* 1989; Clarke and Marsh, 1989a; Taylor and Hennah, 1991; Rubio *et al.,* 1999; Patil *et al.,* 2009; Skoczylas *et al.,* 2014).

The finer particle size produces a higher density briquette (Ellison and Stanmore, 1981a; Ellison and Stanmore, 1981b). However, it significantly increases the surface area to be moistened by the binder (Clarke and Marsh, 1989b), which may require the use of a larger amount of binder to obtain a better mechanical resistance (Pereira *at al.,* 2009). The particle size of material smaller than 2mm is an important factor for the briquetting process, where it is suggested that it must contain at least 50% by weight of the briquette and the superfine particle size lower than 0.149mm should contain a maximum of 5% by weight of the briquette charge (Pereira *et al.*, 2009).

The use of binder above the softening temperature is essential for a better compacting rate due to the increase in the speed at which the binder material moves through the interstices of the coal in view of the increased flowability of the binder (Taylor and Hennah, 1991; Rubio *et al.*, 1999).

Laboratory scale experiments on ellipsoid shaped briquettes found that for all of the briquette dimensions in three different positions, which are horizontal, vertical longitudinal and vertical latitudinal, the compressive strength is independent of the size and mass (Rahman *et al.*, 1989).

The use of soft coke in the conventional type coke oven is not more than 25% by weight of the charge. The ability to use a higher percentage of soft coal in the coal mixture, that is, the greater depletion of the coal mixture in the charge of the coke oven, becomes more efficient when using stamp-charging technology, reaching up to 55% while the maximum utilized briquetting is about 40%. However, the investment required in the use of stamp-charging technology is 10 times greater than the investment in briquetting technology. An increase in the fraction of soft coal implies a lower cost of mixing. Thus, the briquetting process when compared to stamp-charging and the conventional type coke oven becomes an efficient and inexpensive technology (Lima, 2016).

The present work is part of a more intensive study that aims at the use of a mixture of low power coking coal for the manufacture of briquettes with sufficient mechanical and chemical properties to manufacture metallurgical coke within the quality specifications of the market. Special attention was given to the influence of the amount of tar used as binding material as well as the cure time in the physical properties together with the porosity of the briquettes.

2. Materials and methods

2.1 Characterization of coal and tar

The mixture of coals and the tar used in this research comes from Gerdau Ouro Branco. The analysis of particle size of the mixture of coals was made through a methodology of Solvi Insumos. The samples, containing 500g, pass through a series of sieves based on an opening of 3.0mm, 2.0mm, 1.0mm, 0.355mm and 0.15mm with the aid of a ROTAP vibrator, remaining in the sieving system for 15 minutes.

The samples were subjected to proximate and ultimate chemical analysis using standards ISO 17246 and ISO 17247. The rank and proximate analysis of six different coals is shown in Table 1. A commercial coal tar was used as a binding material to produce the briquettes. The characteristic of this coal tar was 343.70K softening temperature, 8.70% insoluble quinoline by weight and 4.80% moisture by weight.

Coal	Fixedcarbon (%)	Moisture (%)	Ash (%)	Volatile matter (%)	Rank
1	58.07	6.80	7.12	34.81	High volatile bituminous
2	70.47	6.72	8.46	21.07	Low volatile bituminous
3	68.79	7.34	9.50	21.71	Medium volatile bituminous
4	62.07	7.47	10.06	27.87	Medium volatile bituminous
5	65.33	6.84	9.27	25.40	Medium volatile bituminous
6	83.80	6.85	3.70	12.50	Low volatile bituminous

Table 1
Proximate analysis of coals.

2.2 Briquetting

The technique of coal briquetting in a double-roll press machine involves particle size distribution, proportional mixing of binding material, compacting and drying. A schematic of the briquetting process is shown in Figure 1.

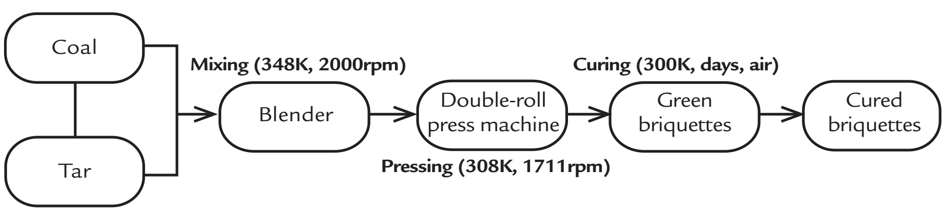

Figure 1
Schematic of briquetting process.

The investigated coals were mixed with different amounts of preheated tar at a temperature of 343.7K using a mixer at a speed of 2000rpm. Thereafter, the blend was compacted in a double-roll press machine. The rollers are symmetric and mounted facing each other and rotate with a speed of 1711rpm in opposite directions. Each briquette was made by pressing a 25g mass of mixture of coal and tar into a double-roll press machine with the dimension shown in Figure 2.

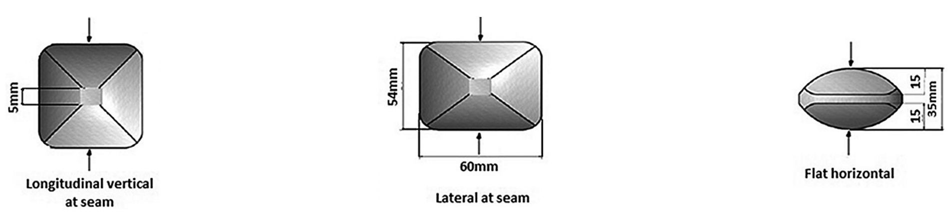

Figure 2
Dimension of briquettes and axial directions in compression testing.

After compacting the material, the green briquettes were ejected from the mold in an ellipsoid shape. Four different briquettes were produced by varying the percentage of tar from 4 to 7% by weight used as binding material. The notation of the briquettes was identified as B followed by the percentage of tar by weight.

2.3 Mechanical testing

The compressive strength test was performed on the three different dimensions of briquettes, as shown in Figure 2, using an Amsler Frères compression testing machine with a 20kN operating capacity, using an adaptation of NBR 5739 standard. The influence of the amount of binding material and the curing time on the physical resistance of briquettes was analyzed. The results were reported as the maximum load supported by the briquette before fracture. The test is used to determine the physical resistance of the briquettes to compressive stresses during storage and handling (Luz *et al.,* 2010).

The impact resistance testing measures the degradation of the briquette simulating the falls that it undergoes during handling and inside the coke oven (Rubio *et al*., 1999; Luz *et al.,* 2010). Each sample suffers three consecutive drops from a height of 3m above the concrete floor. The equipment used is a conveyor belt for Stemmann impact testing. After the test, the material passes through a series of sieves based on an opening of 25mm, 10mm and 5mm. From these results, the impact resistance index (IRI) was calculated according to Equation 1.

$$IRI = \frac{M_2}{M_1} * 100 \qquad (1)$$

Where M_2 is the mass retained above the 10mm sieve after the test and M_1 is the initial mass of the test.

The water resistance testing on briquette consists of immersing the briquette in a vessel with water for 2 hours.The water absorption is then calculated by measuring briquette mass before and after immersion in the water (Richards, 1990; Cunha *et al.,* 2006).

2.4 Real density

The determination of real density of briquettes and coal was carried out using a Quantachrome Instruments model Ultrafon multipicnometer. The analysis was done by placing the sample inside the equipment in which a helium gas with 18psi pressure is passed, capable of penetrating into pores in the order of $1x10^{-10}$m. The determination of the real density expressed by D, Equation 2, is calculated as a function of the weight of the sample (W, in grams) and the volume of the powder (V_p, in cm^3).

$$D = \frac{W}{V_p} \qquad (2)$$

The volume of the powder (V_p), Equation 3, is calculated as a function of the sample cell volume (V_c in cm^3), the reference volume (V_r, in cm^3), the pressure after minimum pressure on the volume (P_1, in psi) and pressure after inclusion of the sample cell volume (P_2, in psi).

$$V_p = (V_c - V_r) * \left[\left(\frac{P_1}{P_2}\right) - 1\right] \qquad (3)$$

2.5 Porosity

The technique of porosity analysis was done through a high resolution computerized microtomography system SkyScan X-ray microtomograph Bruker model 1272. The samples of the briquettes were made in cylindrical format with dimensions of 10mm in diameter by 20mm in height using a saw blade and sandpaper. Then the sample was conditioned inside the equipment at room temperature upon which data was collected for 7 hours. The system visualizes virtual slices of up to 209 Megapixel through the samples using the X-ray detectors with detection details of 0.35μm and 0.45μm in up to three automatic positions, 5μm pixel size, 80kV voltage and 7W power. The technical procedure of microtomography analysis consists of collecting X-ray projection images at different angles of the sample using a SkyScan 1272 software and converting this set of images and sections representing a three-dimensional image. This image was analyzed by CTAN software (Comprehensive Tex Archive Network) in order to obtain the volume of interest and the porosity of the sample.

3. Results and discussions

Since the objective of this study was to obtain mechanically strong briquettes in order to facilitate handling, transport, storage and use as raw material in the coke production, three aspects had to be considered in this study: the mechanical properties, porosity and the minimum percentage by weight of tar used as a binding material. Chemical and physical analysis data of mixture of coals are shown in Table 2.

Particle size distribution		Coal		Ash	
Particle size (mm)	% by weight	Proximate analysis		SiO_2 (%)	54.80
3>x>2	28.61	Moisture (%)	7.1	Al_2O_3 (%)	27.12
2>x>1	32.83	Ash (%)	6.18	TiO_2 (%)	1.48
1>x>0.355	35.8	Volatile matter (%)	22.06	Fe_2O_3 (%)	6.92
0.355>x>0.149	2.74	Fixed Carbon (%)	71.76	MgO (%)	0.76
<0.149	0.02	Ultimate analysis		CaO (%)	1.84
Total	100	Carbon (%)	73.6	Na_2O (%)	0.51
		Hydrogen (%)	4.51	K_2O (%)	1.17
		Oxygen (%)	3.34	P_2O_5 (%)	0.98
		Nitrogen (%)	1.7	ZnO (%)	0.03
		Sulfur (%)	0.76	MnO (%)	0.04

Table 2
Particle size distribution, proximate and ultimate analysis of mixture of coals.

The compressive strength testing was done by starting a load perpendicular to the three different positions of the briquette. An average of five measures was taken every five days to obtain the results. Compression strength is plotted versus the curing time of briquettes as shown in Figures 3, 4 and 5.

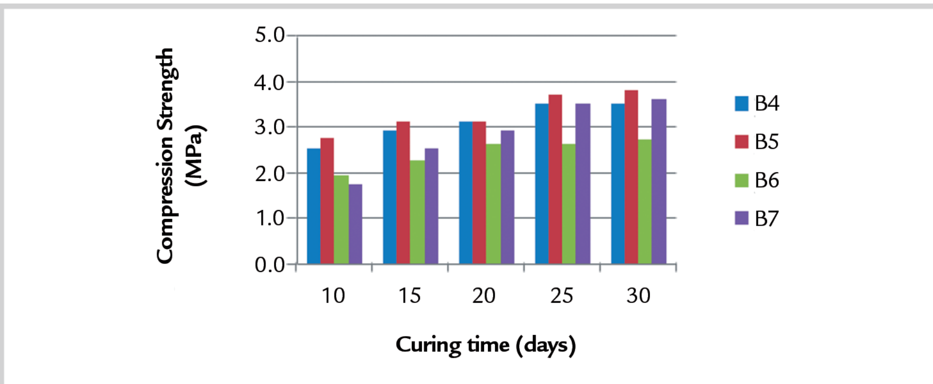

Figure 3
Compressive strength of briquettes in a longitudinal vertical at seam position as a function of curing time.

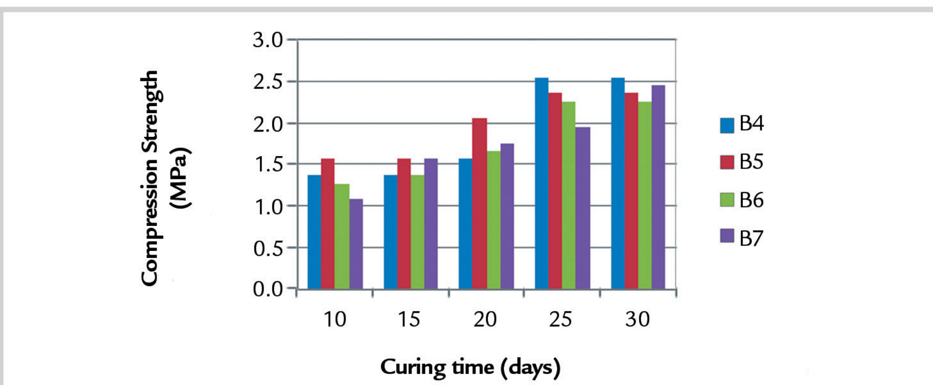

Figure 4
Compressive strength of briquettes in a lateral at seam position as a function of curing time.

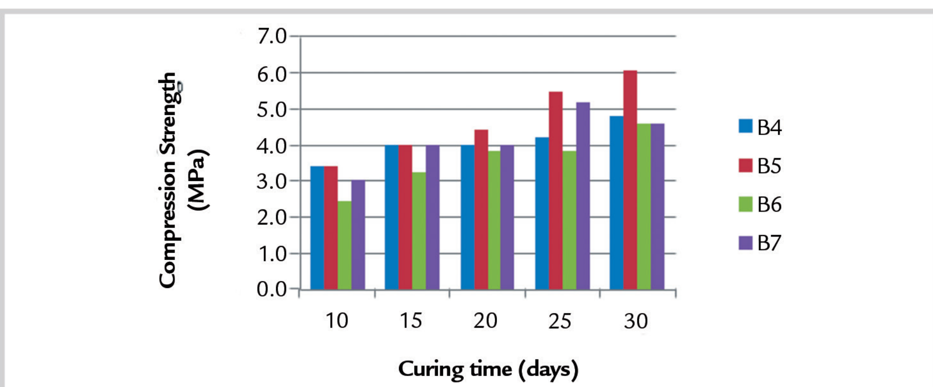

Figure 5
Compressive strength of briquettes in a flat horizontal position as a function of curing time.

From these figures, it can be seen that the compression strength of the briquette falls from a 6% tar by weight present in the briquette (B6). This fact can be explained by a lower agglomeration capacity, coming from a very thick layer of tar, causing a weak bond between the coal particles (Taylor and Hennah, 1991; Rubio *et al.*, 1999). Low amounts of binding material in the briquettes lead to higher pore sizes (Rubio *et al.,* 1999) also decreases their strength as shown in sample B4. For the analysis of compression strength, the best percentage of tar in the briquette corresponds to 5% by weight (B5). The curing time favors the increase in briquette compression strength, showing a significant increase after 20 days. Curing time assists in eliminating the moisture of the briquette, thereby increasing its resistance (Patil *et al.*, 2009).

The impact resistance index (IRI) is plotted versus the amount of tar after 30 days of cure, Figure 6. The target value for IRI is over 90% (Luz et al., 2010). The impact resistance of the briquettes was better for the briquette containing 5% tar by weight with a yield of 96.67%. Values below or above the amount of 5% tar by weight in the briquette did not produce very satisfactory results, since the degradation of the materials was greater than 10%. These values of impact resistance directly linked to the results of the compression strength were expected, since both are part of the analysis of mechanical resistance of briquette.

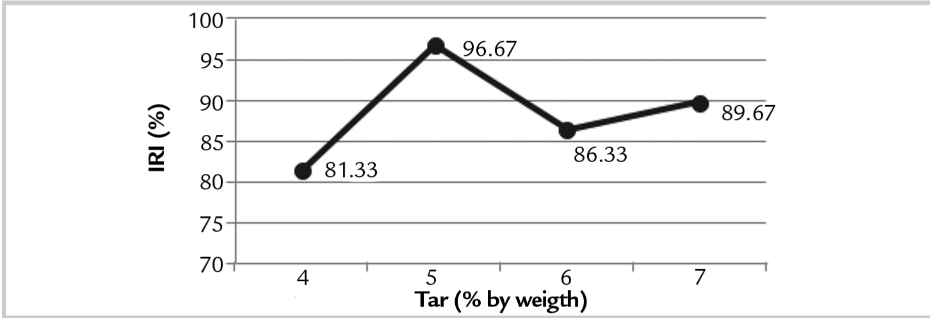

Figure 6
Impact resistance index of briquettes.

When briquetting is made using water-insoluble binding material, such as tar, the briquettes are generally water-resistant. Table 3 displays the water resistance testing results after 2 hours.

Table 3
Water absorption in briquettes.

Sample	Water absorption (%)
B4	1.92
B5	1.59
B6	1.76
B7	1.61

The water resistance testing shows the briquette debonding and also its possible filling. Excess water causes increased coke production costs. It would require more time and heat to remove the moisture in the coking process (Richards, 1990). Also, in the transport of the briquette, since it is hygroscopic and this would imply in unnecessary addition of weight. A maximum value of 5% water absorption would be a reasonable target for most coal briquettes (Richards, 1990). Water absorption was favorable for all briquette samples having an average amount of absorbed water of less than 2%.

The porosity, in addition to other factors, is directly related to the physical resistance of the briquettes. Briquettes with porosity above 24% have low tensile and compressive strength (Skoczylas *et al.*, 2014). Table 4 shows values of the density and porosity of the briquettes. Density and porosity of the samples have a proximity of values, but the sample B4 presented greater porosity. This sample resulted in worse mechanical resistance results. All briquettes have lower porosities and higher density in relation to coal. The density of raw material for coke production can increase by up to 42.5%, but the use of these briquettes to produce coke is limited up to 30% in the charge, due to the increase in expansion pressure during carbonization, which could lead to degradation of the coke oven furnace wall (Lima, 2016).

Table 4
Density and porosity analyzes of briquettes and coal.

Sample	Density (g/cm^3)	Porosity (%)
Coal	0.900	20.52
B4	1.270	18.29
B5	1.283	16.57
B6	1.280	17.65
B7	1.274	17.02

4. Conclusions

Through this study, it can be concluded that the mechanical strength (compression and impact) of briquettes made by low power coking coal and tar depends on the amount of binding material and also on the curing time. All samples proved to be effective in the water resistance testing. The mechanical strength of the briquettes increases with the curing time, with an optimal value in the period of 25 to 30 days. The best results were obtained using briquettes made by 5% tar by weight, which leads to this quantity being an optimum value for coal briquetting in order to produce a briquette with the lowest possible amount of binding material and, at the same time, an excellent raw material for coke production, having the highest density among the others briquettes due to the lower porosity presented. The density of briquettes was higher than the density of coal, showing that it is possible to increase the density in the coke oven, through the partial use of these briquettes leading to increase in productivity.

Acknowledgments

The authors would like to acknowledge Gerdau Ouro Branco, Solvi Insumos, CEFET-MG unit Timóteo, UFOP, REDEMAT and CAPES for supporting the development of this study.

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Received: 9 October 2017 - Accepted: 5 April 2018.