



Influence of low-density polyethylene addition on coking pressure



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HIGHLIGHTS

- Coke production is a viable option to recycle plastics as secondary raw materials.
- Coking pressure rises for low LDPE addition (≤ 3 wt.%); higher amounts reduce pressure.
- LDPE influences the pyrolysis process and the swelling process of the plastic stage.
- A delay in the LDPE degradation is confirmed by DRIFT and SEM.

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ABSTRACT

Different amounts of low-density polyethylene (LDPE) were added to a bituminous coal used to produce metallurgical coke. The effect of the plastic waste on the carbonization process and more exactly, on the coking pressure were investigated. A movable wall oven at semi-pilot scale was used for measuring coking pressure generated. It was found that coking pressure increases for low LDPE addition levels (1–3 wt.%); however higher amounts of LDPE reduce coking pressure. To explain this behavior different blends of the coal and the residue were pyrolysed at three different temperatures (450, 500 and 600 °C) in a Gray-King apparatus. The results show that LDPE causes a modification in the pyrolysis process and also influences the swelling process of the plastic stage.

The increase of the coking pressure at low LDPE addition rates is associated with a less permeable coal plastic layer, which prevents the removal of the decomposition products and causes their retention in the semicoke matrix, evolving them in the post-plastic stage. Coking pressure decrease at high LDPE addition rates can be due to the charge shrinkage and the better permeability to the migration of oil components, which suggest a lower interaction between the coal and the LDPE. A delay in the degradation of LDPE is confirmed by the data provided by DRIFT and SEM.

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1. Introduction

The environmental impact due to waste production, both domestic and industrial, has become a high priority part of environmental policies in developed countries. Even though waste management has undergone a significant evolution in the recent decades, waste recovery continues to be of great importance to achieve sustainable development and compatibility with environmental protection. It has been necessary to allocate time and resources to develop processes to ensure plastic recycling is an economically profitable process, helping to decrease the amount of residue designated to landfill disposal. As a result of recent European legislation for management and recovery of plastic

wastes from packaging, the development of new processes for mechanical and chemical recycling and energy recovery are being promoted in order to achieve the recycling objectives.

The use of plastics as secondary raw materials in metallurgical coke production is an environmentally friendly alternative for recycling municipal plastic wastes. These wastes can be added as minor components to the coal that is used as feedstock in this industrial process [1–3]. Depending on the composition of plastic waste, a different effect can be expected on the fluidity of the coal or coal blends, the semicoke structure and the structure and properties of high temperature cokes [3–9]. Previous investigations have shown the addition of plastic wastes induces a decrease in the Gieseler maximum fluidity. The extent of this reduction depends on the amount, structure and thermal behavior of the plastic residue added [5–9]. Polyolefins (LDPE, HDPE and PP) reduce the fluidity development to a lesser degree, while polymers containing aromatic rings in their structure (PS and PET) significantly decrease the fluidity of the coal. The interactions between coal and plastics also cause modifications on the optical structure of the semicokes,

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decreasing the anisotropic development and the size of the anisotropic components [9]. Moreover, certain types of plastic waste such as polyolefins can be incorporated, in small amounts, into typical coking blends as secondary raw materials without notably modifying coke properties. Preliminary results obtained at a semi-industrial scale at INCAR show that a coking coal blend can tolerate up to 3 wt.% of polyethylene waste without any significant deterioration of the coke quality parameters [3]. Although the quality of the coke is maintained or slightly improved when polyolefins are added in small quantities, single polyolefins or wastes rich in them increase the coking pressure during the process, when there are added at a low addition rates, becoming dangerous carbonizations [10,11]. The mechanism that generates coking pressure is not well understood. The generation of high coking pressure by some coals is due to the combination of two phenomena; the release and the characteristics of the volatile matter evolved and the ability of gas to escape through the plastic layer. Previous authors have related the internal pressure to several factors; the permeability of the plastic layer [12–14], the emission of volatile matter and the plasticity of the plastic layer [15] and the fissure pattern of the semicoke and the pore structure [16–20].

In order to avoid the negative effects of the polyolefins on the coking pressure, different approaches have been proposed. Possible alternatives include carrying out carbonization at a lower bulk density, subsequently reducing yield and quality of the obtained coke; adjusting the amount of polyolefins in the waste [11] or recycling organic wastes and lubricating oils of different origins by adding these wastes to typical coal blends and plastic waste mixtures [21–23].

To investigate the effects of the amount of low density polyethylene (LDPE) on coal fluidity development, coking pressure generation and the coking pressure mechanism, Gray King pyrolysis was carried out at three different temperatures using a coking coal and its blends with up to 10 wt.% additions of LDPE. The structure and morphology of the semicokes and the study of obtained tars is a useful way to elucidate the mechanism of interaction between coal and polyethylene in order to explain the effect of the polyolefins on coking pressure. LDPE was chosen as being representative of the polyolefins contained in municipal, agricultural or other different sectors.

2. Materials and methods

Previous results using different coals and coal blends have shown that single polyolefins (LDPE, HDPE and PP) increase the pressure exerted against the wall in the course of coking process at low addition rates [10,11]. In this work it was decided to use an individual coal instead of a blend in order to isolate the effect of the LDPE on the coking pressure. The coal G has been selected for this study due to its similarity to industrial blends used in the coking industry in terms of volatile matter and fluidity [21,22]. Proximate analysis of the coal was performed following the ISO562 and ISO1171 standard procedures for volatile matter and ash content, respectively. The elemental analysis was determined with the aid of a LECO CHN-2000 for C, H and N, a LECO S-144 DR for sulfur and oxygen was estimated by difference. The main characteristics of the single coal G are presented in Table 1.

Low-density polyethylene (LDPE) from agricultural greenhouse films was selected for this study as it can be taken as representative of polyolefins from other different post-consumer sectors. The nomenclature used in this study is as follows: G is the individual coal, followed by a number corresponding to the addition rate (wt.%) and followed by the initials of the waste, in this case LDPE.

The coking coal and its mixtures with LDPE were carbonised in a semi-pilot moveable wall oven of 15 kg capacity (MWO15). The coking time lasted nearly 3 h with the temperature in the centre

Table 1
Main characteristics of the coal G.

	Coal G
VM (wt.% db)	21.2
Ash (wt.% db)	9.0
C (wt.% daf)	90.6
H (wt.% daf)	5.0
N (wt.% daf)	1.8
S (wt.% daf)	0.6
O (wt.% daf)	2.0
Maximum Gieseler fluidity (ddpm)	423

of the charge reaching a maximum of 950 °C by the end of the process. The MWO15 is described in detail elsewhere [21].

The thermoplastic properties of the coal and its blends with up to 10 wt.% LDPE were tested in a Gieseler plastometer, using the R.B. Automazione model PL2000 and following the ASTM D 2639 standard procedure. The specific parameters for this test are: (i) the softening temperature at which the coal starts to be fluid (T_s); (ii) the temperature of maximum fluidity reached during the thermal heating (T_f); (iii) the temperature at which the fluid mass resolidifies into a semicoke (T_r); (iv) the plastic or fluid range, which is defined as the difference between the resolidification and softening temperatures ($T_r - T_s$); (v) F_{max} , the maximum fluidity, expressed as dial divisions per minute (ddpm).

Samples of the individual plastic (LDPE) and the bituminous coal G were subjected to thermogravimetric analysis (TGA) in a simultaneous TA instrument SDT2960 analyzer. 10 mg of plastic was heated from room temperature up to 600 °C at a heating rate of 3 °C min⁻¹ using a nitrogen flow rate of 100 ml/min to sweep up volatile products. For the coal, the final temperature of the TGA run was 1000 °C.

The coke yields for the coal G and its blends with LDPE were calculated as the mass percentage of residue after heat treatment at 1000 °C at a heating rate of 3 °C/min by means thermogravimetric analysis.

The quality of the resultant cokes was assessed in terms of reactivity towards carbon dioxide at 1100 °C (CRI) and mechanical strength of the partially-gasified coke (CSR) using the Nippon Steel Corporation (NSC) method [24], according to ASTM D5341 standard procedure. Coke reactivity (CRI) was measured as the mass loss of coke after reaction with CO₂ at 1100 °C for 2 h in a dried sample of 200 g with a particle size between 19 and 22.4 mm. The partially-gasified coke was subjected to a mechanical treatment of 600 revolutions at 20 rpm. The amount of coke with a particle size larger than 9.5 mm after mechanical treatment is referred to as the CSR index. The limit values for a good quality coke are CRI < 30 and CSR > 60 [25]. The lower the CRI index and the higher the CSR index, the better the coke quality is.

The pyrolysis of the coal and its blends with LDPE (8 g, <0.212 mm) was also carried out in a Gray King oven at three temperatures, 450, 500 and 600 °C under the atmosphere of evolved gases, applying a heating rate of 5 °C/min and a soaking time of 5 min. The initials GK mean Gray-King pyrolysis and they are followed for the final temperature of the process (GK450, GK500 and GK600).

The design of the Gray King reactor is described in the standard procedure ISO502 modified by Suarez Ruiz et al. [26]. After leaving the reactor, the volatile products were condensed in a trap cooled by an ice salt bath (primary tar). The non-condensable fraction was removed from the reactor by means of an outlet tube. The solid carbon material (semicoke) was removed from the oven after cooling at room temperature.

The obtained semicokes were characterized by elemental analysis, Fourier transform infrared spectroscopy using the diffuse reflectance mode (DRIFT) and scanning electron microscopy (SEM).

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