Fuel 165 (2016) 134–144

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Modelling the Gieseler fluidity of coking coals modified by multicomponent plastic wastes

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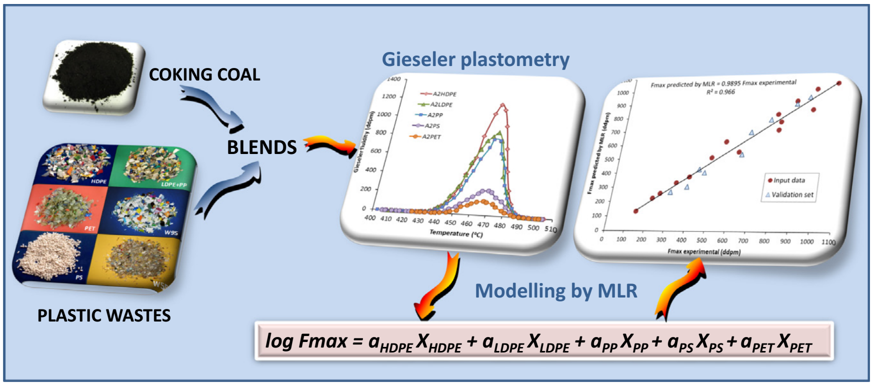
HIGHLIGHTS

- The effect of single/mixed plastics on the fluidity of coking coals is analyzed.
- The composition of plastic wastes is associated to a reduction in coal fluidity.
- A five-parameter model for fluidity prediction was obtained by MLR analysis.
- Validation data showed that the MLR model has good predictive ability.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 31 July 2015
Received in revised form 18 September 2015
Accepted 14 October 2015
Available online 23 October 2015

Keywords:
Coking coal
Plastics
Household packaging wastes
Feedstock recycling
Gieseler fluidity
Cokemaking

ABSTRACT

A novel method for predicting the Gieseler maximum fluidity (F_{max}) of a coal + plastic mixture formed from a relative proportion of the plastics present in a multicomponent waste is proposed. A training set of five most-common thermoplastics in household wastes (HDPE, LDPE, PP, PS and PET), binary and ternary plastic mixtures was used to construct multivariable linear regression (MLR) models. Validation was conducted by means of an external set of mixed plastics and real unsorted plastic wastes. The results obtained from the numerical solution of the MLR models were found to be in satisfactory agreement with the experimental data obtained using a Gieseler plastometer. The F_{max} values fitted the models with determination coefficients of >0.96 and root mean square errors of prediction of 0.048 and 0.058. All the plastic mixtures tested represented a wide spectrum in concentration of the five polymers contained in municipal plastic wastes and a global plastic addition of 2 wt% to the coal was always used. The starting point for this study was to determine the effect of each single plastic on the reduction in fluidity of various coking coals and an industrial coking blend. Afterwards, the exponential functions of F_{max} of the blends of coal and binary/ternary plastic mixtures were useful to analyze the changes in Gieseler F_{max} with varying proportions of components. Based on the results, the coal responses were statistically treated and MLR models were developed.

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1. Introduction

During thermal treatment up to a temperature of around 500 °C in an inert atmosphere, coking coals have one unique property that makes them different from all other coals. When heated at a

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http://dx.doi.org/10.1016/j.fuel.2015.10.053
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relative slow rate of 3 °C/min the solid particles of coal soften and pass through a transient plastic/fluid phase in a narrow temperature range of between 400 and 500 °C, which results in the caking and agglomeration of particles. This physical phenomenon of coking coals is also accompanied by chemical changes in their macromolecular structure, involving the development of molecular-reordering in carbon network before the resolidification of the fluid mass to form a low-temperature graphitizable carbon material (semicoke) [1–5]. The fluidity of a coal or a blend of different coals measured by a Gieseler plastometer has been long recognized and adopted as a key parameter in the evaluation of feedstocks for cokemaking. Any modification of this property caused by the incorporation of additives to an industrial coal blend affects the structure of the semicoke and, in turn, the structure and properties of the high-temperature coke [1–5]. Thus, the degree of compatibility of a given additive with a coal is related to its interaction in the molten or semi-molten state of the coal.

Among non-conventional additives for cokemaking, the effects of individual or mixed polymers from household wastes or other post-consumer sectors on coal fluidity have been discussed in a number of previous studies [6–17]. Of the major thermoplastics contained in household wastes, polyethylene terephthalate (PET) and polystyrene (PS) have been shown to be the strongest inhibitors of the development of fluidity of coking coals, while polyolefins such as high- and low-density polyethylenes (HDPE and LDPE, respectively) and polypropylene (PP) have a more moderate effect on this coal property. The decrease in maximum fluidity is thought to be connected with a combination of: (a) the relative position of the thermal degradation of the polymeric chain in respect to the fluid temperature range of coal [8–14]; and (b) the chemical capacity to participate in the H-transfer reactions as a consequence of the change in chemical structure of the polymeric chain [8,12]. Thus, single PET and PS can be considered as the greatest incompatible additives for cokemaking, because these polymers have a high capacity for accepting hydrogen in the free-radical stabilization process and, in particular, PET promotes chemical cross-linking reactions [12]. Reaction with 9,10-dihydroanthracene (a hydrogen-donor agent) clearly differentiates plastics in terms of hydrogen acceptor ability in the following increasing order: $\mathrm{PP} < \mathrm{LDPE} < \mathrm{PS} < \mathrm{HDPE} \ll \mathrm{PET}$ [12]. Furthermore, the thermal degradation of PS and PET occurs in the final step of the pre-plastic stage before, or close, to the onset temperature of the softening process of coal [8–14]. As a consequence, when PS and PET are added to coking coals at a rate of 10 wt% they cause a greater modification of the optical components in semicokes obtained at 500 °C [12,13]. The addition of these polymers produces more isotropic carbon and small-sized anisotropic components of the mosaic type. Moreover, an addition rate of 10 wt% of PET transforms a coking coal into a non-coking coal, destroying its ability to be fluid [8,12,13].

A different thermal behavior was observed for polyolefins. The degradation of the three polyolefins – HDPE, LDPE and PP – takes place inside the temperature range of the fluid state of coking coals [8–14] and they seem to have a more balanced ability to donate and accept hydrogen during co-carbonization [12].

As a consequence of the thermal behavior of the individual plastics, an order from a higher to a lower reduction in Gieseler maximum fluidity ($F_{\max}$) with the addition of a plastic waste has been established [8,12]:

$$\mathrm{PET} > \mathrm{PS} \gg \mathrm{PP} > \mathrm{LDPE} \geqslant \mathrm{HDPE}$$

Not only have these two groups of polymers a different role in determining the thermoplastic properties of coking coals, but also they differ in the effects they have on the generation of internal gas pressure during coking and on the quality parameters of coke to be used in a blast furnace [16–28]. It has been reported that PS and PET have no significant negative effect on the generation of pressure [16,19,20], but they have a detrimental effect on the quality parameters of coke – negative effect for cokemaking –, while polyolefins have the opposite effect: they increase the pressure generated and maintain or slightly improve the coke quality [16–20,23–25]. In this context, the advances made in plastic waste recycling in cokemaking have been described in detail in two recent reviews [27,28].

As a consequence of such effects the overall conclusion to be drawn is that the relative proportions of the two polymer groups needs to be controlled. Although there is no universal criterion that can be applied to all coking blends and industrial plants, a relative proportion of polyolefins to the PS–PET pair of 65:35 w/w for a total amount of mixed plastic waste of 2 wt% has been established as the limit required to maintain the coking pressure within safe values and to avoid any detrimental effect on coke quality [17].

The degree of the reduction in fluidity seems governed almost entirely by the PS–PET pair, but especially by PET. Therefore, these additives need to be included with caution since the reduced fluidity may have a detrimental effect on coke quality. Much of the published work on the modification of thermoplastic properties of coking coals induced by plastic wastes have been carried out using single plastics [8–15] or a reduced number of mixed plastic wastes [13,15–28], but there is a lack of sufficiently detailed knowledge about how the maximum fluidity of coking coals is affected by the composition of plastic waste and about the effects of the characteristics of the coal employed in the preparation of the blend. In this context lie our goals: (i) to examine coal response during the development of fluidity paying particular attention to how single plastics and simple mixtures affect fluidity; (ii) to determine the behavior of coal when mixed with multicomponent wastes containing the five most common thermoplastics in household streams; and (iii) to develop a mathematical model to estimate the extent of fluidity reduction as a function of the composition of plastic waste.

2. Experimental

2.1. Coal and wastes

Three coking coals (A, B and C) of different rank, maceral composition and rheological properties were selected for the preparation of several mixtures either with common single thermoplastics or real unsorted wastes from household packaging streams. Additionally, a coal blend (M) composed of ten coals from Australia and USA of different rank and rheological properties, employed for the industrial production of blast-furnace coke, was used. The coals were subjected to conventional characterization following the ISO562 and ISO1171 standard procedures for volatile matter and ash contents, respectively. Sulfur determination was performed using a LECO S-144 DR, according to the experimental protocol described in ASTM D-5016. A petrographic examination of the samples was carried out on a MPV II Leitz microscope under reflected white light using immersion objectives (32×), in accordance with the ISO 7404-5 procedure for vitrinite reflectance and the ISO 7404-3 procedure for maceral group determination.

The plastic wastes mixed with the coals and the coal blend at an addition rate of 2 wt% were: (a) five of the most commonly thermoplastics in household packaging streams: high- and low-density polyethylene (HDPE and LDPE, respectively), polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET); (b) a mixture of LDPE and PP (50:50 w/w); (c) binary and ternary mixtures of multicolor HDPE with PET and the mixture LDPE/PP with PS and PET; (d) multicomponent wastes provided by the Spanish recycling company ABORNASA (W95, W93 and W55) and,

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