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Full Length Article

Effect of gasification agent on co-gasification of rice production wastes mixtures

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highlights are the control of the control of

Co-gasification of biomass rice wastes blended with polyethylene (PE).

 \bullet Use of mixtures of air, oxygen steam and CO₂ with different compositions of two or more components as gasification agent.

 \bullet Study of CO₂-blown gasification.

 \bullet Effect of gasification agent on the release of H₂S and NH₃.

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Rice production generates different types of wastes, rice husk and straw and plastics, mainly polyethylene (PE) used in bags for rice packaging and for seeds and fertilizers transport. Due to their contamination they are not suitable for physical recycling and end-up in landfills. Biomass rice wastes may have some utilisations, but each of them has drawbacks. This paper studies the possibility of using fluidised bed co-gasification of all these wastes for their energetic valorisation. Gasification gas composition and heating value is affected by the gasification and fluidisation agent. Mixtures of air, oxygen, steam and CO₂ with different compositions of two or more components were tested. Besides the high cost of producing oxygen, the results obtained showed that the best technical option was the use of steam and oxygen, because the gas was not diluted in nitrogen and thus gas HHV (higher heating value) on dry basis increased around 42%. However, the use of enriched air with up to 40% (v/v) of oxygen may be an alternative, due to the lower cost of producing this gas and also because better results were obtained than those for air-blown gasification. The use of $CO₂$ as gasification and fluidisation agent may be a good route, as the results obtained showed that $CO₂$ reforming reactions were promoted. The increase of $CO₂$ content in gasification agent led to a decrease of 45% in tar content, which was followed by a great increase in gas yield, around 70%. The main drawback of using steam and $CO₂$ is the need to supply the energy for the endothermic reactions, hence a good option could be the use of mixtures of $CO₂$, $O₂$ and steam as gasification agent.

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1. Introduction

Worldwide rice production is about 700.7 million tonnes per year, which produce around 100 million tonnes of rice husk annu-ally [\[1\].](#page--1-0) Rice straw and rice husk are produced in Portugal in significant amounts, but currently, there is no energy recovery of these wastes. In Portugal the production of rice straw was around 37.7 kt year⁻¹, while rice husk was about 23.7 kt year⁻¹ in 2013. The estimated amount of plastics produced due to rice production was around 300 t year⁻¹ [\[2\]](#page--1-0), the main plastic was polyethylene

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<http://dx.doi.org/10.1016/j.fuel.2016.04.048> 0016-2361/@ 2016 Elsevier Ltd. All rights reserved. (PE), used to transport seeds, fertilisers and pesticides which are contaminated with chemical products that prevents their physical recycling. Biomass rice wastes are sometimes incinerated at openair or deposit in landfields, which are not environmentally desirable. Other options are the incorporation in animal food, which is not a good option, due to the high contents of silica, or the use as animal bedding, but again the final disposal is incineration or landfilling. The high energetic content of plastics and biomass rice wastes makes attractive their valorisation by thermochemical processes, namely co-gasification for the production of gaseous biofuel.

There is already some knowledge about gasification of rice husk [\[3–6\],](#page--1-0) nevertheless the presence of plastics brings some challenges.

Calvo et al. [\[3\]](#page--1-0) gasified rice straw in an atmospheric fluidized bed at a temperature up to 850 \degree C and found that it was possible to produce high quality syngas, some bed agglomeration could be avoided by using a mixture of alumina silicate sand and MgO as gasification bed. Murakami et al. $[4]$ used rice straw in a twostage process, hydrothermal treatment was done in the first one and in the second stage steam gasification in a fixed-bed reactor using nickel as catalyst was performed. Yoon et al. [\[5\]](#page--1-0) gasified rice husk in a bench-scale downdraft fixed-bed gasifier and observed that the heating value of the gas and the cold gas efficiency were higher when rice husk was used as pellets.

Fluidised bed gasification is usually a good option to deal with heterogeneous feedstocks like those obtained during rice production. Gasification gas heating value and composition depends on gasification conditions and mainly on the gasification and fluidisation agent, air, oxygen steam and $CO₂$ may be used either alone or in mixtures of two or more components.

As one of the most promising technologies available for energy conversion, air–steam gasification leads to a gas with low heating value, which decreases its potential value, due to the dilution effect of nitrogen [\[7\].](#page--1-0) Air could be replaced by pure oxygen, but the cost of oxygen production for the gasification installation would reduce its economic viability. A compromise option would be the use of enriched air, with oxygen amounts higher than that found in air, as nitrogen dilution effect would be reduced. One of the preferred compositions would be enriched air with 40% (v/v) of oxygen, as this concentration can be produced at a lower cost using membrane technology [\[7\].](#page--1-0)

Campoy et al. [\[7\]](#page--1-0) reported that the use of enriched air with 40% of oxygen and a steam/biomass ratio around 0.3, increased CO and H2 yields, heating value and carbon conversion. A maximum efficiency of 70% was obtained [\[7\].](#page--1-0) Cheng et al. [\[8\]](#page--1-0) also observed that an increase of the oxygen concentration from 21% to 31.4%, increased gas yield, carbon conversion efficiency and gasification efficiency. Under the best operating conditions, the maximum LHV (lower heating value) of the produced gas reached 6200 kJ/ Nm^3 [\[8\]](#page--1-0). Huynh et al. [\[9,10\]](#page--1-0) also reported that increasing oxygen amounts from 21% to 40% (v/v) during gasification of three kinds of wood caused a great increase in $H₂$ production and a smaller one in CO. Thus, an increase in gas LHV of more than 43% was observed [\[9\].](#page--1-0) However, significantly higher amounts of steam were required to control the reactivity of the system at high oxygen levels [\[10\].](#page--1-0) Thanapal et al. [\[11\]](#page--1-0) also observed an increase in CO, H_2 and CH₄ concentrations, while CO₂ decreased when enriched air with 28% (v/v) of oxygen was used for the gasification of dairy biomass wastes.

Wang et al. [\[12\]](#page--1-0) reported that biomass gasification using enriched air up to 99.5% (v/v) of oxygen, but without steam, led to increases in H₂ + CO from about 30% (v/v) to more than 70% (v/v) , but H₂/CO ratio decreased for the highest oxygen concentration. Enriched air with oxygen generated a syngas with higher LHV and higher carbon conversion efficiency, but with a lower yield, corresponding to lower amount of nitrogen on the obtained gas [\[12\]](#page--1-0).

Due to the need of obtaining a viable low $CO₂$ emission energy source, several studies have proposed the use of $CO₂$ as a gasification agent, as it could act as gasification promoter, through gas– solid Boudouard reaction, acting in the solid carbon from the feedstock and through $CO₂$ reforming reactions of the hydrocarbons initially formed. Butterman et al. [\[13\]](#page--1-0) studied biomass gasification with different amounts of $CO₂$ mixed with steam. The use of $CO₂$ significantly enhanced CO evolution and reduced H_2 and CH₄ concentrations. The same authors also reported the increase in carbon conversion, probably due to the ability for $CO₂$ to enhance the pore structure of the residual carbon skeleton after devolatilisation, providing access for $CO₂$ to efficiently gasify the solid [\[14\].](#page--1-0)

Cheng et al. [\[15\]](#page--1-0) observed that by increasing $CO₂/b$ iomass ratio, the mole fraction of CO in the gasification gases increased, while H_2 and $CO₂$ decreased. When $CO₂$ mass percentage in the gasifying agent reached 60%, the fractions of CO and $CH₄$ attained the maximum, as well as gas LHV and cold gas efficiency. Hanaoka et al. [\[16\]](#page--1-0) studied the gasification of aquatic biomass using O_2/CO_2 mixtures with different proportions. The increase in $CO₂$ content led to the increase in the conversion to gas and enhanced CO release, while $H₂$ content decreased.

Several researchers $[11,12,16-18]$ stated that $CO₂$ has a positive effect on char gasification, due to the promotion of the gas–solid Boudouard reaction. These conclusions are corroborated by Huo et al. [\[17\]](#page--1-0) who compared the reactivities of different origin chars during gasification with $CO₂$. Biomass chars with higher BET surfaces than coal or coke char, presented the highest reactivity.

Kirtania et al. [\[19\]](#page--1-0) compared gasification of chars, obtained from wood and algae, with steam or $CO₂$. A significant difference in gasification reactivity, which was attributed to char structure, was reported. These results agree with those presented by Nilsson et al. [\[20\]](#page--1-0), who reported that the char gasification rates measured in a mixture containing both $CO₂$ and $H₂O$, were well approximated to the sum of the individual rates measured with only $CO₂$ or $H₂O$. Guizani et al. [\[21\]](#page--1-0) studied the diffusion–reaction competition for steam and $CO₂$ in the gasification of biomass chars and reported that H_2O had an almost twice higher reactivity and diffusivity than $CO₂$. The combined reactivity obtained with both gasification agents was a linear combination of the two individual reactivities.

Nevertheless, Roberts et al. [\[22\]](#page--1-0) disagreed partially with the former results, indicating that the rate of reaction in a mixture with $CO₂$ and H₂O was not the sum of the two pure-gas reaction rates. A complex combination of the two reaction rates that appeared to be dependent on the blocking of reaction sites by the relatively slow Boudouard reaction was stated. Farzaneh et al. [\[23\]](#page--1-0) also studied the gasification of chars using either $CO₂$ or steam. The experiments led to almost similar results for activation energies of $CO₂$ gasification and steam gasification.

The information about co-gasification of rice straw and husks and plastic wastes blends is still scarce and some problems still need to be solved, due to the heterogeneity and particularities of such wastes. The work presented main objective was to study the viability of using co-gasification of biomass rice wastes and plastic to produce a gas suitable to be used as a biofuel. As the type and composition of gasification agent is an important parameter in the composition of the gas produced by gasification, the effect of using air, O_2 , CO₂ and steam in different compositions was studied. As mentioned before, several authors have studied the effect of gasification agent on gasification performance, however, similar results and trends were not always obtained, which justifies the work presented. The results reported refer to the first stage of the work that has been developed about co-gasification of rice husks and plastic wastes. Other important aspects like controlling gasification bed agglomeration will be addressed in a future work.

2. Experimental part

2.1. Co-gasification bench-scale installation

Co-gasification bench-scale installation is shown in [Fig. 1](#page--1-0). The reactor was a bubbling fluidised bed gasifier made of a refractory steel pipe. The reactor was circular in cross-section with an inside diameter of 80 mm and with a height of 1500 mm. The feeding system was water cooled to avoid some clogging, that might be due to pyrolysis of the feedstock, prior to the entry into the gasifier, especially when PE was used in the feedstock. A nitrogen flow was also

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