



Synergetic and inhibition effects in carbon dioxide gasification of blends of coals and biomass fuels of Indian origin



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HIGHLIGHTS

- Focus on role of catalytic elements in ash in enhancing CO₂ reactivity of fuels.
- Blends of chars of coals and biomasses of Indian origin studied experimentally.
- Systematic reactivity studies using isothermal thermogravimetric analysis.
- Established synergistic effect through 90% conversion time of coal char.
- Inhibition tendency noted at high temperatures for high ash coal.

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ABSTRACT

The present study investigates the enhancement of CO₂ gasification reactivity of coals due to the presence of catalytic elements in biomass such as K₂O, CaO, Na₂O and MgO. Co-gasification of three Indian coal chars with two biomass chars has been studied using isothermal thermogravimetric analysis (TGA) in CO₂ environment at 900, 1000 and 1100 °C. The conversion profiles have been used to establish synergistic or inhibitory effect on coal char reactivity by the presence of catalytic elements in biomass char by comparing the 90% conversion time with and without biomass. It is concluded that both biomasses exhibit synergistic behavior when blended with the three coals with casuarina being more synergistic than empty fruit bunch. Some inhibitory effect has been noted for the high ash coal at the highest temperature with higher 90% conversion time for the blend over pure coal, presumably due to diffusional control of the conversion rate.

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1. Introduction

There has been sustained increase over the last two centuries of atmospheric concentrations of greenhouse gases (GHGs), contributing to a significant rise in the global average atmospheric temperature. This is attributed to too much dependence on fossil fuels to satisfy the ever increasing demand for energy. As a recognition of the devastating consequences associated with global warming, the recent global meet on climate change held in Paris (www.cop21paris.org) declared its intention to strive toward limiting the rise in global temperature to well below 2 °C through a concerted worldwide effort to reduce GHG emissions from anthropogenic sources. India, the third largest emitter of CO₂, has declared its commitment to reducing CO₂ emissions per GDP (gross domestic product) so as not to derail its economic growth. While

there is thus consensus on the need to reduce GHG emissions, there is less unanimity on the measures to be adopted to achieve these. Continued reliance on indigenous, coal-based power generation is inevitable for the next couple of decades for many countries such as India. In such a scenario, reductions in GHG emissions are possible only by resorting to more efficient use of hydrocarbon fuels for power generation. The present work is motivated by this objective and investigates the possibility of blending coal with local biomass to generate syngas, which can then be used to generate power through the more efficient combined cycle route as opposed to using the Rankine cycle.

Biomass is considered as a carbon-neutral form of energy as the CO₂ released during its utilization is equal to the CO₂ absorbed from the atmosphere during photosynthesis (Tillman et al., 2012; Masnadi et al., 2015). Biomass gasification technologies are not viable economically due to the low energy density of biomass and the lack of assured availability. Adding coal to biomass can increase specific energy content of the product. Since biomass

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has relatively small amount of ash, blending it with high ash Indian coals mitigates the ash-related problems in direct usage of coal. There can be benefits arising from reactivity considerations, too. In view of these factors, we consider the kinetics of co-gasification of two fast growing biomasses, namely, empty fruit bunch (EFB) and casuarina equisetifolia, with Indian high ash coals.

The conventional route to syngas generation from solid fuels is through steam gasification. Of late, there has been growing interest in CO₂ as the gasifying agent (Saravanan et al., 2011; Lahijani et al., 2013a,b,c; Naidu et al., 2016). This is partly driven by the prospect of having highly concentrated CO₂ stream readily available on-site in a futuristic scenario in which carbon capture and sequestration (CCS) is employed to minimize CO₂ emissions into the atmosphere. Given India's abundance reserves of coal and seasonal availability of agricultural waste in large quantities, syngas generation through co-gasification appears to be the most appropriate route to efficient utilization of indigenous resources. In view of this, the co-gasification possibilities of three high ash Indian coals and two fast-growing biomasses, namely, empty fruit bunch (EFB) of palm trees and the twigs of casuarina tree (casuarina equisetifolia, referred to henceforth as CE), are of interest in for the present study. Of the five solid fuels selected for the present study, EFB has been studied widely and is known to have much higher reactivity than coals, which is attributed to the presence of catalytic elements in ash (Mohammed et al., 2011; Lahijani et al., 2013a; Naidu et al., 2016). The other biomass considered in the present study is the fast growing tree, casuarina, found on the sea shore along the coastline of India. The needle-like twigs which are continuously shed are currently used for low grade heat generation. The reactivity studies of casuarina are of recent origin (Sharma et al., 2015; Naidu et al., 2016); these indicate that its reactivity is comparable to that of EFB.

Co-utilization of biomass with coal can be achieved by direct co-combustion and co-gasification. The reactivity of co-gasification of the blended coal and biomass can be enhanced due to the catalytic effect of the high content of metal elements in the biomass ash (Lahijani et al., 2013a,b). Biomass is already being used as a supplemental fuel to substitute up to 10% of the base fuel in commercial operations to reduce formation of NO_x and SO_x emissions (Agbor et al., 2014). In the case of direct co-firing, the presence of 80% of volatile matter on dry basis in the biomass has the potential to be very effective reburn fuel so as to reduce the NO_x produced when coal is used as the primary fuel (Sami et al., 2001). Biomass can also be used in indirect gasification mode; this would require additional biomass gasifier which needs to be dynamically coupled and thermally integrated with the rest of the plant. Unless this becomes mandatory, for example, due to severe fouling and slagging problems that may result from incompatible ash characteristics, co-firing is advantageous.

Although adding synthetic catalysts can be very effective in enhancing gasification, cost and recovery of the catalysts are major issues. One possibility to address this is to co-gasify biomass with the coal, in which ash minerals of the biomass, constituting alkali components (e.g. K⁺, Na⁺ and Ca²⁺), act as inexpensive natural catalysts to enhance co-gasification (Lahijani et al., 2013b; Masnadi et al., 2015). Studies reported in the literature (Gil et al., 1997; McLendon et al., 2004; Lapuerta et al., 2008) show that the effect and extent of synergy or inhibition depend on the gasification operating and fuel conditions, such as feedstock type, particle contact, temperature and pressure, and reactor type. Char gasification can be considered as the rate controlling step in the thermochemical conversion of carbonaceous materials. The presence of alkaline (K and Na), alkaline earth (Ca and Mg) metals promotes the carbon reaction rate (Dupont et al., 2011; Lahijani et al., 2013a,b). The ash forming elements in biofuels are salts such as chlorides (KCl), and sulfates (K₂SO₄) (Arvelakis et al., 2003; Fryda et al., 2008;

Mettanant et al., 2009; Serrano et al., 2015). Therefore, for high potassium containing biomass fuels such as empty fruit bunch, gasification or combustion is recommended to be restricted to 800 °C or lower in order to ensure that there is no formation of agglomeration (Lahijani and Zainal, 2011). Recent experimental studies using high ash Indian coals (Datta et al., 2015) showed agglomeration tendency under steam gasification conditions which could be avoided by maintaining proper steam to fuel ratio for a fixed flow rate of air to fuel ratio. The Boudouard reaction is also strongly influenced by the surface morphology. The gasification reaction initiates with the more disorganized carbon where the active sites are activated by the adsorption of the gasifying agent. Therefore, after an initial rise in reactivity to a maximum limit, the reactivity decreases and tends to flatten as time progresses as the available surface area diminishes due to reduction in the number of potential gasification sites (Diez and Borrego, 2013).

In the light of the considerable literature that exists on the effect of biomass on the reactivity of coals, the present study is motivated by the need to study the reactivity enhancement possibilities with two biomasses, namely, empty fruit bunch (EFB) and casuarina equisetifolia (CE) in the context of co-gasification with Indian coals using CO₂ as the gasifying agent. To this end, systematic isothermal TGA studies have been conducted over a range of temperatures and for different proportions of biomass and coal chars. The results show that improvement in reactivity occurs under most conditions for the blended samples; however, inhibition of reactivity too has been found under certain conditions. Details of the experiments and the results obtained are discussed below.

2. Methods

One high ash coal (nearly 48% of ash) obtained from Bilaspur (in the state of Chhattisgarh), denoted as BS, two medium ash coals obtained from Godavari khani and Bellampalli (in the state of Andhra Pradesh) denoted as GK and BM, respectively, and two biomass fuels, namely, empty fruit bunch and casuarina equisetifolia (of nearly 70% of volatile matter), denoted as EFB and CE, respectively, obtained from locally available agro fields, have been used in the present study to investigate the reactivity of blended fuels. The coal was crushed and ground to a size of $-106/+90\ \mu\text{m}$ to minimize diffusional effects during the gasification reaction. The biomass fuels were cut to the size range of 1–2 mm in order to prepare chars. These raw fuels were characterized through proximate and ultimate analyses as per ASTM standards. The calorific value was measured using a bomb calorimeter. The detailed analysis of the fuels is given in Table 1. Ash analysis of the fuels is of particular importance to the present study and it was performed based on ASTM standard for coal and both ASTM and EN standards for the biomass fuels. The high percentage of alkali and alkaline compounds in the biomass mineral matter would not be reflected in the ASTM standard carried out at 900 °C and therefore the ash analysis of biomass fuels was carried out at the lower temperature of 550 °C as per EN standard. The detailed ash analysis of the raw fuels is given in Table 2. The results for high ash coal and EFB compare well with those reported in the literature (Datta et al., 2015; Madhiyanon et al., 2012), respectively. About 80–85% of the ash is composed of silica and alumina in the three coals with silica alone contributing about 60–70%. In the case of EFB, K₂O is the largest contributor to ash but it also has a significant amount of silica. The other biomass (CE) has a large amount of CaO and very little silica and alumina and thus presents a contrasting feature.

All chars were prepared in a muffle furnace at 900 °C for seven minutes as per ASTM standards. In the process, a significant

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