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Optimisation of hydrogen production by steam reforming of chars derived from lumber and agricultural residues

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ABSTRACT

The aim of this study was to develop a process to produce hydrogen-rich gas at temperatures between 650 and 700 °C, by steam reforming of char produced in a pyrolysis reactor. To achieve this, the experimental work was divided into three sections all of which primarily focus on the production of hydrogen by steam reforming. The first section investigated the impact of different chars, produced from different biomasses, and the second and third considered the effects of temperature and residence time on gas composition and yield. Feedstocks selected for this study included apple pomace, birch sawdust, coffee husk, corn cobs, dried distilled grains, hemp seeds, sugar cane and tobacco stalks. It was found that hydrogen yield, calculated as a mass percentage of biomass processed, was highest for chars produced from hemp seeds, tobacco stalk, corn cobs and coffee husk and lowest for apple pomace and sugar cane. A relationship has been derived between char yield and production of hydrogen, especially in the case of hemp seeds. Consequently, a trend was discovered ($R^2 = 0.90$) when comparing the ash free char yield with the carbon content of the feedstock, which is useful to estimate char yields on an ash free basis if the carbon content is known. An inverse relationship was observed, for tobacco stalk, between the quantity of hydrogen produced and its purity, represented as a mole percentage of hydrogen in the gas stream, for the three different steam reforming temperatures (650, 675 & 700 °C). The residence time investigation showed that decreasing the vapour residence time, reduced both hydrogen production and purity, possibly as a result of the water gas shift reaction.

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Introduction

The thermal conversion of biomass by pyrolysis produces three primary products: bio-oil, char and gas. The gas is often

burned to provide energy for the conversion process. Depending on the feedstock type, the liquid bio-oil may be used as a fuel, chemical intermediate, pesticide, food additive or pharmaceutical ingredient. The solid bio-char product is combustible and consists mainly of carbon, oxygen and non-

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volatile inorganics. Currently, bio-oil appears to be the main desired pyrolysis product because of its potential as a green fuel. In addition to fuel, the co-product bio-char may be used to produce fertilizers or activated carbon [3]. However, another potentially attractive use of bio-char is the production of hydrogen for fuel cells, ammonia synthesis, hydrogenation reactions or bio-oil upgrading.

When continuously feeding biomass into a pyrolysis reactor, there can be a continuous bio-char build-up within the bed. Such char build-up within the bed will not only disrupt fluidization, but will also inhibit bio-oil production [8]. Because of this, it is important to minimize the char accumulation within the pyrolysis reactor. There are, therefore, two possibilities to minimize char build-up:

- 1) The pyrolysis process could be run in a semi-continuous fashion. When char builds up to a significant level within the reactor, the biomass feeding is stopped. At this point, air is fed into the reactor to burn the char, leaving ash in the bed and significantly increasing the reactor temperature. The remaining ash is smaller and less dense, and, therefore, more likely to be elutriated from the reactor bed. In order to add further value to the process, it would be beneficial to gasify the char within the reactor instead of simply burning it.
- 2) The pyrolysis process could be run in a continuous fashion. Sand-char mixture is continuously removed from the pyrolysis reactor bed and transferred to an additional unit where char is either combusted or gasified. Clean sand is then returned to the pyrolysis reactor.

When char is exposed to steam at high temperatures, reactions between steam and char produce gases such as hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2) and methane (CH_4). The steam to char ratio can be varied to adjust the H_2/CO product ratio, as shown later. Yan et al. [23] obtained a high yield of hydrogen gas after the steam reforming in a fixed bed reactor with pyrolytic char derived from cyanobacterial blooms, and Smolinski [21] observed good hydrogen production with the steam reforming of coal produced by pyrolysis. There has been some success in bio-char steam reforming by Refs. [11,20,10,19,22,18].

Using steam to increase hydrogen production is not limited to the steam reforming of bio-chars, since the water gas shift reaction simultaneously occurs, as long as steam and CO are present. In the case of cellulose gasification, Asadullah et al. [1] found that increasing steam flow rate led to a more hydrogen-rich product gas due to the water–gas shift reaction. Li et al. [13] also observed the same results when gasifying sawdust, similarly to Kechagiopoulos et al. [12] when steam reforming the aqueous phase of bio-oil. Other researchers [9,16,17,7] also found that increasing the steam flow rate increased the hydrogen produced by gasification of model compounds of pyrolysis oils, vegetable oils, pine sawdust and various other biomass types. Recently, Ma et al. [15] explored the effects of reaction temperature (750–900 °C) and the steam to carbon ratio (from 1 to 4) on hydrogen production using a fluidized-bed reactor. Although there was neither mention of the char type nor investigation of the residence time effects, they were able to produce a hydrogen yield of 0.167 kg/kg bio-char under

optimized conditions. Zheng et al. [24] carried out similar temperature studies to Ma et al. [15]; using a fixed bed reactor, and were able to generate a maximum hydrogen yield of 0.0714 kg/kg bio-char with a hydrogen concentration of 74 vol% at 800 °C.

The objective of this study was to develop a process to produce a hydrogen-rich gas at low temperature utilizing the char that accumulated within a fluidized bed pyrolysis reactor. This work aimed at the study of the gas composition and yield derived from the steam reforming of bio-char produced from the pyrolysis of apple pomace, birch sawdust, coffee husks, corn cobs, dried distillers grains, hemp seeds, sugar cane and tobacco stalks. In addition, the effects of temperature and residence time were investigated.

Experimental

The reactor used was a 7.62 cm diameter fluidized bed equipped with a perforated plate distributor. The inert fluidized medium was silica sand with a Sauter mean diameter of 204 μm . The bed was fluidized in the bubbling regime at a superficial velocity of 0.235 m/s. The reactor was externally heated using electric band heaters positioned along the length of the reactor. Thermocouples protruding the inner reactor wall were used to measure the reactor temperature. An average value was taken from thermocouples placed at the center of the reactor. Fiberglass insulation was used to reduce heat losses from the reactor.

For all feedstocks, except hemp seeds which are naturally suitable for slug feeding, were ground in an IKA Werke model MF10 basic microfine grinder operating at 4500 RPM. The feedstock is dried overnight at 105 °C prior to pyrolysis processing. The elemental analysis was also performed on all the biomasses using a Flash EA 2000NC elemental analyzer. After grinding to particle sizes between 0.21 and 0.85 mm, a slug feeder similar to the one described by Berruti et al. [2] injected 25 g slugs of biomass into the dense phase of the reactor at 1 min intervals. The bio-oil vapors from pyrolysis at 550 °C exited the reactor and were condensed in an aluminum condenser submerged in 4 °C water. The non-condensable gases continued to a vent.

The pyrolysis also created char that remained in the reactor. Once the pyrolysis had finished, the reactor was heated up to 700 °C under a nitrogen blanket. Upon reaching 700 °C, the bed was subsequently fluidized with steam and maintained at 700 °C to achieve steam reforming. The steam was always fed in excess and, at 700 °C, the production of carbon monoxide was always very small, regardless of the char origin. This finding shows that such process is therefore attractive to maximize the hydrogen content in the produced gas. Operating at a low temperature maximizes the production of hydrogen but requires long reaction times. Since this was a batch process, steam reforming was continued until gas production ceased.

In order to observe the effect of the steam reforming temperature, 25 g samples of feedstock were fed into the reactor using the slug feeder and pyrolyzed at 550 °C. Then, instead of heating the reactor to 700 °C, it was heated to 650 °C or 675 °C before steam was injected.

Steam for fluidization and reforming was produced with a 3.8 kW electric Lundberg Blue furnace. Demineralised water

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