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## Entrained flow gasification of straw- and woodderived pyrolysis oil in a pressurized oxygen blown gasifier

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#### ABSTRACT

Fast pyrolysis oil can be used as a feedstock for syngas production. This approach can have certain advantages over direct biomass gasification. Pilot scale tests were performed to investigate the route from biomass via fast pyrolysis and entrained flow gasification to syngas. Wheat straw and clean pine wood were used as feedstocks; both were converted into homogeneous pyrolysis oils with very similar properties using in-situ water removal. These pyrolysis oils were subsequently gasified in a pressurized, oxygen blown entrained flow gasifier using a thermal load of 0.4 MW. At a pressure of 0.4 MPa and a lambda value of 0.4, temperatures around 1250 °C were obtained. Syngas volume fractions of 46% CO, 30% H<sub>2</sub> and 23% CO<sub>2</sub> were obtained for both pyrolysis oils. 2% of CH<sub>4</sub> remained in the product gas, along with 0.1% of both  $C_2H_2$  and  $C_2H_4$ . Minor quantities of  $H_2S$  (3 vs. 23) cm<sup>3</sup> m<sup>-3</sup>, COS (22 vs. 94)  $\text{cm}^3 \text{m}^{-3}$  and benzene (310 vs. 532)  $\text{cm}^3 \text{m}^{-3}$  were measured for wood- and straw derived pyrolysis oils respectively. A continuous 2-day gasification run with wood derived pyrolysis oil demonstrated full steady state operation. The experimental results show that pyrolysis oils from different biomass feedstocks can be processed in the same gasifier, and issues with ash composition and melting behaviour of the feedstocks are avoided by applying fast pyrolysis pre-treatment.

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#### 1. Introduction

The production of transportation fuels from biomass is desirable for a several reasons, including for example the security of supply and environmental considerations [1]. Renewable alternatives for fossil fuels are thus created, which are ideally directly applicable in existing infrastructure. The term biomass applies to a wide variety of resources with diverse properties [2]. Developing a technology (chain) suitable to convert various biomass streams into a flexible product portfolio is therefore very desirable.

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One of the most feedstock flexible approaches to convert biomass into biofuels is believed to be via fast pyrolysis and entrained flow gasification. Fast pyrolysis of biomass has been studied extensively the last decades and several reactor technology concepts have reached commercial scale status [3]. In the fast pyrolysis process, biomass is rapidly heated in an oxygen free environment to form condensable vapours, permanent gases and a solid residue often referred to as char. After rapid cooling of the condensable vapours, up to 700 g kg<sup>-1</sup> of biomass can be converted into a liquid product called pyrolysis oil. With comparable heating values, the overall energy yield from biomass to pyrolysis oil is in the same range (0.7 J J<sup>-1</sup>).

Pyrolysis oil can be derived from virtually any lignocellulosic biomass, and although product yields vary, the pyrolysis oil properties are comparable if produced properly. Pyrolysis oil can readily be used for the production of heat and electricity [4]. More advanced applications require further treatment. The pyrolysis process can thus be regarded as pretreatment process. Pre-treatment of biomass via pyrolysis results in various advantages compared to the direct utilization of biomass. The energy density on volumetric basis is a factor 3-10 higher for pyrolysis oil, decreasing e.g. transportation costs. Via pyrolysis, locally available biomass streams can be exploited as renewable material which could otherwise not be processed economically. Besides economic and ecologic advantages, pre-treatment via pyrolysis also creates technical advantages. Pressurizing pyrolysis oil is much easier and cheaper than pressurizing solid biomass. Also, ash forming elements are reduced by an order of magnitude. The decision to include fast pyrolysis as pretreatment or gasify the powdered biomass in an entrained flow gasifier directly will ultimately depend on biomass properties and the specific scenario.

Entrained flow gasification is widely used to produce syngas from various fossil feedstocks such as coal and natural gas by reacting the feedstock with pure oxygen to form H<sub>2</sub>, CO, CO<sub>2</sub> and H<sub>2</sub>O. Due to the high operating temperature of 1200 °C-1600 °C high conversion rates are obtained and the gas quality is generally good with low tar and methane concentrations [5]. Important aspects for entrained flow gasifiers are the presence of inorganic components in the feedstock. For coal gasifiers, part of the inorganic components present in the feedstock form a liquid slag layer inside the gasifier. This slag layer is used to protect the gasifier wall, and is continuously removed from the system. In certain cases, additives are fed to the gasifier to ensure a proper flow of the slag. These types of gasifiers are called slagging gasifiers. Feedstocks containing no, or limited amount of inorganics, can be processed in non-slagging gasifiers. Examples are entrained flow gasifiers fed with natural gas and the residue gasifiers often found in petrochemical refineries. Generally, the non-slagging gasifiers are a cheaper option than the slagging gasifiers. In this work the contaminants of the original biomass are removed in the pyrolysis pre-treatment process, enabling the use of a non-slagging gasifier.

Even though entrained flow gasification of powdered biomass or pyrolysis oil for the production of syngas for subsequent product synthesis is a process combination often mentioned both in commercial- and scientific environment, actual test results are scarce. Further information on entrained flow gasification of various powdered biomass feedstocks is given by Öhrman [6] and Weiland [7,8]. With respect to gasification of pyrolysis oil, back in 2002, a test was performed to gasify pyrolysis oil in the 1 MW entrained flow gasifier at the site of UET in Freiberg in the Carbo-V<sup>®</sup> process. Results of this work are only recently published [9]. In this test about 1500 kg of pyrolysis oil was produced by BTG from clean wood and was gasified at a rate of 140 kg  $h^{-1}$  for about 10 h until the feed ran out. The temperature in the gasifier was maintained around 1200 °C, resulting in volume fractions of 30% CO, 25% CO<sub>2</sub> and 20% H<sub>2</sub> in the product gas, thus achieving a  $H_2$ /CO ratio of 0.66. About 0.1 kg kg<sup>-1</sup> of pyrolysis oil ended up as soot, which was quite high compared to published laboratory experiments of BTG [9] and Chhiti [10]. Entrained flow biomass gasifiers in particular are in fact known to have issues related to soot formation, as published already in the 1998 [11]. Control of the soot formation and optimization of gasification conditions will thus be important for the entrained flow concept.

A slightly different approach is followed in the so-called BIOLIQ process, under development at the Karlsruhe Institute of Technology (KIT), formally known as the Forschungszentrum Karlsruhe (FZK). In cooperation with the Future Energy company, tests were performed to gasify a pyrolysis oil slurry in 2–3 MW entrained flow gasifier [12]. The slurry stream consists of pyrolysis oil, mixed with pyrolysis char. Mixing both products increases the energy content of the feedstock, and potentially increases the overall energy efficiency of the chain. Downside of this approach is that the mineral matter from the original biomass is also fed to the gasifier, and a slagging gasifier is required. As a result, this route limits the flexibility towards using feedstocks from pyrolysis plants converting different biomass streams, since a proper flow of the slag needs to be maintained at all time during operation. In the period 2002-2005, four test campaigns have been performed in the Future Energy gasifier [12]. A total of about 50 Mg of different slurries were produced for the gasification tests. Slurries included the products of commercial beech wood and wheat straw pyrolysis for charcoal production, as well as from the fast pyrolysis of wood (Dynamotive company). The gasifier operated typically at 2.6 MPa, and temperature between 900 and 1600 °C were achieved for equivalence ratios between 0.36 and 0.65. Dry gas volume fractions varied between 52 and 23% for CO, 32-20% for H<sub>2</sub> and 13-31% for CO<sub>2</sub>, with the first number corresponding to the lower equivalence ratio's, and the second for higher equivalence ratio's. The remaining part consisted of  $N_2$  (9%–17%, primarily purge stream) and small fractions of CH<sub>4</sub> (0-1)%. For temperatures above 1200 °C, carbon conversions over 0.99 kg kg<sup>-1</sup> are reported, although no detailed information is given on the carbon conversion measurement method.

In 2012, preliminary tests were performed in the pressurized entrained flow biomass gasifier (PEBG) at the Energy Technology Centre in Piteå, Sweden (ETC) within the same consortium as the current work. Goal of this first test campaign was to test the suitability of the system to convert a liquid feed. Up to that point the gasifier had only been used to operate on solid feedstock [6,7]. Also, parameters such as the equivalence ratio were varied to determine an optimal

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