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Experimental investigation of an industrial scale black liquor gasifier. Part 2: Influence of quench operation on product gas composition

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ABSTRACT

Pressurised black liquor gasification combined either with a gas turbine or a catalytic fuel synthesis process is a novel technique for production of green electricity or second generation motor fuels. The composition of the gas produced in the gasifier may be important for the performance of either the gas turbine or the catalytic fuel synthesis process and different operating parameters of the gasifier may affect the composition of the produced gas. The aim of this study was to investigate the influence of some operating parameters on the final gas composition with special attention on the performance of the quench in the gasifier. The results show that system pressure, oxygen/black liquor flow rate ratio and the primary spray flow rate in the quench significantly affect the final gas composition. Furthermore, depending on the cooling rate in the quench, the hot reactor gas composition prior to the quench could either be preserved (high cooling rate) yielding the same final gas composition after the quench as in the hot reactor or shifted (low cooling rate) towards a higher concentration of H_2 and CO_2.

1. Introduction

Chemrec AB:s technology for pressurised black liquor gasification combined with a catalytic process for synthetic motor fuel production has the potential to globally produce green motor fuels equivalent to over 45 billion liters/year of gasoline equivalent [1]. Black liquor is a by product of the chemical kraft pulping process and it consists of roughly 30% water, 30% inorganic cooking chemicals (valuable for the pulp mill) and 40% lignin and other organic substances separated from the wood during chemical pulping in a digester. To obtain a proof of the concept, a Development Plant (DP1) with a capacity of 20 ton dry solid of black liquor/24 h (~3 MW_{th}) was commissioned in 2005. The gasifier is located next to the Smurfit Kappa Kraft-liner pulp and paper mill in Piteå, Sweden. The Chemrec DP1 is fully instrumented and is operated continuously by a 5-shift operating team to provide proof that the technology is sufficiently robust for industrial use and to provide valuable data for scale-up of the process. The plant has been operated for more than 11,000 h (October 2009).

The main components of the black liquor gasifier are shown in Fig. 1 [2]. The gasifier consists of a refractory lined oxygen blown entrained down-flow gasification reactor, with a centrally placed gas assisted burner that produces small black liquor droplets (~100 μm) by atomisation with oxygen followed by a direct quench. In the reactor, pressurised (30 bar) high temperature (~1050 °C) gasification takes place, mainly through reactions between the black liquor and oxygen, steam and carbon dioxide producing a syngas (mainly H_2, CO, H_2O, CO_2, CH_4, and H_2S) and a liquid smelt containing Na_2CO_3, Na_2S and NaOH. The product gas and the smelt then enter a quench vessel beneath the reactor where rapid cooling occurs as the products are brought into direct contact with water droplets from several spray nozzles. The smelt droplets are separated from the gas flow and dissolved in a quench pool at the bottom of the gasifier to form green liquor. The saturated gas is thereafter cleaned from particles in a Counter Current Condenser (CCC) gas cooler. The dry (30 °C) and almost particle free (30–100 μg/Nm^3) syngas [3] is then analysed with conventional on-line gas instruments and finally combusted using a flare. In an industrial plant, the cooled syngas will go through an acid gas removal unit to remove the H_2S and CO_2 from the syngas. Thereafter, the H_2/CO ratio will be adjusted in a CO-shift unit that precedes the catalytic conversion unit converting the syngas to high value fuels and chemicals.

The final composition of the syngas is important for the catalytic process design and different operating parameters of the gasifier are known to affect the composition of the product gas that will form the final syngas. In an earlier paper by our group, an advanced gas sampling system was described that makes it possible to investigate the gas composition inside the current hot pressurised reactor [4]. In the first paper of this series, the gas sampling

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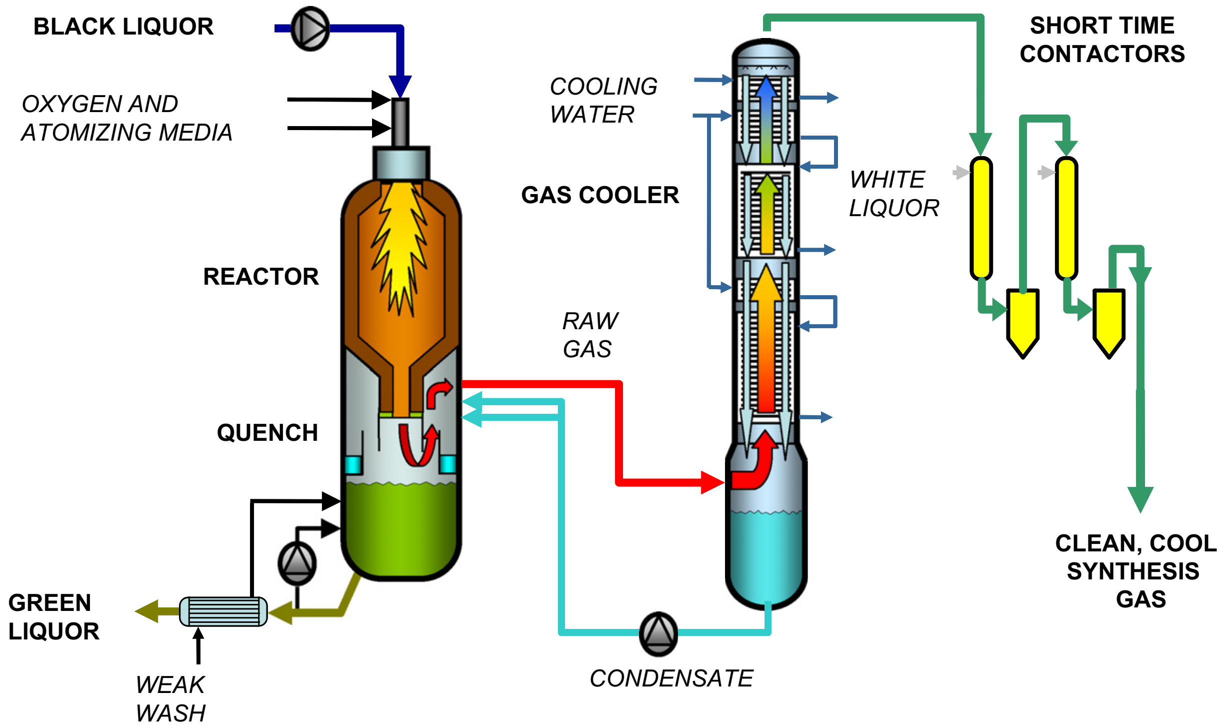

Fig. 1. Schematic drawing of the PEHT-BLG process [2].

system was used to experimentally investigate the influence of process operation on the gas composition inside the hot reactor [5]. In that investigation, it was found that the syngas composition in the hot reactor was affected by the system pressure, black liquor load, black liquor to oxygen mass flow ratio, and the black liquor temperature [5]. However, the composition of the syngas may also be influenced by chemical reactions and/or absorption when the raw product gas and smelt come in contact with the water spray and the green liquor pool in the quench. The aim of the second paper in this series is therefore to experimentally investigate the effect of the quench operation on the syngas composition.

The motivation for this investigation comes from an ambition to better understand the various stages in the current conversion process of black liquor and to provide experimental data that later on can be used to optimise the process and for validation of mathematical models (CFD models and thermo-chemical equilibrium models).

2. The quench

In general, gas from a gasification reactor is contaminated with various components that must be removed before the gas can be used in a synthesis process or for power production [6]. These contaminants, which may be particulates (soot or inorganic ash particles), sulphur or chlorine compounds and tars, must be removed from the gas. All such removal processes operate at a temperature considerably lower than that of the gasifier itself [6]. Therefore, the gas has to be cooled several hundred degrees.

One method often used to cool the gas during slagging gasification is to introduce fine water droplets into the hot gas and thus quench the conversion products by evaporation of the water. In contrast to ordinary gasification processes, the ash product from the current gasification concept (smelt dissolved in water forming green liquor in the quench) is an important by-product from the gasifier that must be recycled to the chemical pulping mill. Hence, proper operation of the quench in black liquor gasification is very important.

2.1. Geometry and function of the quench

The general principle of the quench in the current Chemrec type of gasifier was first proposed by Stigsson and Bernard [7] and a schematic sketch of their quench concept is presented in Fig. 2 [7]. In this paper, a short description of the geometry and the function of the quench are given, adopted from the work by Stigsson and Bernard [7]. The hot product gas with smelt droplets from the reactor is forced to pass through the reactor throat (2 in Fig. 2) and down into the downcoming quench tube (3). At the entrance of the quench tube, cooling liquid (water or condensate from the CCC) is injected into the gas flow through nozzles (primary spray, 4 in Fig. 2). The gas is then cooled by evaporation of the primary spray to a predetermined temperature greater than the steam saturation temperature at the prevailing quench vessel pressure, to provide a gas containing superheated steam. The gas with its superheated steam is then forced to change direction and turn about 180° at the lower end of the downcoming quench tube (5). The gas then flows upwards and out of the outer surrounding tube concentrically arranged around the central downcoming tube.

The entrained particles, containing inorganic alkaline compounds, are forced by gravity and/or inertia to fall into the quench pool where the compounds are dissolved in water to form "green liquor". The separated gas (with superheated steam) is then further cooled by a second cooling liquid (secondary spray, 7 in Fig. 2) and finally bubbled through a water column before it leaves the quench vessel. The gas has now been cooled down to the steam saturation temperature at the prevailing quench vessel temperature. The level of the liquid in the quench pool is an operational parameter, which can be controlled by addition of cooling liquids and by withdrawal of green liquor.

2.2. Chemistry in the quench

The hot gas at the outlet of the reactor mainly consists of H_2, CO, H_2O, CO_2, CH_4, H_2S, and COS [4,5]. A typical gas composition (as dry