FROM BIOMASS TO ELECTRICITY THROUGH INTEGRATED GASIFICATION/SOFC SYSTEM: GREEN FUEL CELL

F. Broust[(1)], P. Girard[(1)], T. Koch[(2)], B. van der Drift[(3)], S. Rouge[(4)]
[(1)] Agricultural Research Centre for International Development (CIRAD)
TA 10/16, 73, avenue J.-F. Breton, 34398 Montpellier, Cedex 5, France
Email: philippe.girard@cirad.fr - Tel : (33) 4 67 61 58 43 - Fax: (33) 4 67 61 65 15
[(2)] TKEnergi A/S, Stationsvej 4, 4621 Gadstrup, Denmark
[(3)] ECN Biomass, PO Box 1, NL 1755 ZG Petten, the Netherlands
[(4)] CEA Grenoble / DTN/SE2T - 17, rue des Martyrs - 38054 Grenoble Cedex 9 - France

ABSTRACT: The ongoing GREEN FUEL CELL project intends to develop an innovative sustainable technology with high “fuel to electricity” efficiency. The objective is to produce a gas suitable for SOFC application through reliable, up-scalable and cost-effective staged gasification of biomass. A basic design will be prepared for a full-scale (1-50 MW_{th}) innovative gasifier and gas treatment system for integrated biomass gasification SOFC systems. The paper presents the research activities, lab tests and pilot scale tests planned in the project. First, two new designed up scalable staged gasifiers are being developed, integrating tar cleaning technologies based on char beds. Secondly, the requirement of the gas quality for an SOFC fuel cell will be determined based on experiments with material from SOFC. According to these specifications, a complete train of gas treatment downstream the gasifier will be implemented. Finally a long term testing of two complete integrated gasification-fuel cell stacks plants will be performed on woody biomass, for at least 100 hours each. This should lead to development and proof-of-principle of the innovative gasifier-fuel cell system.
Keywords: Gasification, gas cleaning, innovative concepts.

1 INTRODUCTION

Innovative sustainable technologies with high “fuel to electricity” efficiency are required to supply the increasing electricity demand and at the same time to fulfil the objectives of reducing the emission of green house gases. Solid Oxide Fuel Cell (SOFC) is one promising technology to obtain a high efficiency in electricity production, which would be sustainable so long as the feeding fuel is produced from renewable energy sources. The hydrogen-rich gas produced by gasification of biomass could be such a sustainable fuel for SOFC. For biomass gasification-SOFC conversion systems the most cost-effective size is expected to be plants from 1 to 30 MWe, with a total electric efficiency expected to be well above 50 %.

However, the main barrier for the development of this technology is that the gases produced by most of the current gasification technologies are still too dirty to be directly fed into SOFC without extensive and expensive gas cleaning systems, while staged fixed-bed gasification producing very low amounts of tar (<50 mg/Nm^3) cannot be up-scaled to more than 10 MW_{th}**.** Moreover, until now most research, development and demonstration of gasification has been focused on the potential for CHP, based on engines and turbines where the requirements for the purity of the gas are not as elaborate and extensive as for an SOFC fuel cell.

On the other hand, the current state-of-the-art for SOFC technology is only based on experience with synthetic gases or from natural gas reforming and similar very uniform and clean fuels. SOFC specifications towards syngas are still to know.

The ongoing European project “Green Fuel Cell” aims at enabling the use of gasification gas from biomass as feed gas in SOFC fuel cells, which requires

i) knowledge of the requirements for gas quality,
ii) appropriate gasification process and subsequent gas cleaning system.

Extensive research activities regarding theses aspects, lab tests and pilot scale tests, and finally a long-duration test of the integration of the gasifier and fuel cell should lead to development and proof-of-principle of the innovative gasifier-fuel cell system. Figure 1 shows a schematic overview of the different steps investigated within the Green Fuel Cell concept.

The main objective is to produce a gas suitable for SOFC application through reliable, upscalable and cost-effective staged gasification of biomass. A basic design will be prepared for a full-scale (1-50 MW_{th}) innovative gasifier and gas treatment system for integrated biomass gasification SOFC systems, with the following expectations:

- Low tar content of the gas (<10 mg tar/Nm^3),
- Cold-gas efficiency of more than 85% for the total gasification process,
- Carbon conversion of more than 99%
- Minimal process waste streams and by products, to reduce the environmental impact of the waste from the gasifier and the operational cost.

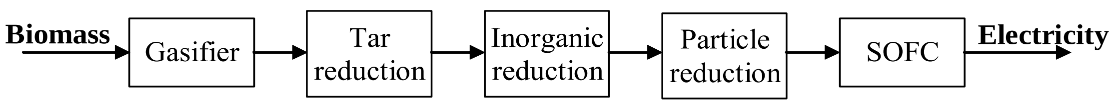

Figure 1: Biomass-to-electricity concept developed in Green Fuel Cell

2 TECHNICAL APPROACH

2.1 Design of innovative char-beds for tar reduction

Char has been proven to be suitable as catalytic agent for the reduction of tar concentration at high temperatures (900°C or higher). The principle of removing (and cracking) tar with a char-bed is illustrated on Figure 2.

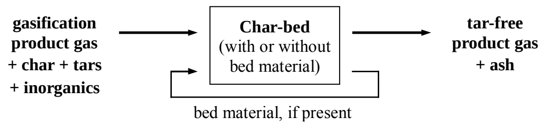

Figure 2: Concept of tar removal with char bed in Green Fuel Cell

In the char-bed, three continuous processes take place:

- ✓ The char from pyrolysis or gasification is captured in the bed;
- ✓ Tars in the gas are adsorbed on the char and destructed due to the catalytic effect of the char and/or mineral components. In the cases where an inorganic bed material is present, this will be internally recycled;
- ✓ Due to the long residence time of the char in the bed, most of the char is gasified due to the presence of H_2O and CO_2 in the gas. The ash formed is entrained by the gas and will be removed in a downstream gas filter. Similarly, residual char will be removed downstream.

The technical idea of this project is to design an upscalable char bed that can be integrated into existing gasifiers in order to reduce tar concentrations to a level low enough to avoid tar-related problems in an SOFC-system. Two ways of creating the necessary char bed will be developed, tested and compared within the project.

- *TKE-concept of char-bed (without bed material)*

The first char-bed design (TKE-design) consists of a dense stratified char bed where a major part of the char from the gasification/pyrolysis will end up. The hot gas is blown through this zone and the tar is decomposed.

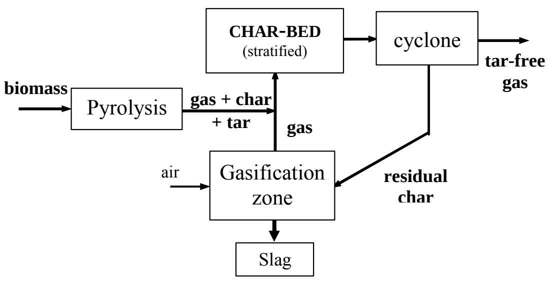

Figure 3: TKE concept of char bed for tar removal

This design presents two major innovations (Figure 3):

i. Char is “trapped” in a conical stratified fixed char bed (the lower part of the bed is spouted and the top is fluid). This design ensures that no char particles can move through the gasifier without resting in the hot zone of the char bed. The advantages foreseen of this design are that it ensures very low tar content in the gas and it can be upscaled, contrary to fixed bed.

ii Char loss is prevented by recycling the char and ash that leaves the char bed reactor to a combustion chamber, where the char is gasified or combusted. The hot gas from the combustion chamber is reintroduced to the process below the char bed reactor.

- *ECN-concept of char-bed (with bed material)*

The other char-bed design, called TREC-reactor (Tar REduction by Char), is a moving char/sand bed based on granular filter technology, to be mounted at the outlet of an existing circulating fluid bed gasifier (Figure 4).

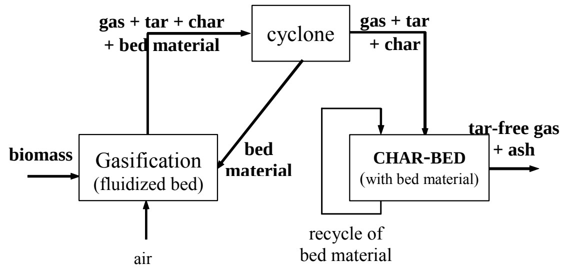

Figure 4: ECN concept of char bed for tar removal

The innovations of this design are:

i. Char particles entrained by the gas are trapped in a fixed bed containing (inert) bed material. The expected advantages are:
- good contact between gas (tar) and char where tars are destroyed catalytically,
- low pressure drop given a minimum amount of (inert) bed material,
- the remaining char is gasified by the product gas to achieve higher char conversion.

ii three main disadvantages of a conventional and commercially available gasifier, being a low carbon conversion (CFB gasifier in average only 90%) and C-rich ashes and a gas with a high tar content, are all solved by the coupling of the char-bed as described

iii Due to the down-stream positioning of TREC reactor without integration to the gasifier, the principle is generic and applicable in systems with other types of gasifiers.

2.2 Fundamental research on tars and inorganics

In order to make a comprehensive transfer of the technical principles into basic-designs for full-scale installations, the technological developments of char beds and gas cleaning system are supported by fundamental research activities directed towards understanding of the mechanisms of tar reduction with char and behaviour of volatile inorganics (metals and salts) with respect to cooler fouling. The main scientific objectives are to:

- Gain knowledge on tar formation and on their destruction in char beds in order to minimize the tar content in the gas.
- Determine relations between char properties (*e.g.* extent of devolatilisation, ash content, and composition) and catalytic effect for tar reduction;
- Understand the fate of volatile metals and salts in the gasification process and upon cooling in the gas cooler.

2.3 SOFC research

In order to complete the total design from biomass to electricity, the maximum allowable concentrations of organic (tars) and inorganic (metals, salts, H_2S, HCl, …) impurities will be determined, with regards to the operation of an SOFC. The operation parameters for a dry gas cleaning system will be identifed to ensure that also the inorganic impurities are removed to a level where the gas can be fed directly into a SOFC fuel cell.

2.4 Gas cleaning system

According to the specifications, a complete train of gas treatment downstream the gasifier will be implemented. Typical impurities are: solids, volatile metals (*e.g.* alkaline metals), tars, ammonia, chloride, traces of sulphur (H_2S, COS). Innovative technologies allowing gas cleaning at high temperature will be investigated to minimize loss of thermal efficiency between the gasifier outlet and the SOFC (both operate at around 800-900°C).

The gas cleaning train probably constitutes the bottleneck of the project.

2.5 Operation and evaluation of process

Final tests will be performed with the complete chain of unit operations from biomass to electricity as shown in Figure 1. Gas cleaning units (tar, inorganics, particles) will be operated in such a way that the experimentally determined maximum concentrations for the SOFC are met. The achievement will be the two gasifiers coupled to fuel cells stacks for at least 100 hours each.

Complete process evaluations will be performed on the basis of the project results in order to assess the systems in the market and compare both concepts (see pictures 5 and 6).

3 WORK IN PROGRESS

3.1 Char bed designs

The two different designs are in progress of development by TKE and ECN. In both cases, cold models have been built and led to experimental data useful to the design and construction of hot lab-scale pilots which are currently being tested.

- *TKE-concept of char-bed*

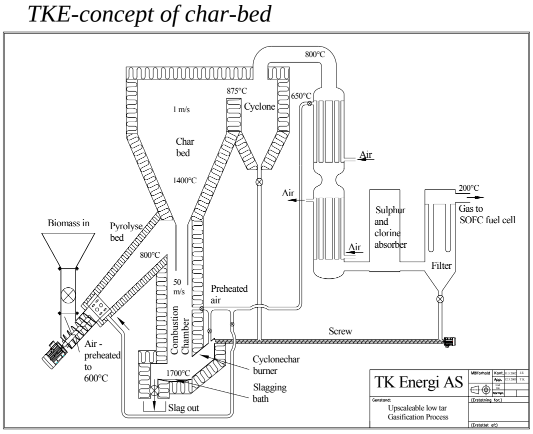

Figure 5: TKE upscalable staged gasifier

Experiments have been conducted with both cold models and a hot pilot scale facility [1]. The concept of the char bed reactor has proven successful with different kinds of materials. The hydrodynamics and temperature distribution in the system has been examined for a wide range of process parameters and a window of operation has been established. Recycling the char and ash from the char bed reactor makes it possible to conduct the gasification without char loss.

Based on these experiments a design for the total plant has been made. The updraft gasifier has finally been preferred to a slagging chamber, because fewer problems are expected due to temperature differences in the system.

The pyrolysis unit and the updraft gasifier have been designed, constructed and are currently being tested. A temporary stainless steel connection chamber (connecting the pyrolysis unit and the gasification unit) has also been built. Several full scale designs have been compared by CFD simulations. This tool was very useful to obtain the highest grade of mixing of the supplied air and pyrolysis gas before the mixture enters the char reactor [2].

- *ECN-concept of char-bed*

A small cold flow 2-dimensional transparent model has been designed and constructed in order to gain knowledge about material flows of granules (sand) as well as char flow, pressure drop, char distribution and scale-up variables.

The cold flow tests have proven that it is possible to separate the majority of char particles from a gas flow in the TREC-reactor concept (particle filtering efficiency up to 99%). Thus, reducing the particle concentration is confirmed to be an extra-advantage of TREC reactor.
It appeared that the dimensions of the reactor must be designed carefully so that the pressure drop increase over the bed can be limited to an acceptable level during stationary operation. More details about the TREC reactor design are available in ref [3].

The experiences with the cold-flow reactor have led to a detailed design of a high-temperature lab-scale TREC reactor to be placed downstream an existing 30 kW_{th} lab-scale fluidised bed gasifier producing 8 m_n^3/h of gas. The tests are currently in progress. The main conclusion so far is that TREC reactor is able to reduce the tar concentration significantly. It is expected that a significant amount of tar and some hydrocarbons react to soot. Soot caused operational problems in the TREC reactor. Further work will focus on the prevention of soot formation or the handling of the soot.

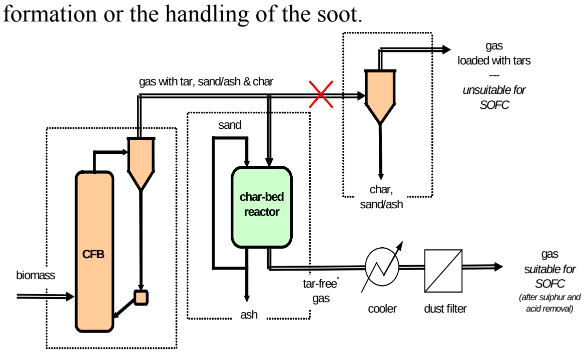

Figure 6: Schematic positioning of the TREC reactor downstream the gasifier (ECN).

3.2 Tar research

Several works are carried out simultaneously by the consortium to gain knowledge on tar formation and destruction in char beds.

- *Tar release*

A method has been developed by which the residual amount tar in a specific char is an indicator for the char quality. To determine the relationship between char quality and residual tar volatiles, char is produced in a la-scale facility at similar heating rates but with end-temperatures ranging from 250 to 1000 ^{o} C. The results are detailed in [4]. Simple models for tar release from pyrolysis have been applied and modified in order to describe the tar release as observed in the pyrolysis experiments [5].

Furthermore, residual tar analysis will be carried out on samples of char produced at varying pyrolysis temperature on a pilot plant recently at CIRAD, consisting of a 75 kWth pyrolysis reactor made of an externally heated screw conveyer (single first zone of a two-stage gasifier described in [6]) fed by woodchips. The lab-scale results will be compared to pilot scale operation

- *Tar destruction*

The aim of this task is to provide experimental data in order to determine a simplified model of chemical mechanisms of tar reducing in char bed. The present work will focus more particularly on the influence of the nature of tars (primary or tertiary tars respectively produced from pyrolysis or gasification processes) and of char (origin, ash content…) on the efficiency of tar destruction in char beds.

To achieve this, one approach investigates the chemistry and the dynamics of the char bed and relates this to the reduction of the concentration of tar compounds. ^{13}C labelled compounds are used for quantifying the incorporation of tar compounds into the char bed as well as for tracing the formation of simpler gaseous compounds especially CO [7]. Furthermore deuterium labelled compounds are used to investigate possible reversible binding of PAHs to the char bed.

Two kinds of chars will be used in bed experiments:

i. A pyrolysis-char, which has a low ash content and is representative of TKE-concept
ii. A gasification-char (90-95% conversion), which has a high ash content and is representative of ECN-concept

3.3 Inorganics behaviour modelling

Thermodynamic calculations have been performed to evaluate the composition of syngas at equilibrium, taking into account the inorganic composition of biomass and the conditions of gasification. For condensable species in the gas, the range of temperature where condensation occurs is determined for each species. This is of importance for corrosion risks evaluation and also for gas cleaning strategy. From the gas composition, interactions are calculated with materials for gas cleaning sorbents, and materials for SOFC anodes.

Details of the procedure and the results of the calculations are presented in [8].

Besides, laboratory experiments are being carried out to compare the efficiency of sorbents for removing chloride and sulfide compounds from simulated gas mixture in temperature range from 200 to 800 °C. The best sorbents will be chosen for the future SOFC application according to several criteria like efficiency, %mass retention, regenerability, cost.

3.4 SOFC research

During the project first year, research activities performed in the field of SOFC aimed at determining its sensitivity with respect to organic compounds. A setup has been constructed in order to carry out a parametric study of organic compounds with single SOFC cell tests on synthetic pre-mixed gases. Degradation of the cell due to carbon deposition is likely to occur and should be avoided by increasing the H_2O content and/or limiting the maximum allowable concentration of the organic compounds.

4 CONCLUSION

Both gasification concepts suggested in the project are innovative technologies which are expected to produce very low tar-content gases. Moreover, an appropriate dry clean system will solve inorganic contamination. For these reasons, various other applications requiring very clean gases can be considered including fuel synthesis.

More generally, this project should lead to the development and proof-of-principle of the innovative gasifier-fuel cell system. All together the different tasks will contribute to clear the path for biomass in a future based on the “hydrogen economy”.

REFERENCES

Conference papers related to this project in the 14th European Biomass Conference and Exhibition: Biomass for Energy, Industry and Climate protection:

[1] Developing an up-scalable fixed bed gasifier with no char loss. Nielsen M., Friehling P.B., Teglbjærg J.S. and Koch T.
[2] Optimization of Gasification Reactor using Computational Fluid Dynamics. Nielsen M.B., Noes, J. and Nielsen M.
[3] The Trec-Module Integration of Tar Reduction and High-Temperature Dust Filtration Carbo M.C., van der Drift A., van der Meijden C.M.
[4] Time Resolved Pyrolysis of Char. Egsgaard H., Ahrenfeldt J., Henriksen U.B.
[5] Pyrolysis of Pine Wood, experiments and theory. Fjellerup J., Ahrenfeldt J. and Henriksen U.B.
[6] Char Quality and Tar Formation Interdependance : Experiments in a New Two-stage Gasifier. Fassinou W.F., Martin E., Van de Steene L., Broust F., Teglbjærg J.S., Hansen F., Girard P.
[7] The thermal history of char as disclosed by carbon isotope ratios. Egsgaard H., Ambus P., Ahrenfeldt J. and Henriksen U.B.
[8] Minor Inorganic Species in Biosyngas and SOFC Material Interaction – Thermodynamic Calculations. Froment K., Rougé S., Bertrand C., Ravel S.

ACKNOWLEDGEMENTS
This project is funded by the European Commission 6th Framework Program (STREP, contract n°503122) and conducted in collaboration with: CEA (F), ECN (NL), TK Energi A/S, DTU, RNL, FORCE Tech. (Dk), ICT (Cz).