

# ENTRAINED FLOW GASIFICATION OF BIOMASS

## Ash behaviour, feeding issues, and system analyses

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## Acknowledgement/Preface

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Preliminary results have been presented during a workshop in December 2003 in the presence of people from Shell, Novem, SDE, TU-Delft, TU-Eindhoven, Gasunie and ECN [1].

## Key words

entrained flow, gasification, synthesis gas, syngas, ash, slag, feeding, pressurizing, piston

## Abstract

Biomass is heading for a great future as renewable energy source. Not only electricity and heat can be produced, also gaseous and liquid fuels may be synthesized from biomass. The present work focuses on entrained flow gasification, which is commercially available on large scale (mainly for coal and liquid fuels) and also can reach the highest efficiency from biomass to so-called biosyngas. As feeding and ash behaviour are expected to be the major hurdles, these issues have been subject of the present study.

Two types of entrained flow gasifiers can be distinguished: slagging and non-slagging. It has been concluded that biomass should be converted in a slagging gasifier. Slagging gasification of biomass requires fluxing material in order to obtain the proper slag properties at reasonable temperatures. Silica or clay seems to be the obvious choice. Furthermore, slag recycling might be needed to obtain enough wall coverage.

As far as feeding is concerned, there are various options to get biomass into an entrained flow gasifier. Milling wood to 100  $\mu\text{m}$  particles similarly to coal would consume very much electricity (approx.  $0.08 \text{ kW}_e/\text{kW}_{\text{th wood}}$ ), which makes this option unattractive. It has been shown however, that torrefaction (a mild thermal treatment) can reduce the energy demands upon milling to only 0.01-0.02  $\text{kW}_e/\text{kW}_{\text{th wood}}$ . It may however be possible to take advantage of the relatively high reactivity of biomass compared to coal. It has been made plausible that biomass particles as large as 1 mm would suffice. This not only means a low electricity consumption upon milling, it also means that screw feeding is possible, which is very attractive compared to pneumatic feeding. This means less electric energy consumption for compression of inert gas, only little syngas dilution by inert gas and resultantly a higher efficiency. Furthermore, if piston feeder systems are used to pressurize the biomass fuel, advantages become even more pronounced.

On the basis of efficiency and the above-mentioned facts and opinions, one option is selected as the best choice for the production of syngas from biomass at large-scale. In this system, biomass is milled to 1 mm particles, compressed by piston feeder and subsequently fed by screw into the gasifier. This option has the lowest amount of unit operations and has the highest efficiency. It has been calculated that the efficiency from wood with 35% moisture to 40 bar syngas with  $\text{H}_2/\text{CO}=2$  is 81% (LHV). If net electricity production is included (calculated to primary energy with 40% efficiency), the overall efficiency is 84%. As alternatives, two options are selected where biomass is pre-treated: torrefaction or fast pyrolysis to produce torrefied wood or oil/char-slurry respectively. These systems show lower efficiency (overall efficiency is approximately 75%), higher investment and more complexity compared to the "1 mm" option, but pressurizing and feeding becomes conventional.

The next steps towards large-scale biosyngas plants by entrained flow gasification should focus on suitability of 1 mm particles, applicability of screw feeders and piston feeders, soot formation, gas cooling and ash properties. Economic evaluations should be done. A phased approach towards a large-scale plant is proposed consisting of lab-scale, pilot-scale and co-firing experiments.



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## SUMMARY

Biomass is heading for a great future as renewable energy source. Not only electricity and heat can be produced, also gaseous and liquid fuels may be synthesized from biomass. One way to achieve this is by gasification of the biomass fuel as an intermediate step. Recent studies have shown the attractiveness of entrained flow reactors for the gasification of biomass. The technology is not only commercially available on large scale (mainly for coal and liquid fuels), it also can reach the highest efficiency from biomass to so-called biosyngas. Biomass however, is different from coal in many respects. The most relevant differences refer to ash behaviour, feeding, and pressurizing properties. The underlying report is the result of a project carried out by ECN and Shell, where mentioned issues are addressed. On the basis of tests, calculations and interviews, design guidelines are obtained and systems have been selected for the entrained flow gasification of biomass to produce biosyngas.

### *Entrained flow gasification*

Two types of entrained flow gasifiers can be distinguished: slagging and non-slagging. In a slagging gasifier, the ash forming components melt in the gasifier, flow down the walls of the reactor and finally leave the reactor as a liquid slag. Generally, the slag mass flow should be at least 6% of the fuel flow to ensure proper operation. In a non-slagging gasifier, the walls are kept free of slag. This type of gasifiers is suitable for fuels with only little ash. The question is whether clean wood (approximately 1% ash) should be converted in a slagging or a non-slagging gasifier.

### *Ash behaviour*

Different lab-scale tests and thermodynamic modelling have shown that ash from biomass, particularly from clean wood, does not or hardly melt at typical operating temperatures of entrained flow gasifiers (1300-1500°C). This is caused by the fact that the ashes are rich in CaO, while alkali metals (which generally reduce melting temperatures) are removed by the gas phase. Despite the high ash melting temperatures of (clean) biomass, a *slagging entrained flow gasifier* is preferred over a non-slagging gasifier. The most important reasons are that (1) little melt (fouling) can never be avoided and (2) a slagging entrained flow gasifier is more fuel flexible. Slagging gasification of biomass requires fluxing material in order to obtain the proper slag properties at reasonable temperatures. Silica or clay seems to be the obvious choice. Furthermore, slag recycling might be needed to obtain enough wall coverage.

By adding the envisaged flux material to biomass, coal-based slag (generally coal ash with added limestone) is mimicked and slag properties become comparable. Experts therefore have the opinion that the gasification process does not bring about substantial problems if biomass is used instead of coal. This means that existing coal-based plants can be used for biomass tests or even transformed into a biosyngas production plant. This coincides with the foreseen increasing role of coal gasification in the coming decades. All experts however, agree on the fact that feeding of biomass will be the biggest challenge.

### *Feeding solids*

If biomass has to comply with the demands of coal-based (thus: existing, conventional and proven) systems, it is estimated that it should be pulverised to particles of 100 µm in size. This would consume as much as 0.08 kW<sub>e</sub>/kW<sub>th wood</sub> of electric energy relative to the heating value of the wood. Calculated to primary energy this means approximately 20%, which is unacceptable. Torrefaction is a mild thermal treatment, which renders biomass into a brittle product resembling coal. It has been shown experimentally that torrefaction pre-treatment can reduce the electricity consumption upon milling to only 0.01-0.02 kW<sub>e</sub>/kW<sub>th wood</sub>. At the same time,

torrefied wood powder has proven to be suitable for pneumatic handling. In an optimised system, dry wood can be converted to torrefied wood with 95% efficiency.

Trying to mimic coal however might not be the best approach to convert biomass into biosyngas. Due to the relatively high reactivity, biomass particles as large as 1 mm can probably be sufficiently converted upon entrained flow gasification. This has two major advantages: (1) pre-treatment (e.g. torrefaction) is not necessary since direct size reduction of biomass to 1 mm only consumes 0.01-0.02  $\text{kW}_e/\text{kW}_{\text{th wood}}$  and (2) screw feeders suffice (pneumatic feeding will not work). The latter advantage not only means less technical complexity compared to pneumatic feeders, it also means less inert gas consumption resulting in less electric energy consumption for compression and higher gasifier efficiency.

#### *Pressurizing solids*

The conventional way to pressurize solids is to use lock hoppers. Due to the low density and low heating value of biomass, lock hoppers have two major disadvantages: (1) large amounts of inert gas are required and must be compressed and (2) gasification efficiencies drop due to the dilution of the syngas. This becomes even worse if the fuel has to be pneumatically transported and injected into the gasifier. Pressurizing and subsequent pneumatically feeding of biomass fuel powder into a 40 bar reactor consumes approximately 0.025  $\text{kW}_e/\text{kW}_{\text{th wood}}$  electricity and reduces the efficiency from wood fuel to syngas with almost 0.04  $\text{kW}_{\text{syngas}}/\text{kW}_{\text{th wood}}$  (relative to the heating value of the fuel). This is hardly better than atmospheric gasification and downstream syngas compression. If wood particles of 1 mm suffice, the energy penalty is much less because screw feeders instead of pneumatic feeders can be used: 0.02  $\text{kW}_e/\text{kW}_{\text{th wood}}$  electricity and 0.012  $\text{kW}_{\text{syngas}}/\text{kW}_{\text{th wood}}$  syngas efficiency penalty. Moreover, if piston feeders instead of lock hoppers are used, the inert gas consumption reduces even further. The total efficiency penalties can reduce with more than 50%.

#### *System choice*

Different options to produce syngas from wood have been selected and efficiencies have been calculated using the flow sheet simulation package ASPEN-plus. On the basis of efficiency and the above-mentioned facts and opinions, option A as shown in Figure A is selected as the best choice for the production of syngas from biomass at large-scale. In this system, biomass is milled to 1 mm particles, compressed by piston feeder and subsequently fed by screw into the gasifier. Option A has the lowest amount of unit operations and has the highest efficiency. It has been calculated that the efficiency from wood with 35% moisture to 40 bar syngas with  $\text{H}_2/\text{CO}=2$  is 81%. If net electricity production is included (calculated to primary energy with 40% efficiency), the overall efficiency is 84%. Efficiencies are calculated on lower heating value.

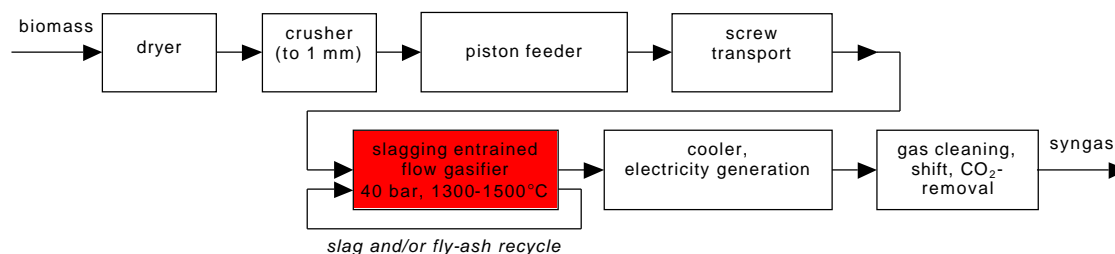


Figure A *Option A: the preferred system for the large-scale production of biosyngas*

Two main technical questions exist for option A. Firstly, it is not clear if (and how) a screw feeder can feed the solid fuel particles sufficiently constant into the gasifier. Secondly, it needs



to be proven experimentally that 1 mm biomass particles can be converted sufficiently in a slagging entrained gasifier.

If one of the questions cannot be answered satisfactorily, option B or C as shown in Figures B and C respectively will be preferred. In these options, the biomass is pre-treated in such a way that pressurizing and feeding will become conventional: torrefied wood resembles coal and slurry feeding is state-of-the-art. Options B and C can have an overall efficiency (including the electric energy flows, calculated to primary energy) of approximately 75%. The biggest challenge in both cases is to economically construct and operate large-scale torrefaction and pyrolysis processes.

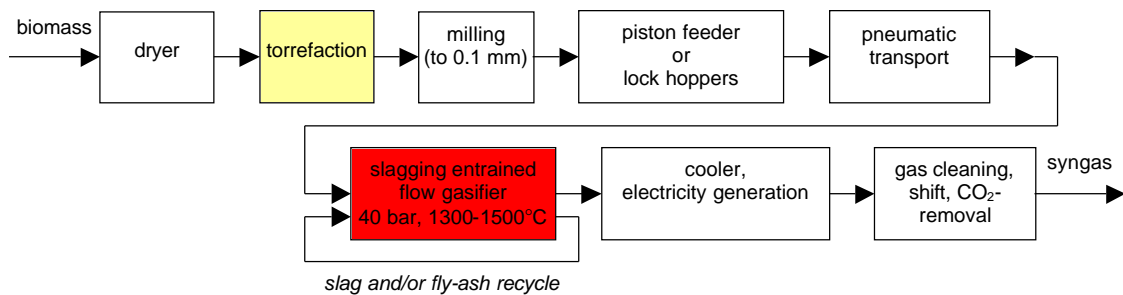


Figure B *Option B: alternative option for the large-scale production of biosyngas: pre-treatment by torrefaction to resemble coal*

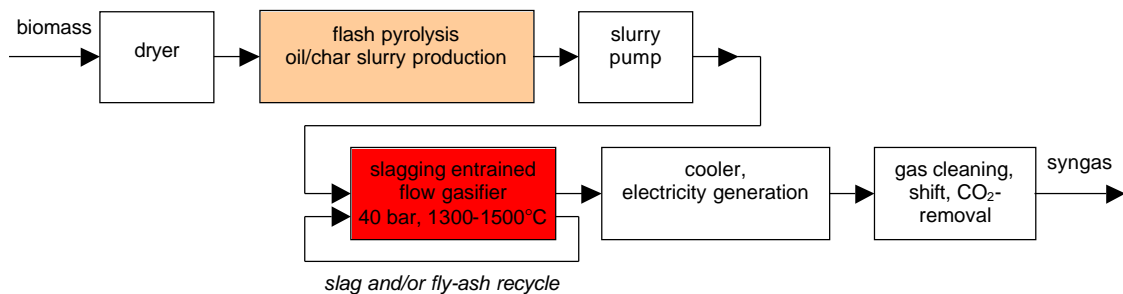


Figure C *Option C: alternative option for the large-scale production of biosyngas: pre-treatment by flash pyrolysis to produce oil/char-slurry*

It must be realized that oil/char-slurry not only is easy to compress and feed, but also transport and transshipment is relatively cheap compared to solid wood. If cheap and remote biomass is considered, option C can therefore be economically more attractive than the other options.

One alternative system needs to be mentioned because of its high efficiency: option D. In this option, biomass is first converted at relatively low temperature in an oxygen-blown fluidised bed reactor and the resulting product gas (including small solid particles consisting of unconverted fuel and ash) is introduced in an entrained flow gasifier. In this system, feeding is relatively easy since fluidised beds can handle fuel up to several centimetres. Furthermore, the overall efficiency is high: up to 85% depending on the feeding system. Because of the direct coupling of fluidised bed and entrained flow reactor, the system of option D will however be

relatively vulnerable for malfunctioning and therefore probably less reliable than options A, B and C. Furthermore, oxygen-blown pressurized fluidised bed gasification on large-scale is not state-of-the-art technology.

In all options discussed above, biomass ash ends up in the entrained flow gasifier. Therefore ash behaviour will be similar for these options. Generally spoken, this means that the choice for a slagging entrained flow gasifier, including flux addition and slag recycle, is valid for all options A-D. Although not subject of present work, gas cooling and ash utilisation issues also are expected to be similar for options A-D.

#### *Biosyngas development*

It has been shown that pre-treatment by fast pyrolysis to produce a liquid oil/char-slurry (option C) has a relatively low efficiency and therefore is less attractive than option A for expensive biomass. It must be realised however that feeding and pressurizing is not expected to be problematic in option C. Since ash related issues are supposed to be similar to other options containing an entrained flow reactor, option C therefore can be considered as a suitable method to research ash related items without having to deal with feeding and pressurizing issues in a first stage of development towards large-scale biosyngas production. Although ash related issues are not supposed to be critical by the experts, development and acceptance might benefit from this relatively simple first step.

At the same time, technical development should focus on the following issues related to feeding/pressurizing issues: (1) the chance that 1 mm biomass suffices, (2) the suitability of screw feeders for entrained flow gasifiers, and (3) the suitability of a piston feeder/disintegrator. Furthermore, soot may be mentioned as a research subject since the formation or suppression of soot significantly influences the efficiency. Also gas cooling and ash quality, which were not part of the scope of the reported work, are subjects that might need further attention. Finally, economic calculations need to be done to ultimately discriminate between different options. A phased approach towards a large-scale plant is proposed consisting of lab-scale, pilot-scale and co-firing experiments. Industrial participation is essential.

## 1. INTRODUCTION

### *Background*

Biomass is heading for a great future as renewable energy source. It not only is available in large quantities, it also is the only renewable energy source that is suitable for the production of (generally carbon containing) transportation fuels and chemicals. A promising option to do so is to convert biomass into a biosyngas by gasification and subsequently synthesize the required products.

In 2002, a joint project of Shell, ECN and BTG with input from Utrecht University and Ecofys has been performed in which different routes to produce biosyngas have been evaluated [2,3]. Important assumptions in this study were: clean wood as fuel, wood is available in the Baltic States at a local price of 4 €/GJ, large-scale syngas production in Rotterdam (8000 MW) to be realised within 8 years or in other words: the technologies must be relatively mature. Three routes were evaluated:

1. wood is collected and transported to Rotterdam, wood is dried and pulverised and converted in an oxygen-blown pressurised entrained flow gasifier, the syngas is cleaned and conditioned;
2. idem as 1, but the conversion takes place in an oxygen-blown pressurised circulating fluidised bed with subsequent entrained flow reactor (tar cracker);
3. wood is first converted into pyrolysis oil, the oil is transported to Rotterdam and converted in an oxygen-blown pressurised entrained flow gasifier followed by gas cleaning and conditioning.

It was concluded that the biomass to syngas efficiency was of decisive importance for the economics. Route 1, with entrained flow gasification of pulverized wood, turned out to be the winner. CFB-gasification of wood chips followed by entrained flow gasification was slightly more expensive. The third route via pyrolysis oil is approximately 30% more expensive per GJ of biosyngas, but this route might be attractive if cheap biomass is available at locations that require long distance road transport. The pyrolysis oil option becomes better if the oil is mixed with char to form a slurry [4]. In any case, entrained flow gasification is worth to be analysed into in more detail.

### *Problem definition*

Although many (large-scale) commercially operating entrained flow gasifiers exist (Appendix A), there is very little experience with biomass. What is clear however is that biomass cannot simply replace currently used fuels (coal, oil/residues). This is related to many differences like energy density, mass density, ash content, ash melting behaviour, etc.

### *Objective*

The objective of the underlying work is to obtain design guidelines for entrained flow gasification of biomass as part of an integrated biosyngas production plant. The major issue as originally defined, was ash-behaviour and the choice between slagging and non-slagging gasification. During the project, it has become clear that biomass feeding is another area of importance. Both issues will be addressed in this report. Furthermore, efficiencies of different options have been calculated to support the choice between technologies.



## 2. DEFINITIONS AND ASSUMPTIONS

The focus in the project is on ash behaviour in entrained flow gasification systems. In Appendix A, entrained flow gasification and some typical characteristics are described. During the project, the attention has been extended upstream towards feeding and pressurising.

### *Scale*

It is assumed that entrained flow gasification takes place on a large scale because (1) envisaged products (transportation fuels) have huge markets, (2) large-scale generally means lower specific costs and (3) heat integration is easier. The main arguments often used for small-scale (decentralised units need less logistics if fuels and/or products are available/needed locally) do not hold since (1) biomass is not locally available in quantities needed to serve the market of transportation fuels and (2) logistic costs of the liquid fuel product are negligible.

### *Biomass fuel*

Only clean wood is considered as a fuel, since this will be available in large quantities during a longer period. Given the limited availability of local clean wood in the Netherlands, the wood is supposed to be imported from e.g. the Baltic States.

For the tests, the following wood fuels have been used: beech, willow, a wood mixture<sup>1</sup>, and torrefied beech. Appendix B gives the chemical composition and heating values of mentioned wood fuels. Also shown is the composition of pyrolysis oil/char slurry.

### *Pre-treatment*

Generally, biomass needs to be pre-treated to be converted in an entrained flow gasifier. This often is related to the (physical) properties required for proper feeding and/or conversion. The following pre-treatment processes are considered in this study: (1) direct wood pulverization, (2) production of brittle solid by torrefaction<sup>2</sup> and subsequent pulverisation, (3) production of oil/char-slurry by flash pyrolysis, and (4) production of gaseous fuel by low temperature fluidised bed gasification. In the present study, it is assumed that the above-mentioned pre-treatment options do not influence the inorganic composition of the fuel. The conclusions regarding ash behaviour in the entrained flow gasifier therefore are assumed to be generally applicable.

The feeding method of the fuel into the entrained flow gasifier does however highly depend on the pre-treatment process. Relevant technical and economic issues will be addressed in this report.

### *Cooling*

The hot gas leaving the entrained flow gasifier at 1300-1500°C must be cooled before it can be cleaned, conditioned and used. Since the hot gas might contain molten particles, fouling of heat exchanger surfaces may occur. First installations will therefore probably use a gas or water quench. Quenching to 500°C probably will completely avoid ash related fouling. For the economic calculations, a gas quench to 900°C is assumed. Cooling technology is no subject of this project. It is realised however that biomass is different from coal in this respect and cooling biosyngas certainly needs further attention since it can have significant effects on economics.

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<sup>1</sup> "Wood mixture" is the fuel as currently used in a commercially operated biomass-to-electricity plant in the Netherlands.

<sup>2</sup> The torrefaction process has been studied in a separate SDE-financed project of ECN, Shell and TU/e called "Torrefaction for entrained flow gasification of biomass" (Torrefactie ten behoeve van entrained-flow vergassing van biomassa). In this report, some results will be mentioned. Complete results can be found in [8].

### *Ash/slag application*

The project will neither deal with the application of the produced ash or slag, nor will the possible applications be used as an argument for one or another gasification technology.

It has been said that a liquid slag encapsulates the inorganic elements from the biomass and prevents them from being recycled as fertiliser. This might break the mineral cycle and forces to produce fertilisers in an alternative (energy intensive) way. In the present study however, it is assumed that the mineral cycle is not an issue because (1) there is no need for fertilisation in long-rotation forests as leaves, needles and small branches are left in the wood and these parts contain the larger fraction of the essential minerals and (2) part of the minerals introduced in the high-temperature gasifier will end up as fly ash particles, which are very small (1-10  $\mu\text{m}$ ), better leachable and probably suitable as fertiliser.

Ash and slag application issues might be worth analysing once material is available and characteristics no longer have to be guessed.

### 3. ASH BEHAVIOUR

The ash behaviour has been said to be the major hurdle if biomass is converted in an entrained flow gasifier. Two types of entrained flow gasifiers can be distinguished, see Appendix A: slagging and non-slagging. In a slagging gasifier, (part of) the ash forming components melt in the gasifier, flow down the walls of the reactor and finally leave the reactor as a liquid slag. The amount of slag should be sufficient to ensure continuous slag flow. Generally, the slag mass flow should be at least 6% of the fuel flow. In a non-slagging gasifier, the walls are kept free of slag. Generally, this type of gasifiers is suitable for fuels with only little ash. The question is whether clean wood (approximately 1% ash) should be converted in a slagging or a non-slagging gasifier.

This chapter describes the results of lab-scale tests and modelling with the focus on ash behaviour. Furthermore, the opinion of different experts in this field are given.

#### 3.1 Experiments

##### 3.1.1 Ash melting temperatures

Ash from willow, produced at 850°C, has been subjected to ash melting temperature tests according to DIN 51730 in reducing atmosphere and performed by DBI-AUA GmbH in Freiberg. In addition, three mixtures with pure silica have been subjected to the same ash fusion tests. Table 3.1 shows the results. Pure willow ash does not melt at typical entrained flow temperatures (1300-1500°C). The addition of silica can significantly reduce the melting temperatures.

Table 3.1 *Measured ash melting temperatures of willow ash/silica mixtures*

ratio SiO <sub>2</sub> / willow ash [kg/kg dry]	0	0.9	1.9	2.4
sintering temperature	1380	1130	1080	1040
softening temperature	1540	1170	1140	1160
hemi-spherical temperature	1550	1200	1220	1410
flowing temperature	1560	1260	1340	1490

The most relevant temperature for slagging entrained flow gasifiers (flowing temperature) has its minimum for mixtures with 0.9 kg SiO<sub>2</sub> per kg dry wood ash. The value of 1260°C seems perfect for use in mentioned reactor types. The relation between the amount of silica and the melting temperature is in good agreement with thermodynamic modelling results, see Paragraph 3.2.

##### 3.1.2 LCS lab scale simulator

The slag behaviour of selected wood streams has been characterised under simulated entrained-flow gasification conditions. The atmospheric ECN lab-scale test facility LCS<sup>3</sup> has been used for this. The LCS has concentric burners, fired with different gases, to create the correct atmosphere and temperature for the inner burner where fuel particles are introduced. This way, entrained flow gasification is mimicked. At a certain distance from the burner, particles/ash can be collected on a hot “probe” consisting of a (ceramic) deposit plate. By examining the deposit

<sup>3</sup> LCS: Lab-scale Combustion and gasification Simulator

plate by SEM/EDX, information can be obtained on the deposition and slagging behaviour of the fuel and ash. Appendix C describes the LCS test facility.

The LCS has been operated at 1400-1450°C with a flame temperature of over 2000°C. The residence time of the particles in the hot zone is approximately 0.2 second. The residence time from burner to probe is approximately 0.1 second. The fuels used were: beech, torrefied beech, willow and a wood mixture. All fuels were milled until all mass passed a sieve of 250 µm. See Appendix B for chemical composition.

The main conclusion from the LCS tests is that wood ash is not prone to form a fluid slag at typical entrained flow gasifier temperatures. On the deposit plate, only few particles were trapped. Most of these particles were non-molten, irregularly shaped and CaO-rich. Only a small fraction has been molten. This fraction is characterised by Ca-silicates while also complex mixtures enriched in Ca, P and Si were observed in case of willow. Figure 3.1 shows a typical deposit where most material is not molten. Appendix C describes the results obtained in more detail.

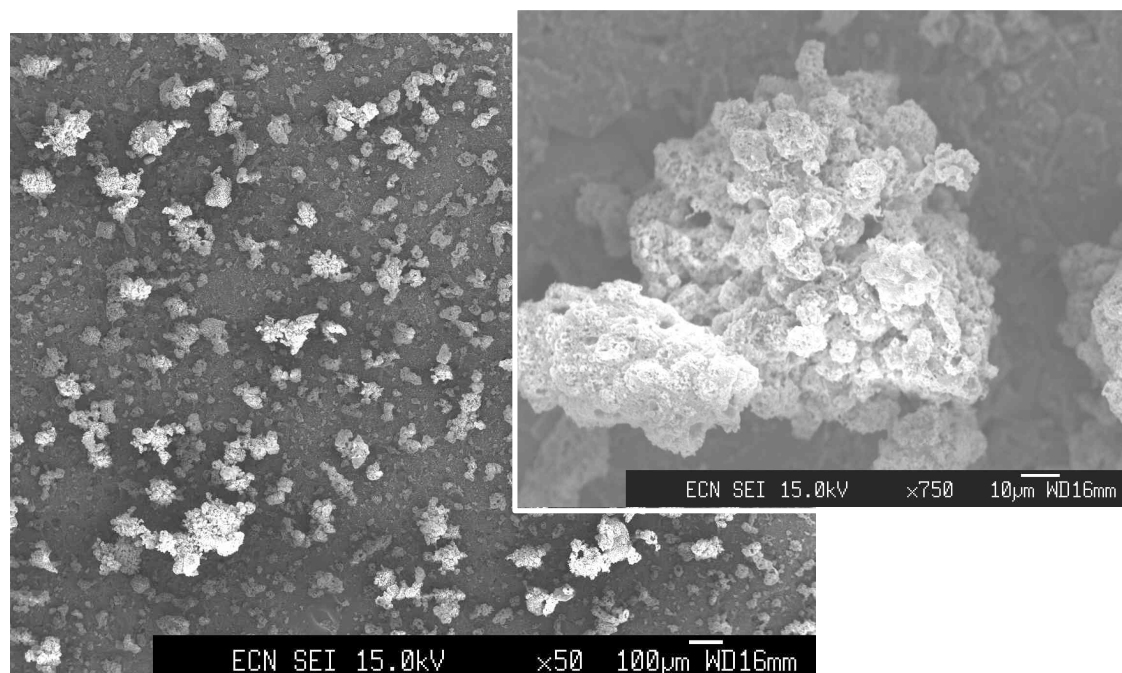


Figure 3.1 SEM pictures of typical deposit after gasification test with beech in the LCS facility, right picture is enlargement of picture on the left; material is hardly molten

An attempt to increase the fraction of melt by addition of pure silica (30 µm) to beech wood (3 kg silica/kg ash) resulted in the formation of a slag, which was molten to a higher extent than in the case without silica. About 50% was molten and consisted of mainly Ca-silicates as expected. The non-molten particles were either almost pure CaO or SiO<sub>2</sub>. Better mixing probably would have increased melt fraction.

Furthermore, torrefied beech and “normal” beech show similar results as far as ash slagging tendency is concerned. Using torrefied beech resulted in an even lower fraction of molten particles. This might be because of a reduction of silicon content upon torrefaction, see Appendix B. The conversion after 0.1 s was 96% for beech, 98% for the wood mixture and 88%



for torrefied beech<sup>4</sup>. It is expected however, that these biomass fuels will be completely converted in a large commercial gasifier, having typically 10 times longer residence times<sup>5</sup> and higher flame temperatures. Also pressure might have a positive effect on conversion, see Appendix D. Furthermore, particle residence times in slagging gasifiers can be further increased as (large) particles are trapped in the slow flowing liquid slag. The higher conversion in large-scale commercial plants compared to the LCS has been shown before<sup>6</sup>.

Finally, deposition plates in the LCS have been pre-treated to simulate commercial slagging gasifiers. The deposition plates were:

1. SiC,
2. SiC pre-oxidised in air at 1200°C, and
3. SiC covered with ash and sintered at 1200°C in reducing atmosphere.

It appeared that the amount of trapped particles increased going from 1 to 3. This confirms the idea that the “degree of stickiness” (increasing from 1 to 3) is of major importance for the chance that fuel/ash-particles bounce off the surface. For practical applications, this means that wood ash particles, once they will be trapped in a pre-existing melt on the wall of the gasifiers, can melt by interacting with low-temperature melting species.

From the LCS tests it can be concluded that wood ash is hard to melt at typical entrained flow reactor temperatures of 1300-1500°C, but a little melt cannot be avoided. Keeping the particles away from the walls might theoretically be possible, but in practice hard to achieve. Once a sticky layer is initiated, more particles are easily trapped. A non-slagging reactor therefore seems not applicable.

### 3.1.3 PEFG

The PEFG is a similar lab-scale test facility as the LCS. The main difference is the option to operate under pressure. The PEFG allows also for longer residence times than the atmospheric facility. A test has been performed using torrefied beech at 10 bars and with operating conditions (in terms of flame- and furnace temperature) similar to those used in the atmospheric LCS reactor. The PEFG facility and the results are described in Appendix C. The main conclusion is that the slagging behaviour under pressure is similar to the slagging behaviour as observed when using the atmospheric LCS: absence of a molten slag layer and occurrence of many CaO-rich particles.

## 3.2 Modelling

Slagging/melting tendencies of the selected fuels have been studied using a thermodynamic equilibrium model (FACTSAGE) minimising Gibbs free energy, and applying it to a hypothetical (pressurised) entrained-flow gasification system. The main results are given below. More elaborate results can be found in Appendix E and [5].

It appears that only 10-25% of the ash forming components of beech is liquid at typical operating temperatures of 1300-1500°C. The reason is due to the dominance of CaO, which melts at temperatures higher than 1700°C. At these conditions, and due to the absence of

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<sup>4</sup> The volatile content, expressed as wt% of dry and ash-free mass, is 84%, 85% and 77% for beech, wood mixture and torrefied beech respectively (Appendix B). The conversion expressed as fraction of volatile-free mass then becomes respectively 75%, 87% and 48%.

<sup>5</sup> Typical mean gas residence times for entrained flow gasifiers vary between 0.6- 2 seconds. Particle residence times can be very different depending on the type of gasifier. For example the power plant in Buggenum has tangentially fuel injection at the bottom. This results in relative long solids residence times compared to the gas. The effect of swirl on particle residence time is quantified in [35].

<sup>6</sup> Coal has been tested in the LCS under identical conditions as the biomass fuels mentioned in this report. Measured conversion of coal was 60-65%, whereas exactly the same fuel has 99% conversion in the large-scale entrained flow gasifier in Buggenum.

network-forming element (i.e., slag forming elements like Si), alkali's like K will stay in the gas phase at temperatures higher than 1000°C. This prevents the formation of K-containing components, which generally have low melting temperature.

By adding silica or clay (aluminium-silicate), the melting point reduces due to the formation of Ca-silicates and/or the presence of Al-silicates. Furthermore, by adding silica or clay, the material gains the ability to partly bind alkali's (mainly K), which further reduces the melting point. By adding silica with a ratio of 0.6-1.2 kg/kg fuel ash (Si/Ca molar ratio of 1-2), 50-80% of the ash melts at 1400°C. Adding clay results in similar effects. Figure 3.2 shows the calculated melting behaviour of beech ash with different additives.

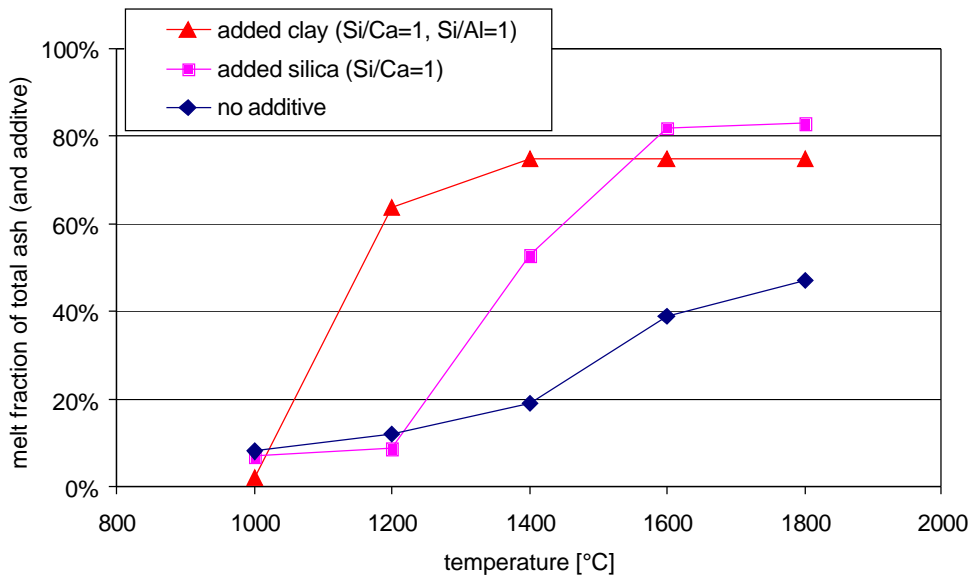


Figure 3.2 Calculated fraction of ash (plus additives) that is present in a liquid phase as function of temperature

### Viscosity

The slag viscosity is an important property for determination of the performance in a slagging gasifier, since the slag must be continuously removed and the slag must therefore be kept in a liquid phase. The slagging behaviour is critical to protect the refractory-lined walls of the gasifier from the harsh environment within the gasifier. Inadequate slagging can also lead to excessive refractory wear. Therefore, knowledge of the slag properties versus temperature and composition is essential.

For continuous tapping, the viscosity of the slag must be in the range 8-15 Pa·s at typical gasifier operating temperatures (tapping criterion). In addition, the tapping temperature must be high enough to avoid crystallisation of the slag in the tapping hole.

Most of the data available on slag viscosity (in terms of predictive models based on fuel ash composition) is related to coal ash. Coal ash however differs substantially from wood ash. It contains a relatively high content of clay minerals and low content of alkali and alkaline-earth metals. For ashes with high concentration of alkali metals, the available models therefore are hardly applicable. Another restriction is that P, which is present in some biomass ash, is not taken into account in viscosity models for coal ash. Therefore application of the slag viscosity models to wood ash is questionable, and no other good models or experimental data are currently available. It is believed that viscosity of the (fluxed) wood ash might be a relevant issue for future research in order to optimise reactor design and operating performance.

### 3.3 Expert opinions

Different experts have been consulted to get an answer to the question whether biomass should be converted in a slagging or non-slagging entrained flow gasifier. The experts are representatives of Shell, Future Energy, TKE and Schwarze Pumpe. All have the opinion that biomass should be converted in a slagging entrained flow gasifier. The main reasons are the high fuel flexibility of this type of gasifier and the chance that a little melt cannot be avoided completely. In order to obtain the proper slag properties, fluxing materials should be added. Furthermore, slag recycling may be necessary to obtain enough slag inside the reactor to ensure sufficient wall coverage.

### 3.4 Conclusions

It has been shown both experimentally and theoretically that ash from biomass, particularly clean wood ash, does not or hardly melt at typical operating temperatures of entrained flow gasifiers: 1300-1500°C. This is caused by the fact that the ashes are rich in CaO. A non-slagging entrained flow reactor therefore seems to be an obvious choice, since this also is the cheapest option. Nevertheless, a slagging entrained flow gasifier is preferred for biomass. The most important reasons are that (1) a little melt can never be avoided and (2) a slagging entrained flow gasifier is more fuel flexible. Fuel flexibility even extends to coal. The addition of fluxing agents like silica or clay is necessary. Furthermore, slag recycling might be needed.

The preference for a slagging reactor holds for the systems where biomass minerals end up in the entrained flow gasifier. This is the case for the system where pulverised biomass is introduced directly into the reactor, but also for the systems where biomass firstly is converted into oil/char-slurry by flash pyrolysis or torrefied wood by torrefaction or gas by low temperature fluidised bed gasification.

Choosing for a slagging entrained flow gasifier reactor for biomass has the consequence that coal-based know-how is (partly) applicable. This even goes further: by adding the envisaged flux material to biomass (silica-rich material or clay), coal-based slag (generally coal ash with added limestone) is mimicked and slag properties can become similar. Experts therefore have the opinion that the gasification process itself does not bring about substantial problems if biomass is used instead of coal. This not only holds for the slag behaviour, but also coal burners are assumed to be applicable for biomass. This means that existing coal-based plants can be used for biomass tests or even transformed into a biosyngas production plant. This matches with the foreseen increasing role of coal gasification in the coming decades<sup>7</sup>. Presently, tests with biomass are performed in different coal-fired gasification plants. The Buggenum plant (NUON-owned, entrained flow gasifier developed by Shell) has co-fired up to circa 6% biomass [6] and plans to co-fire as much as 25% biomass on energy basis in 2005 [7].

All experts however, agree on the fact that the feeding of biomass will be the biggest challenge. Pulverizing and subsequent transport and feeding at an elevated pressure proved to be very difficult for biomass so far. The next Chapter will deal with this subject.

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<sup>7</sup> Main reasons: contrary to coal combustion, CO<sub>2</sub>-sequestering is relatively easy/cheap and the product (syngas) can also be used for other applications like the production of transportation fuels. Furthermore, the compatibility with biomass (in slagging reactors) combines nicely with the various existing renewable energy objectives.



## 4. PARTICLE SIZE AND FEEDING

In the previous Chapter, it has been argued that no major hurdles are expected if biomass is converted to syngas in a *slagging entrained flow reactor*. With the proper fluxing materials, biomass and coal slag behaviour becomes similar. The analogy however does not hold for conversion and feeding of the fuel. The particle size plays a key role in both processes. Almost any fuel has to be reduced in size to be suitable for the entrained flow gasification process. The question is to what size. In the next paragraph it will be shown that biomass size reduction is very energy consuming. This means that there is an incentive to keep the biomass fuel particles large.

### 4.1 Size reduction

Pulverisation of biomass requires dedicated milling equipment. Electric power consumption turns out to be very dependent on required size reduction. At ECN, tests have been performed with dried biomass of different type and initial size in a standardised test set-up. Both dried wood and torrefied wood have been used. Figure 4.1 shows that electricity consumption starts to rise significantly if wood is milled to sizes below 1 mm. Pulverisation of wood to particles of 200  $\mu\text{m}$  requires as much as 0.05  $\text{kW}_e$  per  $\text{kW}_{th}$  (combustion enthalpy of the fuel). If based on primary energy, this is more than 10%!

From Figure 4.1 it appears that torrefaction can decrease electricity needs with 80%. At the same time (not shown), the capacity of a milling plant increases significantly. This means that torrefaction can reduce the costs of milling biomass if particle sizes of less than several 100  $\mu\text{m}$ 's are needed. For details on torrefaction and pulverizing torrefied wood, see [8].

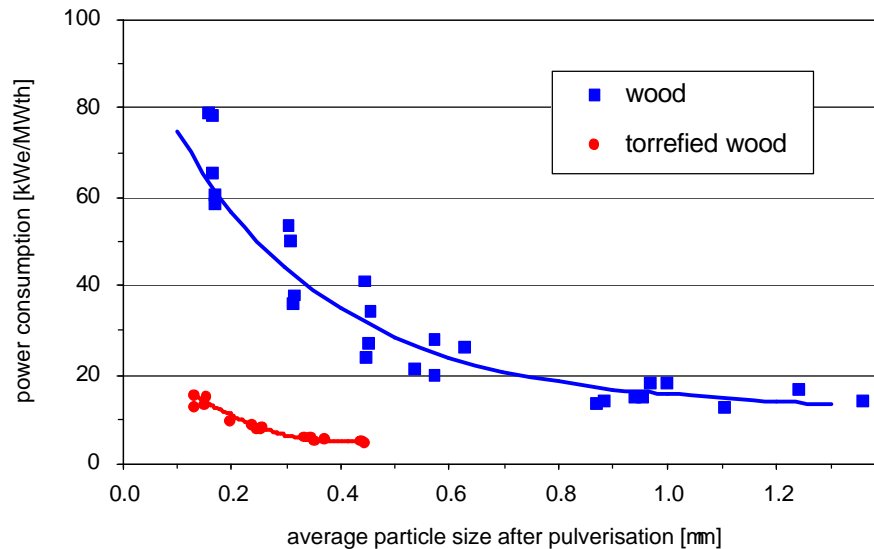


Figure 4.1. Measured power consumption upon size reduction in ECN standard test facility, more details in [8]

## 4.2 Particle size and conversion

The residence time of particles entrained by the gas flow in an entrained flow gasifier generally is limited to a few seconds. Most of this time, the particle will have the average gas temperature of 1300-1500°C<sup>8</sup>. This is the “optimum” temperature when using coal. At a lower temperature, coal is not converted completely, whereas higher temperatures lead to unnecessary energy losses. If biomass is used instead of coal, the minimum temperature is expected to remain 1300°C. This is assumed to be the temperature needed to crack hydrocarbons (including tar), which are formed as soon as biomass is heated.

In general, coal-fired entrained flow gasifiers (and also pulverised coal combustors) are operated on coal powder with a size of typically 50-100 µm. This ensures practically complete conversion; 99% is considered to be high. Because biomass is much more reactive than coal<sup>9</sup>, it is expected that size demands for biomass are less stringent. It is stated here, that biomass particles can be as large as 1 mm as far as conversion is concerned. Facts and calculations substantiating this statement are:

- Future Energy reports 97.6% conversion for 1.1 mm straw particles after 1.4 seconds at 1580°C in their entrained flow gasifier [9]. The straw fuel even contained particles up to 8 mm in size.
- In the Risø pilot-scale entrained flow reactor, tests have been done using both coal and straw [10]. It appeared that coal of approx. 150 µm was converted for about 80% in 2 seconds at 1100°C and 10 bar in 40% steam. Straw particles of 1-3 mm were converted for as much as 98% under the same conditions.
- Measurements at ECN in the LCS<sup>10</sup> with park wood particles up to 1 mm, resulted in a conversion of 92% within 130 ms. The conversion after 90 ms was already 87% [11,12]. Simple extrapolation would result in complete conversion of the fuel within 0.3 second.
- Tests in the coal-fired entrained flow gasifier of Elcogas in Puertollano in Spain with MBM (Meat and Bone Meal) of 500 µm grain size did not show any signs of insufficient conversion [13].
- Calculations by Twente University have been performed taking into account heat transfer and kinetics for conversion of biomass [14,15]. It has been calculated that 1 mm particles can be converted for over 99% within 1 second at 1300°C [16]. A particle of 2 mm would need 2.5 seconds for the same conversion.
- LCS-tests performed in this project (Chapter 3.1.2 and Appendix C) have shown that beech particles of 210 µm<sup>11</sup> are 96% converted at 1450°C in 0.1 s. This would mean that in about 0.2 seconds, conversion would be practically complete<sup>12</sup>. Extrapolation to larger particles, using the above-mentioned method of Twente University, would mean that 1 mm particles will be completely converted within 2 seconds.
- Tests with wood saw dust of approximately 1 mm in a pilot-scale CFB-gasifier<sup>13</sup> at ECN at 860°C reveal a conversion of 97%. The residence time of most of these particles is probably less than 1 second. It has to be mentioned that there is a chance that some particles are

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<sup>8</sup> In the flame close to the burner however, temperatures can be as high as 2500 or even 3000°C in a commercial plant.

<sup>9</sup> The reactivity of biomass during gasification is much higher than of coal. One of the reasons is that the volatile content is much higher (typically 80% and 35% of dry and ash-free mass respectively). The volatiles escape during heating and therefore represent “free” conversion. The higher volatile content also means that the remaining char is more porous resulting in less transport limitations. Another reason for the relatively high reactivity of biomass is the presence of elements like K in the fuel, which act as catalyst for gasification. Appendix J shows some experimental results, which prove the above-mentioned statements.

<sup>10</sup> LCS: Lab-scale test facility at ECN, see Appendix C for description.

<sup>11</sup> The median size is 210 µm, but 10% is larger than 700 µm

<sup>12</sup> If the conversion is expressed as wt% of volatile-free mass (volatile content: 84%), this is 75%. So, in another 0.1 seconds, 75% of the remaining 4 wt% will be converted. Total conversion will then be 99%.

<sup>13</sup> CFB: circulating fluidised bed, the ECN pilot plant has a capacity of 100 kg/h and generally uses silica sand as bed material

trapped by the circulating bed material of the CFB and have considerably longer residence time.

From the above given data, it might be concluded that biomass particles of 1 or even 2 mm can be accepted as fuel for entrained flow gasification at 1300-1500°C as far as conversion is concerned. This statement becomes even stronger if realised that (1) the burner zone in a full-scale gasifier is very hot and this will increase conversion compared to calculations and lab-scale tests, (2) larger particles can be trapped in the liquid slag in a slagging gasifier, which extends the residence time, and (3) due to the extreme full-scale burner conditions of high heating rates (typically  $10^5$  °C/s), there is a chance that biomass particles will explode into many smaller fragments when releasing volatiles. Furthermore, it can be said that possibly unconverted fuel may get a second chance since the slag and/or fly ash *must* (because of slag quantity, see Chapter 3) be recycled. Finally, it must be realised that in any case incomplete conversion has to be technically and economically compared with methods to increase conversion.

In conclusion: it can be stated that from a conversion point of view, biomass can be as large as 1 mm in size. This statement has been made plausible and holds for gasification in entrained flow gasifiers operating at temperatures of 1300-1500°C.

### 4.3 Particle size and feeding

In pulverised fuel reactors like an entrained flow gasifier, the feeding system sets limits to the particle size of the fuel.

#### *Coal-based feeding*

Pulverized fuel (generally: coal) feeding systems for pressurised reactors like the entrained flow gasifiers of NUON in Buggenum and Future Energy in Freiberg consist of the following three parts:

1. lock hopper(s) to pressurise the fuel; inert gas is used as pressurising gas, see Chapter 5;
2. a fluidised bed operated on inert gas;
3. a dense phase pneumatic transport line to feed the reactor; the fluidisation gas (plus additional gas if necessary) functions as transport gas.

In this system, it is essential to operate the fluidised bed in a non-bubbling fluidisation regime. The presence of bubbles would result in non-continuous feeding and corresponding fluctuations in temperature and other gasification conditions. In order to be able to fluidise without bubbles, particles should be so-called A-class according to Geldart [17]. In case of coal, this means that the maximum size is approximately 150 µm. Biomass may theoretically be a little larger due to its lower particle density: approximately 300 µm.

So, in order to use existing coal-based feeding systems, biomass particles should theoretically not be larger than approximately 300 µm. In practice however, this turns out not to be the only demand. Feeding tests at Future Energy with pulverised willow wood, having a median size ( $d_{50}$ ) of 210 µm, showed that proper fluidisation was impossible due to the fibre-like structure and the resulting high cohesion forces between the particles, see Appendix F. Another reason for the difficult fluidisation behaviour may have been formed by the broad particle size distribution of the powder (measured particle size values:  $d_{10}$ : 35 µm,  $d_{50}$ : 210 µm,  $d_{90}$ : 700 µm), probably also related to the presence of fibres. It can be speculatively concluded that coal-based feeding might only be suitable for biomass if fibres can be avoided or are small. This may be achieved by (1) “extreme” pulverisation or (2) pre-treatment of biomass in such a way that fibres are avoided. The first option is a theoretical option, it would cost too much energy (see

Figure 4.1). The second option can be realised by using torrefaction<sup>14</sup>: the brittle product can be pulverised relatively easy and also fibres break up. Both at Future Energy (Appendix F) and at ECN [8] fluidisation tests have been performed with pulverised fuel. It has been shown, that torrefaction indeed helps to get a more suitable material for fluidised bed/pneumatic feeders.

It can be concluded that *if existing coal-based feeders should be used*, biomass must be either pulverised to very small sizes (estimation: 100 µm or even smaller) or pre-treated by torrefaction and pulverised to particles of this size. Since milling biomass to 100 µm consumes 0.08 kW<sub>e</sub>/kW<sub>th</sub> and only 0.01-0.02 kW<sub>e</sub>/kW<sub>th</sub> is needed if the fuel is first torrefied (see Figure 4.1), the latter is preferred. This statement assumes that biomass should replace coal without major modifications in the feeding system. It may however be better to use a dedicated feeding system for biomass.

#### *Dedicated biomass-based feeding*

In Paragraph 4.2 it has been made plausible that biomass particles of 1 mm are small enough to be used in entrained flow gasification as far as conversion is concerned. Size reduction in this case will cost relatively little electricity: between 0.01 and 0.02 kW<sub>e</sub>/kW<sub>th</sub> according to Figure 4.1. Particles of approximately 1 mm cannot be fed through fluidised bed/pneumatic transport feeders as developed for coal powder, see above. A simple screw feeder however would suffice according to TKE [18]. Also ECN has good experience with their lab-scale fluidised bed gasifiers generally fired with 0.7-2 mm biomass particles.

Compared to pneumatic feeders, screw feeding is much simpler and it also has the advantage that less inert gas is introduced in the gasifier together with the solid fuel. This means that (1) less electricity is needed to compress the inert pressurising gas and (2) the gasifier efficiency increases due to less dilution. Appendix G elaborates on inert gas consumption and the consequences for electricity consumption and gasifier efficiency.

In conclusion, it can be said that two ways of feeding exist if solids are introduced into the gasifier, see also Table 4.1:

- The 100 µm approach: biomass, preferably (or actually: inevitably) pre-treated through torrefaction, is pulverised to approximately 100 µm particles and is fed through pneumatic feeding systems in analogy with systems based on coal as fuel.
- The 1 mm approach: biomass is reduced in size to approximately 1 mm and is fed by screw feeder.

Table 4.1 *The main advantages and disadvantages of the two approaches (see text) to use solid biomass for entrained flow gasification*

approach	advantages	disadvantages
100 µm approach (coal-mimic)	<ul style="list-style-type: none"> <li>• coal-based: proven technology, minimum modifications needed</li> <li>• complete fuel conversion in gasifier</li> </ul>	<ul style="list-style-type: none"> <li>• high electricity consumption upon milling <i>or</i> torrefaction needed as pre-treatment</li> <li>• high inert gas consumption</li> </ul>
1 mm approach (biomass-dedicated)	<ul style="list-style-type: none"> <li>• little pre-treatment necessary</li> <li>• low electricity consumption for size reduction</li> <li>• low inert gas consumption</li> </ul>	<ul style="list-style-type: none"> <li>• incomplete fuel conversion in gasifier?</li> <li>• new developments needed?</li> <li>• stability?</li> </ul>

From Table 4.1 and the efficiency difference as presented in Chapter 6, the “1 mm approach” seems preferable. It however should be proven that 1 mm particles could be converted

<sup>14</sup> Torrefaction is a mild thermal treatment (approximately 250°C and 30 minutes in absence of oxygen) resulting in a relatively brittle product



sufficiently in entrained flow gasification. Also questions arise if the screw feeder requires additional technical developments if used in the non-conventional combination with entrained flow gasification.

#### 4.4 Conclusions

If solid biomass has to comply with the demands of coal-based (thus: existing, conventional and proven) systems, it should be pulverised to particles of approximately 100  $\mu\text{m}$  in size. This would consume as much as 0.08  $\text{kW}_e/\text{kW}_{\text{th}}$  of electric energy. Calculated to primary energy this means approximately 20%, which is unacceptable. Torrefaction has been shown to be a pre-treatment process that can reduce the electricity consumption upon milling to only 0.01-0.02  $\text{kW}_e/\text{kW}_{\text{th}}$ . At the same time, torrefied wood powder seems to be suitable for pneumatic handling similarly as when using coal powder. In the optimum system, dry wood can be converted to torrefied wood with 95% efficiency.

Trying to mimic coal however might not be the best option for biomass. Due to the relatively high reactivity, biomass particles of as large as 1 mm can probably be sufficiently converted in entrained flow gasification. This has two major advantages: torrefaction is not needed since direct size reduction of biomass to 1 mm will cost only 0.01-0.02  $\text{kW}_e/\text{kW}_{\text{th}}$  and screw feeders suffice. The latter advantage not only means less technical complexity compared to pneumatic feeders, it also means less inert gas consumption resulting in less compression energy and higher gasifier efficiency.



## 5. PRESSURISING

Entrained flow gasification generally takes place at elevated pressure for two main reasons: (1) it concerns large-scale processes and (2) a high pressure is preferred in downstream synthesis for thermodynamical reasons. Solid fuels therefore must be pressurized prior to the injection into the gasifier.

### 5.1 Lock hoppers

Often, lock hoppers are used to pressurise solid fuel. This is a batch-wise system, where a vessel is filled with fuel, closed and subsequently pressurised with an inert gas. Once the desired pressure is reached, the vessel is emptied in a second vessel, generally needed as a buffer to compensate for the discontinuous solids flow resulting from the batch-wise operation of the lock-hopper system. From the second vessel (also called: feed bin), the solids can be transported to the reactor by pneumatic transport (as in coal-based plants), screw feeder or other methods.

Lock hoppers need inert gas as pressurising gas. This often is  $N_2$  and sometimes flue gas is used. In the case where biomass is converted into syngas, the obvious gas to use is  $CO_2$  since  $CO_2$  is present in large quantities in the gas and has to be removed anyway in order to increase (economic) efficiency. In addition,  $CO_2$  to some extent suppresses soot formation during gasification.

The amount of inert gas needed to pressurize solids in a lock hopper system strongly depends on the density of the fuel. Furthermore, the feed bin must be kept inert in order to avoid poisonous syngas to escape from the feeding system. This means that inert gas is needed to fill the volume of fuel particles removed from the feed bin. If pneumatic feeding is used, the gas needed for transportation prevent syngas to flow the wrong direction.

Appendix G contains calculated inert gas consumption quantities for different fuels. It is calculated that pressurizing biomass through lock hoppers needs 8-10  $m^3$  gas per ton of fuel or 0.5-0.6  $m^3/GJ$ . The electricity consumption for compression to 40 bar is approximately 25  $kW_e$  per  $MW_{th}$  combustion enthalpy of the fuel. If coal is used, the specific inert gas use and energy consumption drops with a factor of approximately 3, due to the relatively high density and high heating value of coal.

### 5.2 Piston feeding

TKE is specialised in biomass feeding under pressure. They developed and operate a piston feeder capable of feeding biomass against 20-bar pressure. The piston feeder is an alternative for the lock-hopper system and has the advantage of little volume and low inert gas consumption. Furthermore, less inert gas will enter the gasifier and gasification efficiency will hardly be influenced. Appendix G shows the calculated values based on experimental data in comparison with the lock-hopper system.

The test feeder of TKE consists of an atmospheric supply bunker, a piston feeder and a pressurised tank. The piston presses the fuel through a tapered opening, thus creating high radial pressures in the fuel plug. The feeder is designed in such a way that by releasing the piston (to feed another batch of fuel) the plug will stay in its position. The dense plug falls into the pressurised tank of the test facility. In real applications, this would contain a disintegrator and a mechanism to transport the disintegrated “briquettes” into the reactor. The piston feeder has

been tested successfully with different fuels. TKE is involved in the large EU-project “Chrisgas” where the lock hopper system of the pressurised CFB-gasifier in Värnamo in Sweden is replaced by a piston feeding system.

In order to assess and quantify the positive effects of a piston feeder, torrefied park wood has been sent to TKE for tests. The material has been produced at ECN.

The result of the test is described in Appendix I. In conclusion, it appeared that the standard test unit was suitable for feeding torrefied wood chips against 20-bar pressure. The resulting plug could easily be disintegrated. The material after feeding (the plug) has been subjected to milling tests at ECN to measure the electricity consumption. It turned out that the electricity consumption is approximately 25% less than the electricity consumption upon milling of the original torrefied wood chips. This probably is due to the size reduction already taking place during compression in the piston feeder.

The gas leakage through the piston feeder will be approximately  $0.1 \text{ m}^3/\text{ton}$  for a large-scale 40 bar system. Assuming that the disintegrated solids can be fed by screw, and the inert gas consumption is twice the leakage flow<sup>15</sup>, electricity consumption of CO<sub>2</sub>-compression would be only  $0.5 \text{ kW}_e/\text{MW}_{\text{th}}$ . The piston feeder would require  $8 \text{ kW}_e/\text{MW}_{\text{th}}$ . The cold gas efficiency will drop 0.05% due to the addition of inert gas into the gasifier. Both values are considerably lower than when using a lock-hopper system, see Appendix G.

### 5.3 Conclusions

Entrained flow gasification of biomass will generally require pressurizing equipment for the solids. The conventional way is the use of lock hoppers. Due to the low density and low heating value of biomass, lock hoppers have two major negative effects: (1) large amount of inert gas is required and must be compressed and (2) gasification efficiency drops due to the dilution of the syngas. It has been calculated that the total penalty on efficiency<sup>16</sup> is approximately 10% if wood powder is fed through lock hoppers and pneumatic feeders against 40 bar. If wood particles of 1 mm suffice, the energy penalty is approximately 6% because of the possibility to use screw feeders instead of pneumatic feeders. If however, lock hoppers are replaced by piston feeder, the inert gas consumption dramatically reduces. The energy penalty can be lower than 3% for 1 mm solids.

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<sup>15</sup> This doubling is meant to completely avoid syngas escaping from the feeder, it means that half the inert gas will flow through the feeder and the other half will flow into the gasifier.

<sup>16</sup> The efficiency penalty is the sum of the electricity consumption for compression of the inert gas (calculated to primary energy assuming 40% efficiency) and the reduction of cold gas efficiency of the gasifier due to the dilution of the syngas.

## 6. SYSTEMS

Different options to produce syngas from wood have been selected and efficiencies have been calculated using flow sheet simulation package ASPEN-plus. In all cases, clean wood with 35% moisture is used as fuel and syngas is produced at 40 bar with a H<sub>2</sub>/CO-ratio of 2. CO<sub>2</sub>-removal is part of the system. The specified syngas may be used for Fischer-Tropsch synthesis.

Seven options have been evaluated. In all options the wood is dried with waste heat, which is supposed to be available on the site. Figure 6.1 shows a schematic representation of the different options. Option 0 is the reference case in which wood is pulverized to 100 µm powder. As stated before, this consumes a lot of electricity and furthermore, it might not even be technically possible to feed the material pneumatically. The options A up to D are meant to be improvements on both issues. They differ in the way that wood is pre-treated for entrained flow gasification.

In the calculations, it is assumed that soot production in the gasifier is limited to 1% on energy basis. From recent tests at ECN simulating option D, it however has become clear that soot can be formed in large quantities [19]. Generally, it is concluded that soot formation is not understood and research is needed to quantify soot related effects in practice for the different options.

The overall efficiency is calculated for each option and included in Figure 6.1. The efficiency is defined as the lower heating value<sup>17</sup> of the syngas plus the net electricity production (calculated to primary energy with 40% efficiency) relative to the lower heating value of the wood (35% moisture). Electricity consumption includes (if applicable) pulverization, piston feeding, gas compression (CO<sub>2</sub>, O<sub>2</sub>, syngas) and O<sub>2</sub> production. In Appendix H, the different options are described in more detail together with the relevant assumptions used for the system modelling.

It can be concluded that option 0 has a very low efficiency, which can easily be improved by various pre-treatment options. As might be expected, option A has the highest efficiency. Moreover, this option is also the simplest as far as the amount of unit operations is concerned. A total efficiency of 84% is calculated, being the sum of 81% biosyngas and 3% electricity (as primary energy). Another attractive option from an efficiency point of view is option D where a “low temperature” gasifier is used to treat the wood prior to entrained flow gasification. The advantage of feeding wood chips instead of wood powder becomes visible. Option D can become even more attractive if piston feeders are used instead of lock hoppers.

Pre-treatment by torrefaction (option B) or flash pyrolysis (option C) appears to significantly decrease the total efficiency. This is caused by the fact that these processes have limited efficiency (95% and 85% respectively) and pressurizing and subsequent feeding of torrefied wood requires large amounts of inert gas.

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<sup>17</sup> The lower heating value (LHV) is the combustion enthalpy assuming that water in the flue gas remains in the gas phase (no condensation).

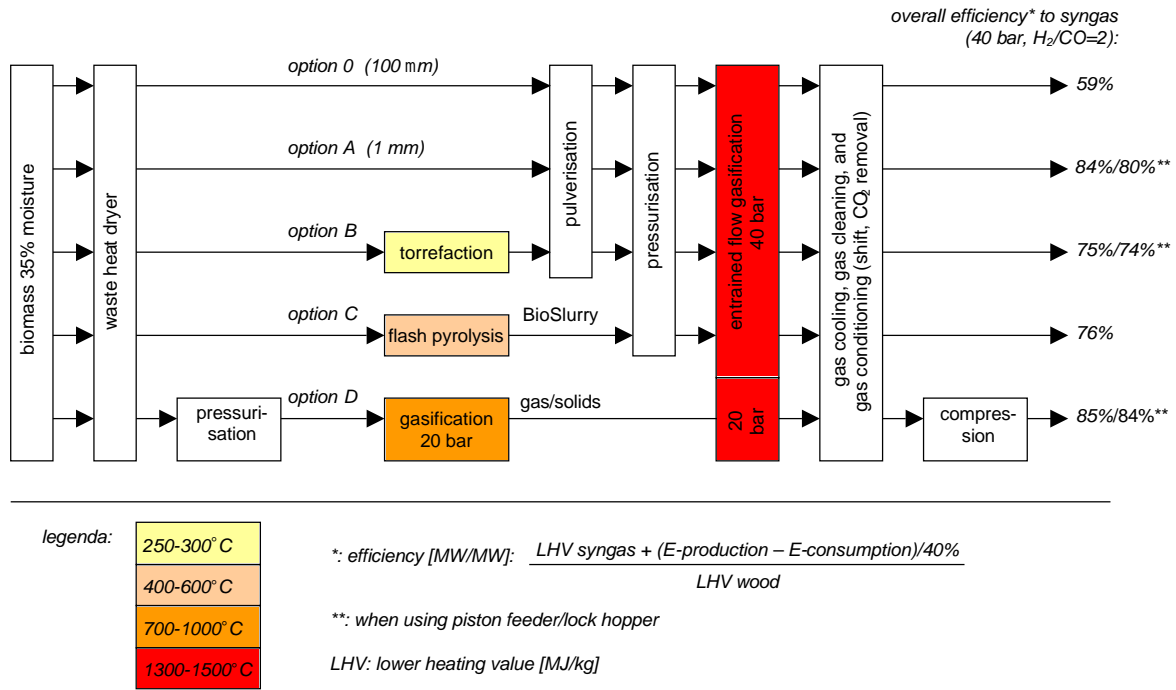


Figure 6.1 The biomass-to-syngas options considered including calculated overall efficiency to syngas (40 bar with H<sub>2</sub>/CO=2) and net electricity (calculated to primary energy); option 0 is the reference where biomass is pulverized to 100 mm particle size

## 7. DISCUSSION AND CONCLUSIONS

### *Ash behaviour*

Different lab-scale tests and thermodynamic modelling have shown that ash from biomass, particularly from clean wood, does not or hardly melt at typical operating temperatures of entrained flow gasifiers (1300-1500°C). This is caused by the fact that the ashes are rich in CaO, while alkali metals (which generally reduce melting temperatures) are removed by the gas phase. Despite the high ash melting temperatures of (clean) biomass, a slagging entrained flow gasifier is preferred over a non-slugging gasifier. The most important reasons are that (1) little melt (fouling) can never be avoided and (2) a slagging entrained flow gasifier is more fuel flexible. Slagging gasification of biomass requires fluxing material in order to obtain the proper slag properties at reasonable temperatures. Silica or clay seems to be the obvious choice. Furthermore, slag recycling might be needed to obtain enough wall coverage.

By adding the envisaged flux material to biomass, coal-based slag (generally coal ash with added limestone) is mimicked and slag properties become comparable. Experts therefore have the opinion that the gasification process does not bring about substantial problems if biomass is used instead of coal. This means that existing coal-based plants can be used for biomass tests or even transformed into a biosyngas production plant. This coincides with the foreseen increasing role of coal gasification in the coming decades. All experts however, agree on the fact that feeding of biomass will be the biggest challenge.

### *Feeding solids*

If biomass has to comply with the demands of coal-based (thus: existing, conventional and proven) systems, it is estimated that it should be pulverised to particles of 100 µm in size. This would consume as much as 0.08 kW<sub>e</sub>/kW<sub>th wood</sub> of electric energy relative to the heating value of the wood. Calculated to primary energy this means approximately 20%, which is unacceptable. Torrefaction is a mild thermal treatment, which renders biomass into a brittle product resembling coal. It has been shown experimentally that torrefaction pre-treatment can reduce the electricity consumption upon milling to only 0.01-0.02 kW<sub>e</sub>/kW<sub>th wood</sub>. At the same time, torrefied wood powder has proven to be suitable for pneumatic handling. In an optimised system, dry wood can be converted to torrefied wood with 95% efficiency.

Trying to mimic coal however might not be the best approach to convert biomass into biosyngas. Due to the relatively high reactivity, biomass particles as large as 1 mm can probably be sufficiently converted upon entrained flow gasification. This has two major advantages: (1) pre-treatment (e.g. torrefaction) is not necessary since direct size reduction of biomass to 1 mm only consumes 0.01-0.02 kW<sub>e</sub>/kW<sub>th wood</sub> and (2) screw feeders suffice (pneumatic feeding will not work). The latter advantage not only means less technical complexity compared to pneumatic feeders, it also means less inert gas consumption resulting in less electric energy consumption for compression and higher gasifier efficiency.

### *Pressurizing solids*

The conventional way to pressurize solids is to use lock hoppers. Due to the low density and low heating value of biomass, lock hoppers have two major disadvantages: (1) large amounts of inert gas are required and must be compressed and (2) gasification efficiencies drop due to the dilution of the syngas. This becomes even worse if the fuel has to be pneumatically transported and injected into the gasifier. Pressurizing and subsequent pneumatically feeding of biomass fuel powder into a 40 bar reactor consumes approximately 0.025 kW<sub>e</sub>/kW<sub>th wood</sub> electricity and reduces the efficiency from wood fuel to syngas with almost 0.04 kW<sub>syngas</sub>/kW<sub>th wood</sub> (relative to the heating value of the fuel). This is hardly better than atmospheric gasification and

downstream syngas compression. If wood particles of 1 mm suffice, the energy penalty is much less because screw feeders instead of pneumatic feeders can be used:  $0.02 \text{ kW}_e/\text{kW}_{\text{th wood}}$  electricity and  $0.012 \text{ kW}_{\text{syngas}}/\text{kW}_{\text{th wood}}$  syngas efficiency penalty. Moreover, if piston feeders instead of lock hoppers are used, the inert gas consumption reduces even further. The total efficiency penalties can reduce with more than 50%.

#### System choice

Different options to produce syngas from wood have been selected and efficiencies have been calculated using flow sheet simulation package ASPEN-plus. On the basis of efficiency and the above-mentioned facts and opinions, option A as shown in Figure 7.1 is selected as the best choice for the production of syngas from biomass at large-scale. In this system, biomass is milled to 1 mm particles, compressed by piston feeder and subsequently fed by screw into the gasifier. Option A has the lowest amount of unit operations and has the highest efficiency. It has been calculated that the efficiency from wood with 35% moisture to syngas at 40 bar with  $\text{H}_2/\text{CO}=2$  is 81%. If net electricity production is included (calculated to primary energy with 40% efficiency), the efficiency is 84%. Efficiencies are based on lower heating value.

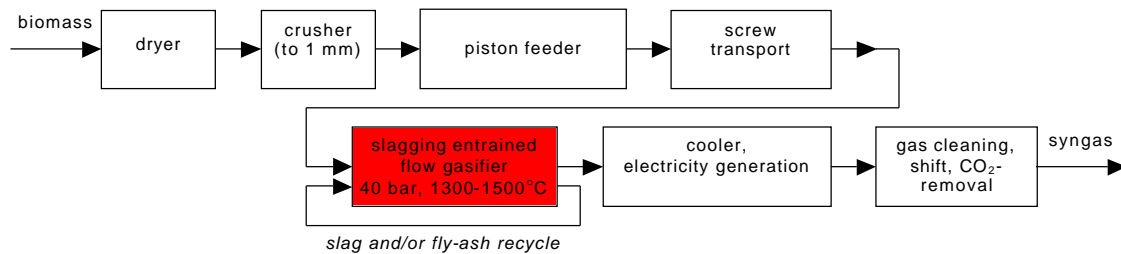


Figure 7.1. Option A: the preferred system for the large-scale production of biosyngas

Two main technical questions exist for option A. Firstly, it is not clear if (and how) a screw feeder can feed the solid fuel particles sufficiently constant into the gasifier. Secondly, it needs to be proven experimentally that 1 mm biomass particles can be converted sufficiently in a slagging entrained gasifier. If one of the questions cannot be answered satisfactorily, option B or C as shown in Figure 7.2 and Figure 7.3 respectively will be preferred. In these options, the biomass is pre-treated in such a way that pressurizing and feeding will become conventional: torrefied wood resembles coal and slurry feeding is state-of-the art. Option B and C have an overall efficiency (including the electric energy flows, calculated to primary energy) of approximately 75%. The biggest challenge in both cases is to economically construct and operate large-scale torrefaction and pyrolysis processes.

It must be realised that oil/char-slurry not only is easy to compress and feed, but also transport and transshipment is relatively cheap compared to solid wood. Especially if cheap and remote biomass is considered, option C might be economically more attractive than the other options.



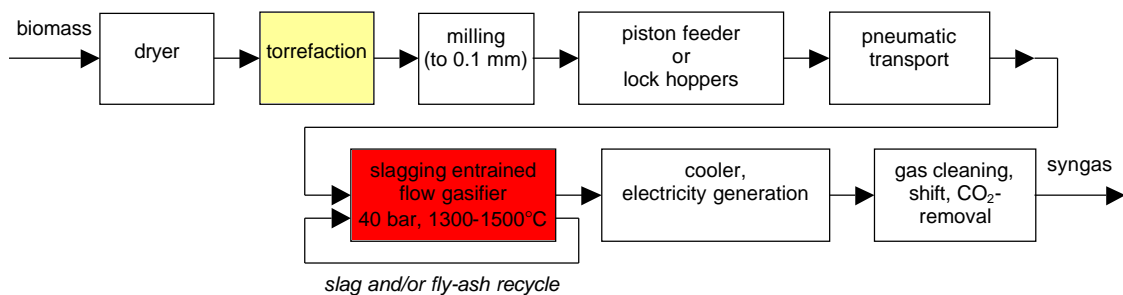


Figure 7.2. *Option B: alternative option for the large-scale production of biosyngas: pre-treatment by torrefaction*

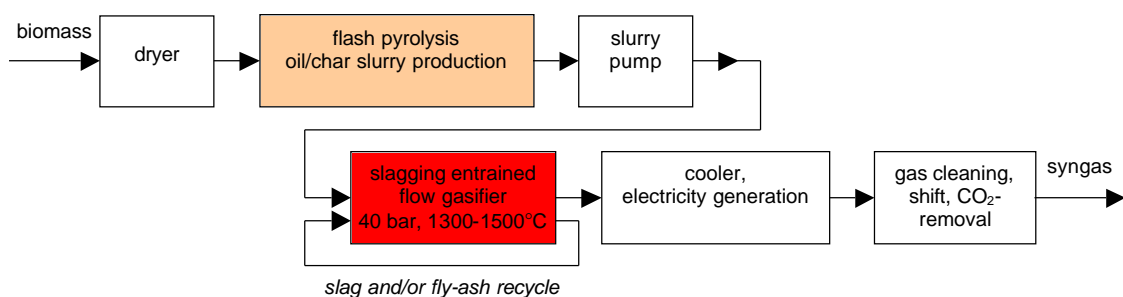


Figure 7.3. *Option C: alternative option for the large-scale production of biosyngas: pre-treatment by flash pyrolysis*

One alternative system needs to be mentioned because of its high efficiency. In option D, biomass is first converted at relatively low temperature in an oxygen-blown fluidised bed reactor and the resulting product gas (including small solid particles consisting of unconverted fuel and ash) is introduced in an entrained flow gasifier. In this system, feeding is relatively easy since fluidised beds can handle fuel up to several centimetres. Furthermore, the overall efficiency is high: up to 85% depending on the feeding system. Because of the direct coupling of fluidised bed and entrained flow reactor, the system of option D will however be relatively vulnerable for malfunctioning and therefore probably less reliable than options A, B and C. Furthermore, oxygen-blown pressurised fluidised bed gasification on large-scale is not state-of-the-art technology.

In all options mentioned above, biomass ash ends up in the entrained flow gasifier. Therefore ash behaviour will be similar for these options. Generally spoken, this means that the choice for a slagging entrained flow gasifier, including flux addition and slag recycle, is valid for all options A-D. Although not subject of present work, gas cooling and ash utilisation issues also are expected to be similar for options A-D.

#### *Biosyngas development*

It has been shown that pre-treatment by fast pyrolysis to produce a liquid oil/char-slurry (option C) has a relatively low efficiency and therefore is less attractive than option A for expensive biomass. It must be realised however that feeding and pressurizing is not expected to be problematic in option C. Since ash related issues are supposed to be similar to other options containing an entrained flow reactor, option C therefore can be considered as a suitable method to research ash related items without having to deal with feeding and pressurizing issues in a first stage of development towards large-scale biosyngas production. Although ash related

issues are not supposed to be critical by the experts, development and acceptance might benefit from this relatively simple first step.

## 8. FUTURE WORK

### *Technical issues*

In the previous Chapter, the best options have been presented. In order to make the final decision in favour of the selected option (option A: 1 mm biomass particles directly fed into entrained flow gasifier), two items need clarification:

- find out what the **conversion** can be of 1 mm biomass particles in a slagging entrained flow gasifier,
- find out whether **screw feeders** can fulfil the demands of an entrained flow gasifier.

Generally, **piston feeding** is the better option for pressurizing solid fuels as far as efficiency is concerned. Although different tests have been successfully performed with different fuels (in this project torrefied wood chips have been tested), it needs to be proven that it can indeed be used in the selected options. Also the **disintegrator** is of major importance. Furthermore, **soot** formation and related steam consumption in entrained flow gasifiers for different (pre-treated) fuels needs to be quantified. Finally, **economic** calculations should be performed.

### *Vision*

The ECN approach for the research and development trajectory is aimed at the implementation of large-scale biosyngas production. This approach is the concretisation of the ECN “Biosyngas Visions”. In the ECN vision it is believed that biosyngas will be an important intermediary for the production of renewable chemicals and fuels. Typically, biosyngas will be produced and consumed in large centralised industrial areas, located close to ports for effective biomass logistics. For cost-effective use of biomass, high biomass-to-biosyngas efficiencies are required and the optimum gasification technology to establish this is pressurised slagging entrained flow gasification. Woody and straw-like materials will be the main biomass feedstock for biosyngas production, for which different pre-treatment and feeding options have to be applied.

The research and development activities of ECN are concentrated on the biosyngas production from solid woody biomass materials. Wood is, and will be in the future, the main source of biomass for energy applications. Based on wood, the highest biosyngas efficiencies can be obtained. Grass-like materials however, have the benefit of a lower price and also are available in large quantities worldwide. It is therefore realised that grass-like fuels, pre-treated by flash pyrolysis into an oil/char-slurry, may also play an important role in future biosyngas production.

### *Development and time scale*

Within ECN, a phased approach has been set up to reach the objective of a large-scale slagging entrained flow gasifier operated on 100% solid woody biomass. Figure 8.1 contains a schematic representation of this approach. Figure 8.2 contains a time schedule. It is realised that industrial participation is essential to reach the final stages of development.

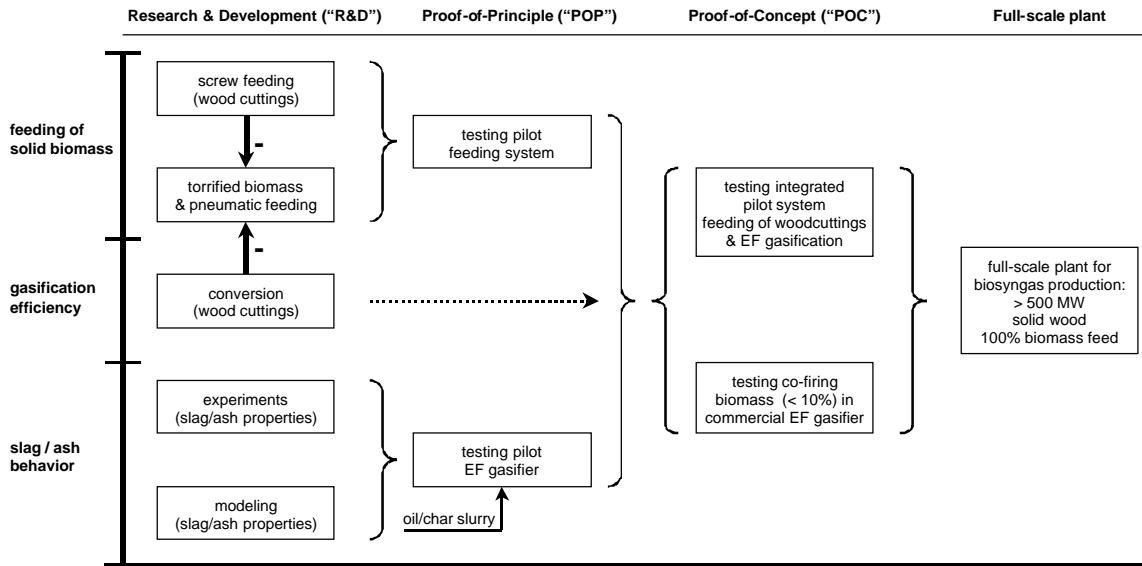


Figure 8.1. Approach to come to large-scale biosyngas plant on solid biomass; EF: entrained flow; see Figure 8.2 for time schedule

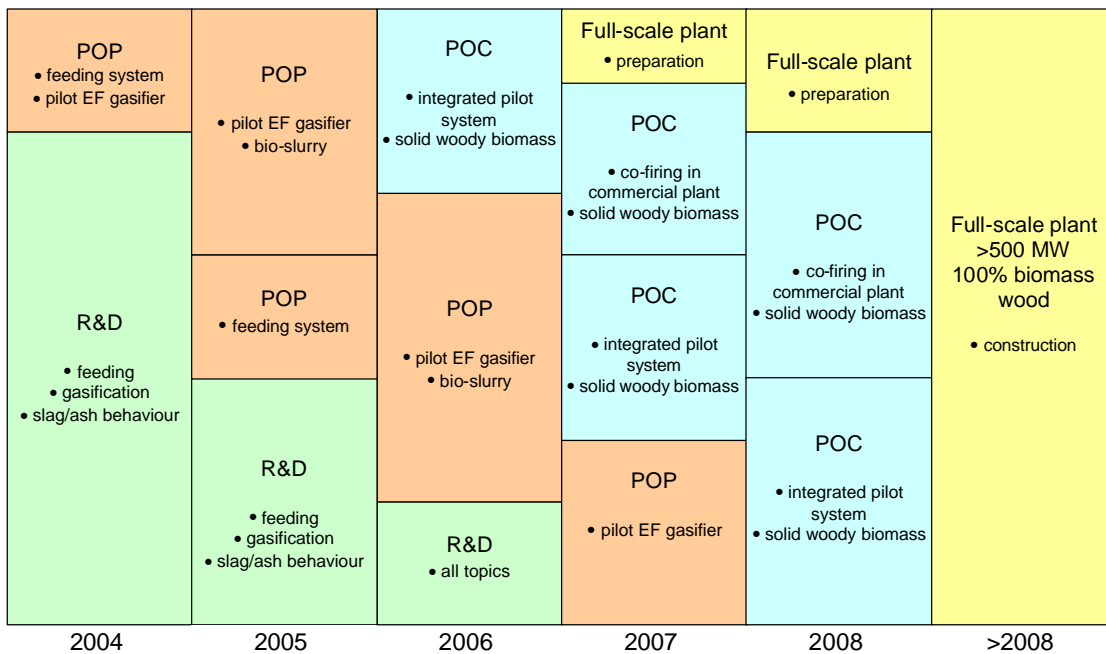


Figure 8.2. Time schedule for the different phases as indicated in Figure 8.1

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## APPENDIX A ENTRAINED FLOW GASIFIERS

An entrained flow gasifier (German: Flugstromvergaser, Dutch: stofwolkvergasser) is characterized by fuel particles dragged along with the gas stream. This generally means short residence times (typically 1 second), high temperatures (typically 1300-1500°C) and small fuel particles (solid or liquid, typically <100 µm). Furthermore, entrained flow gasifiers often are operated under pressure (typically 20-50 bar) and with pure oxygen. The capacity often is in the order of several hundreds of MW.

Pulverized solid fuels generally are introduced in the gasifier (after being pressurized, mostly using a lock hopper system) by pneumatic feeding. The powder is moved by inert gas and injected in a so-called burner in the gasifier. The burner intends to realize a good mixing between solid fuel and oxygen. Often vortex flow patterns are created in the burner. Local temperatures in the burner zone can be 2500°C or even higher. In the case of liquid fuels or slurries, pressurizing systems can be simply pumps. The liquid fuel subsequently is atomised and fed to the burner similarly to solid fuel powder.

Since entrained flow gasifiers operate at high temperature, it results in a CO- and H<sub>2</sub>-rich gas (and inevitably also CO<sub>2</sub> and H<sub>2</sub>O). Depending on the type of fuel, also soot may be formed. Soot formation often is suppressed by adding steam, typically 0.1 kg steam per kg oxygen.

Two types of entrained flow gasifiers can be distinguished:

1. slagging
2. non-slagging.

### *Slagging entrained flow gasifier*

In a slagging gasifier, the ash forming components melt in the gasifier. The molten particles condense on the relatively cold walls and ultimately form a layer being solid close to the wall and liquid on the inner side. This slag layer serves as a protective layer for the wall. The liquid slag is removed from the bottom of the gasifier.

In order to generate a liquid slag with the right viscosity at the given temperature, generally so-called fluxing material must be added. For coal fired plants, this often is limestone or another Ca-rich material.

Slagging entrained flow gasifier manufacturers are Shell, Texaco, Krupp-Uhde, Future-Energy (formerly Babcock Borsig Power and Noell), E-gas (formerly Destec and Dow), MHI (Mitsubishi Heavy Industries), Hitachi and Choren (formerly UET).

One example of a large-scale entrained flow gasifier is the 600 MW<sub>th</sub> coal-fired Shell gasifier in Buggenum, the Netherlands [20]. It is owned by the utility company NUON and produces electricity with a net efficiency of 43%. Tests have been performed with different kinds of biomass like wood, sewage sludge and chicken manure up to approximately 6% on energy basis. It is planned to co-fire 25% biomass on energy basis in 2005 [7,21]. Shell has signed several contracts to make similar coal gasifiers for fertilizer industries in China [22].

Another example is a 130 MW<sub>th</sub> gasifier (25 bar) made by Noell (presently Future Energy), operating on waste oil and sludges on the premises of the Schwarze Pumpe in Germany [23]. Figure A1 shows this gasifier, where the bottom half of the reactor is gas cooler by water quench.

The largest coal-fired entrained flow gasifier is located in Puertollano and owned by the utility company Elcogas [24]. It can produce 320 MW<sub>e</sub>. The technology is from Krupp-Uhde (Prenflo) and similar to the Shell technology. Tests have been performed with MBM (Meat and bone meal) up to 4.5wt% [13].

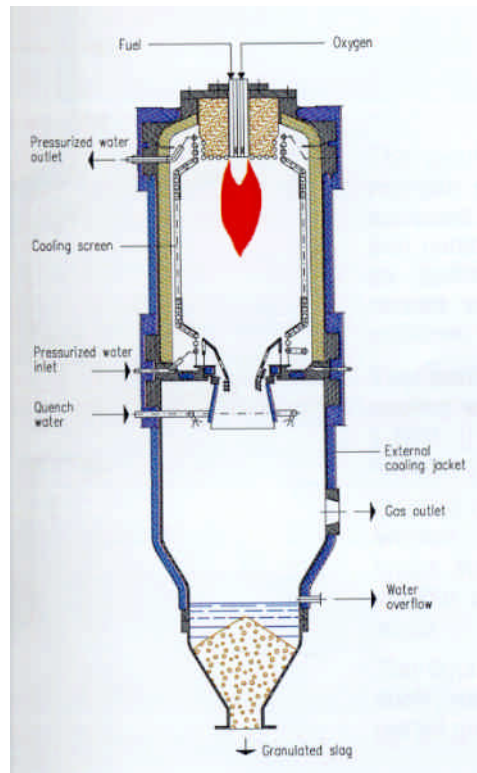


Figure A1 *Schematic representation of the entrained flow reactor of Noell (presently Future Energy) [25]*

*Non-slugging entrained flow gasifier*

In a non-slugging gasifier, slag is not produced. In practice, this means that fuels should contain only little amounts of minerals/ashes. Generally 1% is the maximum allowable ash content. An example of this type of gasifier is the SGP (Shell Gasification Process) on oil residues in Pernis, the Netherlands [20]. A certain amount of soot often is deliberately produced to generate condensation surface in the gas to prevent fouling of the walls.

## APPENDIX B FUEL COMPOSITION

Table B1 *Chemical composition of fuels, wet: on wet basis, dry: on dry basis, daf: on dry and ash-free basis*

		willow	beech	torrefied beech	wood mixture	pyrolysis oil <sup>(a)</sup>	oil/char- slurry <sup>(b)</sup>
water	wt% wet	5	9	5	5	22	19
volatiles	wt% daf	84	84	77	85	67	60
ash	wt% dry	1.9	1.0	1.3	1.5	0.05	2.3
HHV	MJ/kg daf	19.8	20.2	21.9	19.2	24.3	25.0
LHV calculated	MJ/kg wet	17.1	16.8	19.2	17.6	17.3	18.3
C	wt% daf	50.0	49.0	56.1	49.3	57.2	60.5
H	wt% daf	6.1	6.2	5.7	6.1	6.8	6.3
O	wt% daf	42.9	43.4	40.4	44.3 <sup>(c)</sup>	35.6 <sup>(c)</sup>	32.8 <sup>(c)</sup>
N	wt% daf	0.6	0.2	0.2	0.3	0.3	0.3
S	wt% daf	0.06	0.02	0.02	0.02	0.05	0.05
Cl	wt% daf	0.020	0.004	0.007	0.019	0.04	0.04
Al	mg/kg dry	60	48	42	95		
As	mg/kg dry	1	1	1	1		
Ca	mg/kg dry	5720	3100	3900	2450		
Cd	mg/kg dry	2	0.1	0.1	0.3		
Co	mg/kg dry	0.6	0.5	0.1	1.7		
Cr	mg/kg dry	14	3	2	5		
Cu	mg/kg dry	13	1	2	3		
Fe	mg/kg dry	68	47	42	110		
K	mg/kg dry	2894	1150	1450	970		
Mg	mg/kg dry	524	370	460	252		
Mn	mg/kg dry	10	56	64	72		
Mo	mg/kg dry	0.5	0.7	0.7	3.7		
Na	mg/kg dry	210	9	8	53		
Ni	mg/kg dry	26	4	12	2		
P	mg/kg dry	708	90	106	190		
Pb	mg/kg dry	238	1	1	2		
Sb	mg/kg dry	3	26	10			
Se	mg/kg dry	1		0.5			
Si	mg/kg dry	618	170	182	420		
Sn	mg/kg dry	2	0.1	0.1	<0.5		
Sr	mg/kg dry	14	5.1	6.1	10		
Te	mg/kg dry	1					
Zn	mg/kg dry	97	4	4	19		

<sup>(a)</sup> average value taken from [26]

<sup>(b)</sup> calculated values using composition of flash pyrolysis oil and low temperature wood char [26], assumed ratio of oil/char is 4.6 (70% and 15% respectively, based on dried wood) on LHV-basis

<sup>(c)</sup> calculated from [C]+[H]+[O]+[N]+[Cl]+[S]=100%

## APPENDIX C LAB-SCALE GASIFICATION TESTS AT ECN

This Appendix is a summary of the lab-scale tests performed at ECN. The complete results of these tests can be found in [5]. The tests have been performed together with thermodynamic modelling. The modelling results are summarized in Appendix E.

### LCS

The lab-scale test rig (Lab-scale Combustion and gasification Simulator, LCS), applied in this study is schematically depicted in Figure C1. The ring-shaped, concentric, staged gas burner is used to simulate the (high) initial heating rates, and serves as a source for the reaction atmosphere. The alumina reactor, placed in a two-stage electrically heated furnace, is designed to mimic the temperature-time history that coal/biomass/char particles experience in entrained-flow gasifiers. The residence times possible to achieve in the system are a couple of hundreds milliseconds scale. For the tests, the reactor furnace temperature was set at 1450°C, while flame temperature has reached approx. 2050°C.

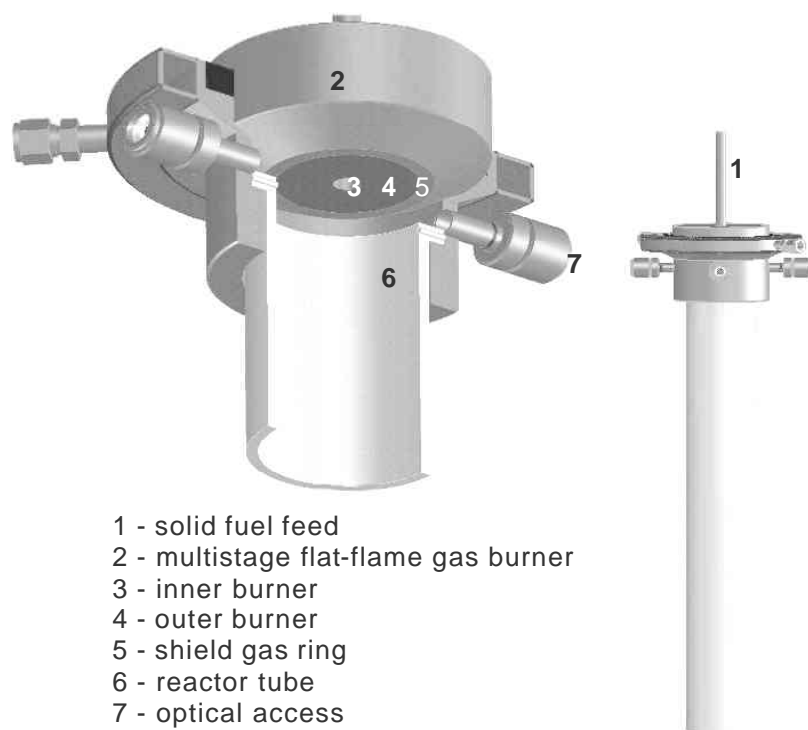


Figure C1 Schematic representation of the LCS test rig

The slagging behaviour of the ash in the LCS has been characterised by means of a deposition probe (Figure C2), on top of which an alumina deposition plate was placed, kept uncooled. The probe can be set axially at different positions along the reactor, thus simulating different particle residence times. The probe, couple with a deposit plate, had been extensively used in other studies [27] to evaluate the slagging behaviour of different fuels under combustion and gasification conditions. SEM/EDX techniques have been applied to characterise the slag sample obtained.

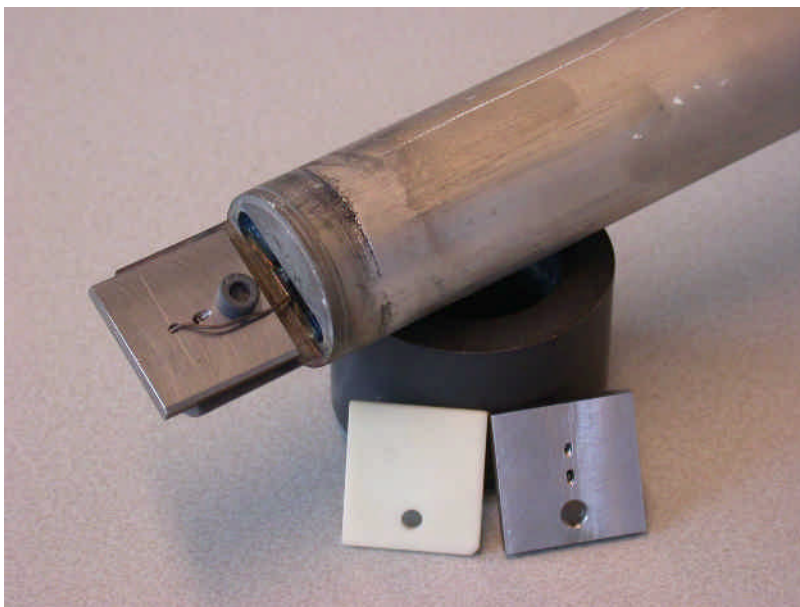


Figure C2 Photograph of the deposition probe and substrates

### *PEFG*

The PEF (pressurised entrained flow gasifier) of ECN is similar to the LCS, but can operate at elevated pressures [28,29]. Figure C3 shows the PEF schematically. The operation conditions have been chosen in such a way to be as close as possible to the conditions set in the atmospheric installation LCS in terms of temperature profile and residence time. The main difference between the two installations is therefore the operative pressure (10 bar).

Experiments at the PEF were performed with the insertion of an alumina deposition plate at the top of the probe. SEM/EDX techniques have been applied to characterise the slag sample obtained.

### *Fuels*

Experiments in the LCS have been performed with four different woods: beech, torrefied beech, willow, and a mixed wood that is commonly utilized in a Dutch power station. Fuel composition is shown in Appendix B. The feeding rate was kept constant at 2.0 g/hr throughout all the experiments. Experiments in the PEF have been performed with torrefied beech.

### *Additives*

One test has been done in the LCS using beech with added silica sand. The silica sand contains about 99% SiO<sub>2</sub> and the particle size was less than 30 µm. The ratio silica sand to wood ash was approximately 3.

### *Results*

All wood samples turn out to show similar ash behaviour, both in PEF and LCS. The main conclusion is that wood ash is not prone to form a fluid slag at typical operating temperatures of slagging gasifiers (1300-1500°C). Figure C4 shows the top view of the deposit plate after beech gasification after an experiment of 2 hours duration time. On the top of the deposit probe no uniform melt/ash layer was found, but rather single (clusters of) ash particles. Figure C5 shows a SEM picture of a single particle of beech ash, in which the original wood particle structure is still recognizable.

Although the bulk material on the deposition probe is not molten, a few particles can be found which actually have been molten. The melt is characterised by a relatively high concentration of silicon (wood mixture) or phosphor (willow). The ash and slagging behaviour of torrefied beech

was similar to beech. Wood mixture appears to generate a little more melt than the other wood fuels.

The test with beech and added silica sand resulted in a deposition, which was molten for about 50%. The molten parts mainly constituted of Ca-silicates, the non-molten particles were mainly CaO and SiO<sub>2</sub>.

Some tests have been done with deposit plates, which were covered with sintered ash prior to the gasification test in order to create a sticky substrate similar to a slag layer in a commercial entrained flow gasifier. It turned out that the ash particles reaching the substrate are completely encapsulated in the pre-existing melt. Furthermore, the ash particles are unrecognisable and probably have chemically interacted with the molten layer.

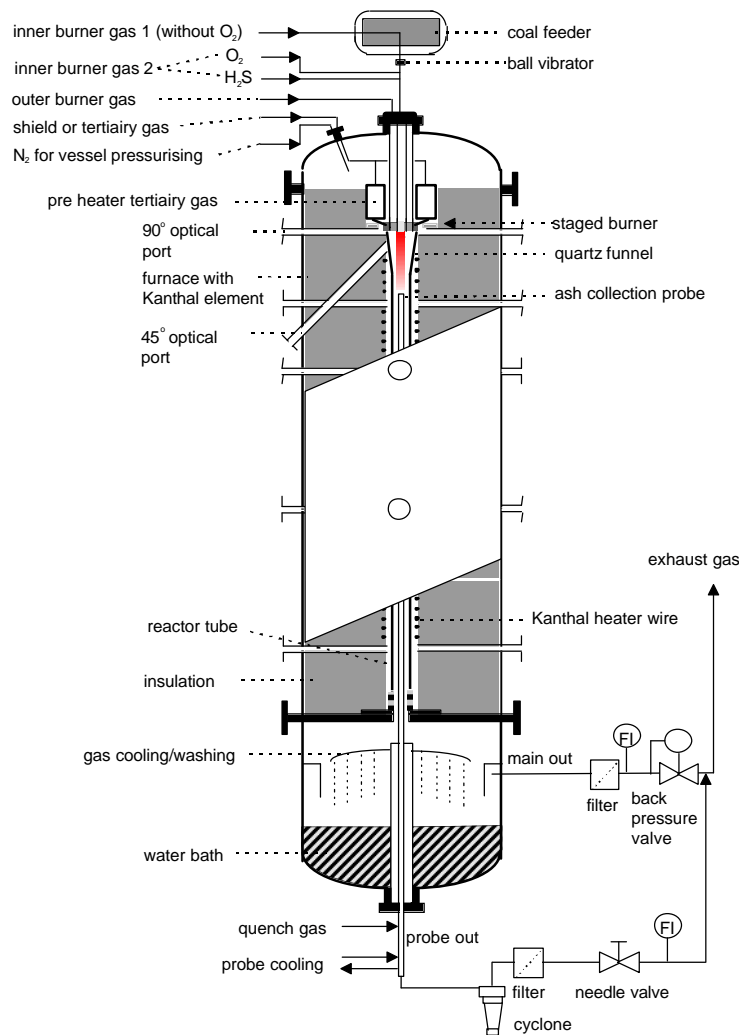


Figure C3 *Schematics of the ECN Pressurised Entrained Flow Gasification simulator (PEFG)*

### Conclusions

Wood ash hardly forms a liquid melt at typical entrained flow conditions. The ash mainly consists of CaO, which has a melting temperature of 1700-1800°C. All clean wood tested show similar results. The addition of Si turns out to lower the melting temperature resulting in a significant fraction of molten ash. In Appendix E thermodynamic modelling results are

presented with different additives and also at higher pressure. Furthermore, some remarks are made on slag viscosity.

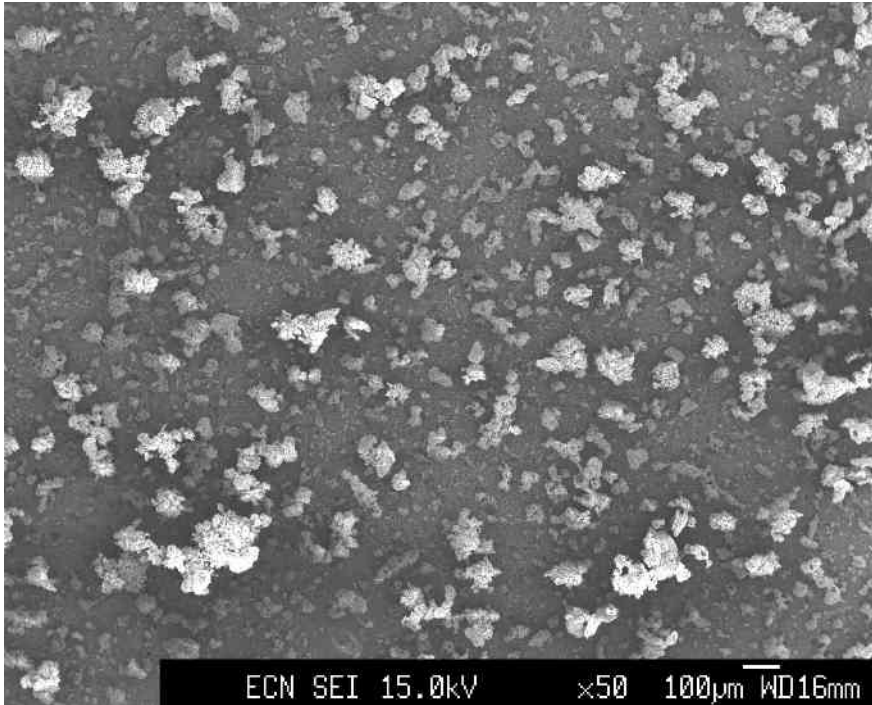


Figure C4 *Micrograph of a deposit plate (top view) after beech gasification in LCS*

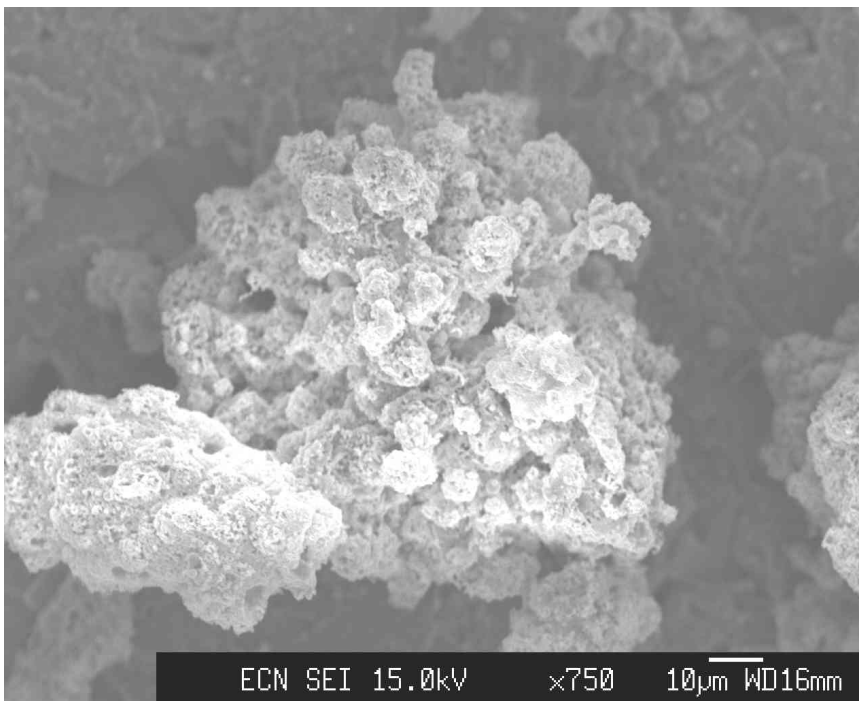


Figure C5 *Micrograph of a beech gasification slag deposit in LCS, enlargement of fraction of Figure C4*

## APPENDIX D CONVERSION AND PRESSURE

Increasing pressure in a gasifier results in two main effects:

- Partial pressures of reactive components like H<sub>2</sub>O and CO<sub>2</sub> get higher. This increases gasification reaction rates.
- Thermodynamically, it will get harder to increase the amount of gas molecules, which is what happens during gasification of solid fuel.

The first effect will result in an *increase* of conversion; the second effect will *decrease* conversion if pressure is increased. There are some additional effects worth mentioning:

- Also oxidation reactions get faster at higher pressure because of increased partial pressure of oxygen. This means that flame temperatures get higher and conversion is *increased* for this reason.
- The morphology and reactivity of the resulting char changes with pressure. It has been shown that the char produced at high pressures has a relatively high degree of large macropores. It also contains a high percentage of cenospheric (small particles of few μm) particles [30], thus resulting in a total decrease of the particle size. In addition, also microporosity, and therefore available surface area, is increased by an increase of total pressure. This positively affects the reactivity of the chars [31]. Mentioned effects of pressure will probably result in an *increase* of conversion. It must be realised however, that most of the knowledge existing on this topic is based on work done on (Australian bituminous) coal and might not be applicable to biomass.

Generally, literature on entrained flow gasification kinetics is limited. Kinetic correlations at high pressure have been generally derived by studies carried out on samples generated at atmospheric pressure or slow heating conditions and therefore may not be completely applicable for pressurised entrained flow gasifiers with very high particle heating rates. In addition, most of the literature available is related to coal gasification. Biomass has not given much attention so far.

In conclusion, it can be said that there are several arguments promoting the statement that increasing pressure will increase conversion. It must be realised however, that (1) thermodynamics are against this and (2) the arguments originate from experience and calculations based on coal.



## APPENDIX E MODELLING OF ASH BEHAVIOUR

This Appendix is a summary of the thermodynamic modelling performed by ECN in order to better understand the experiments and to be able to extrapolate the results to commercial gasifiers. The complete results can be found in [5].

### *The model*

Knowledge on slagging/melting tendencies of the selected fuels has been studied using an equilibrium model (FACTSAGE computer model) minimizing Gibbs free energy and applying it to an hypothetical (pressurised) entrained-flow gasification system. The simulations have been focussed to obtain knowledge on the formation of the slag at typical operating temperature of an entrained flow gasifier and on the distribution of the ash-forming constituents over a wide range of temperatures corresponding to a theoretical temperature profile of an entrained flow gasifier.

The calculations have been performed assuming a fuel to oxygen ratio similar to large-scale commercial entrained flow gasifiers (equivalence ratio ER is 0.25). A sensitivity analysis has been carried out in order to assess the impact of ER in the range 0.1-0.4 on the slag behaviour. The results (not reported here) show that there is little influence on the behaviour of the slag in the high temperature region (1300-2000°C).

### *Simulation of lab-scale test*

Calculations with beech (see Appendix B for composition) indicate that at 1400°C only 15-20% of the ash forming constituents is molten. CaO is the dominant component in the system. This is in good agreement with the observations during the lab-scale tests, see Appendix C. Calculations with wood mixture indicate approximately 50% melt. This matches with the experimentally observed trend. In reality, calculated melt fraction values are not reached. Probably chemical reactions and/or mixing is too slow.

Alkali metals exclusively form gaseous species (calculated for the ranges of 1000-2000°C and 5-30 bar) and will not appear in the slag.

### *Effect of pressure*

Calculations have been performed with pressures up to 30 bar, being the typical pressure for large-scale entrained flow gasifiers. The total slag amount only slightly increases with increasing operating pressure. The only relevant effect seems to be the formation of carbonates at higher pressure. This only affects the melting behaviour at low temperatures (1000-1200°C). It can be of major interest however for the fouling of heat exchange surface upon cooling.

### *Additives*

Some calculations have been performed with silica or clay added to beech wood in order to enhance the melting behaviour. It appears that the addition of 0.5 – 1 kg silica per kg ash results in a minimum melting temperature, being about 1400°C and hardly dependent on pressure. This corresponds to a Si/Ca molar ratio of 1-2. Calculations show that the formation of Ca-silicates directly reduces the melting temperature, but also alkali metals are more easily encapsulated in the matrix. This increases the enhanced melting effect of the addition of silica.

Calculations have been performed with the addition of clay (aluminium-silicate). It is shown that clay can be a good fluxing material for clean wood, may be even better than silica. If clay with assumed Al/Si molar ratio of 1 is added to wood resulting in a molar ratio of Si/Ca of 1, melting temperatures can be reduced to approximately 1200°C.

### *Slag recycle*

One important issue for the design of an (entrained-flow) slagging entrained flow gasifier is the protection of the refractory wall of the reactor. Normally this protective function is achieved by means of the running slag itself, flowing down to the bottom of the gasifier. Entrained flow slagging gasifiers have been so far generally designed for a fuel ash content of minimum 6%. So when designing an entrained flow gasifier for wood with a typical ash content of only 1%, modifications are required. Recycling of slag seems to be the obvious solution. This will require crushing and milling of the slag and subsequent transport and injection into the gasifier. Another (or additional) option can be the recycling of fly ash. This already are small particles and is easy to handle.

Preliminary thermodynamic calculations have been performed with beech and silica. It has been assumed that the slag and fly-ash is recycled with a mass flow, which is 8 times the wood ash mass flow. Manual iterations result in the observation that concentrations hardly change. This can be explained by the fact that only a small fraction of the wood ash leaves the system in the gas phase and also the recycle hardly changes the distribution of elements between gas phase and condensed phase.

Additional calculations need to be performed to evaluate the recycle option with other assumptions. Especially the option with slag recycle and without fly ash recycle (or the other way around) might be interesting. Experimental data on the distribution of elements between slag and fly ash will be necessary.

### *Viscosity*

Entrained flow reactors have criteria on the melting temperature of the fuel (and additives). For a slagging gasifier to operate well, the slag must flow down the walls continuously. Furthermore, the slag must be removed from the gasifier in the liquid phase. For these reasons, viscosity and temperature of critical viscosity are relevant parameters.

It is generally reported that viscosity of the slag must have values typically between 8-15 Pa·s [32,33] to achieve sufficient fluidity to allow free slag flow. A number of empirical correlations relating the viscosity of the slag to its composition and temperature have been reported in literature [32]. These correlations have been extensively developed for coal ash. However, availability of data on slag flow properties in the composition range of wood ash with flux streams is limited. More fundamental work is needed to assess quantitatively the characteristics of the slag flow in terms of viscosity versus slag composition and temperature.

### *Conclusions*

Thermodynamic modelling can effectively be used in understanding and predicting melting behaviour of wood ash (and additives). Model results match fairly good with experimental observations. Adding 0.5-1 kg silica or clay (Al/Si is 1 mol/mol) per kg wood ash greatly enhances melting behaviour (reduce melting temperature). Existing viscosity models developed for coal appear to be not applicable for biomass ash (and additives).

## APPENDIX F FEEDING TESTS BY FUTURE ENERGY

Future Energy GmbH has performed some lab-scale standard feeding tests with pulverised willow wood. It has been pulverised at ECN until all mass passed a 250  $\mu\text{m}$  sieve. The resulting sample has a  $d_{50}$  diameter of 210  $\mu\text{m}$  ( $d_{10}$  is 35  $\mu\text{m}$ ,  $d_{90}$  is 700  $\mu\text{m}$ ). Future Energy concluded that this material is absolutely not suitable to be fed for their fluidised/pneumatic feeding system. Due to the large fraction of fibre-like structures and the corresponding high cohesion forces between particles, it appeared to be impossible to fluidise the material. Channels were formed. With the use of an agitator, fluidisation at 2.5 cm/s was possible. At this relatively high velocity however, the bed already inclined to form bubbles.

Pulverised *torrefied* willow has been subjected to the same “feeding” tests at Future Energy. Although this material has also been milled until all mass passed a 250  $\mu\text{m}$  sieve, similar to the non pre-treated biomass, the  $d_{50}$  particle size is only 90  $\mu\text{m}$  ( $d_{10}$  is 15  $\mu\text{m}$ ,  $d_{90}$  is 340  $\mu\text{m}$ ). Furthermore, the structure is much less fibre-like. The torrefied wood could be fluidised in the test facility at Future Energy. It also appeared however, that the material tend to form “walls” and also slight bubbling occurred. Future Energy expected however that at higher pressure and higher bed height, bubbling may be avoided. The inclination to form walls would mean that effectively only part of the feeding reactor could be used.

In conclusion: it seems difficult, or even maybe impossible, to produce a powder from biomass that can really meet the requirements of a pneumatic feeding system. The particle size distribution and the high cohesion forces between the fibre-like structures of pulverised biomass avoid the proper functioning of existing fluidised/pneumatically coal-based feeding systems. Torrefied and pulverised wood proved to be much better. Although not ideal, it is expected by Future Energy that torrefied wood can be used in the existing pneumatic feeding system.

## APPENDIX G INERT GAS CONSUMPTION IN PRESSURISED FEEDING

In the Table below, calculated inert gas consumption is given for different fuels. Depending on the density of the fuel, 4-6 m<sup>3</sup> gas per ton is needed to pressurize the biomass fuel in lock hoppers and keep the pressure constant. On top of this, 2 m<sup>3</sup> gas per ton is needed to avoid synthesis gas flowing into the feeding equipment (in case of wood chips where screw feeder suffices) or even 6 m<sup>3</sup> gas per ton if the biomass has to be pneumatically transported similar to coal. In the worst case, 11 m<sup>3</sup> inert gas is needed in total.

Compressing 11 m<sup>3</sup> inert gas to 40 bar consumes more than 0.02 kW<sub>e</sub>/kW<sub>th</sub>. Based on primary energy, this is 5%. Because part of the inert gas ends up in the gasifier, also the gasification efficiency is influenced. As shown in the lower line, this can be almost 4% reduction of cold gas efficiency. In case of wood chips, this is only just above 1%.

Due to the large differences in density of biomass and coal, the negative effects of using inert gas driven pressurizing and transporting systems are relatively small.

Table H1 *Calculated values for inert gas use (assumed CO<sub>2</sub>) in pressurised feeding systems for different solid fuels with different feeding systems; no gas re-use from lock hoppers is assumed*

		wood chips <sup>(a)</sup>	wood chips <sup>(a)</sup>	wood powder	torrefied wood powder	torrefied wood powder	coal powder
pressurizing system <sup>(b)</sup>		lock-h.	piston f.	lock-h.	lock-h.	piston f.	lock-h.
feeding system <sup>(c)</sup>		screw	screw	pneum.	pneum.	pneum.	pneum.
density solid fuel particle	kg/m <sup>3</sup>	500		500	475	475	1400
bulk density solid fuel	kg/m <sup>3</sup>	175		250	240	240	700
net CO <sub>2</sub> use lock hopper for pressurizing	m <sup>3</sup> /ton fuel	4.3		2.4	2.6		0.9
CO <sub>2</sub> needed in feed bin to avoid syngas in-flow	m <sup>3</sup> /ton fuel	2.0	0.1	2.0	2.1	0.1	0.7
extra CO <sub>2</sub> to avoid syngas in-flow in feed bin	m <sup>3</sup> /ton fuel	2.0	0.1	-	-		-
CO <sub>2</sub> use pneumatic feeder	m <sup>3</sup> /ton fuel	-		6.0	6.3	6.3	2.1
total CO <sub>2</sub> use	m <sup>3</sup> /ton fuel	8.3	0.2	10.4	11.0	6.4	3.7
CO <sub>2</sub> to gasifier (reducing gasifier efficiency)	m <sup>3</sup> /ton fuel	2.0	0.1	6.0	6.3	6.3	2.1
consequences (calculated for 40 bar):							
electr. consumption for compression inert gas <sup>(d)</sup>	kW <sub>e</sub> /kW <sub>th</sub>	0.021	0.009 <sup>(e)</sup>	0.027	0.025	0.023 <sup>(e)</sup>	0.007
reduction of cold gas efficiency of gasifier	%points	1.2%	0.05%	3.7%	3.9%	3.9%	1.3%

(a) in order to be able to feed the solids by screw feeder, the minimum size is approximately 0.5 mm

(b) lock-h.: lock hopper system; piston f.: piston feeder system

(c) screw: screw feeder; pneum.: pneumatic transport/feeding

(d) expressed per unit lower heating value (LHV) of the solid fuel; LHV of wood chips and wood powder is assumed to be 17 MJ/kg, torrefied wood: 19 MJ/kg, coal: 24 MJ/kg

(e) including electricity consumption needed for the hydraulic piston feeder system (assumed to be 0.008 kW<sub>e</sub>/kW<sub>th</sub>)

## APPENDIX H MODELLING OF BIOSYNGAS OPTIONS

Different biomass-to-syngas options have been modelled using flow sheet simulation package ASPEN-plus in order to calculate efficiencies. The following general assumptions have been made:

- Fuel is clean wood with 35% moisture on wet basis and 1.6% ash on dry basis. The chemical composition on dry and ash-free basis is: 50.8% C, 6.1% H, 42.7% O, 0.3% N, 0.06% S and 0.04% Cl. The higher heating value (HHV) at 298 K and 1 atmosphere is 20.4 MJ/kg dry and ash-free wood. The lower heating value (LHV) of the wood “as received” is 11.4 MJ/kg.
- Wood is dried to 7% moisture prior to every syngas option and stored temporarily at room temperature. Waste heat is used as energy source. This is assumed to be abundantly available and therefore is not included in the overall efficiency. The lower heating value (LHV) at 298 K and 1 atmosphere of the dried wood is 17.3 MJ/kg wet wood.
- In every option, the raw gas is conditioned in a shift reactor and CO<sub>2</sub> is completely removed, the end product is a synthesis gas at 40 bar with a H<sub>2</sub>/CO-ratio of 2.
- CO<sub>2</sub> is available at atmospheric pressure and is used as inert gas (if necessary) for compression and feeding of solid fuel.
- Oxygen is 99% pure.
- Oxygen production electricity consumption is 0.25 kW<sub>e</sub>/m<sub>n</sub><sup>3</sup>, oxygen is available at atmospheric pressure.
- Steam is used as moderator in entrained flow gasifiers with steam/oxygen ratio of 0.1 on mass basis in all options.
- Fuel conversion in entrained flow gasifier is 99%.
- Entrained flow gasification takes place at 1300°C.
- Entrained flow gasification has 1% heat loss.
- Cooling the entrained flow gasifier takes place with a cold syngas quench to 900°C to avoid fouling problems. Further cooling is done in a conventional steam boiler. Steam is used for own use and the rest is used for the production of electricity. Steam of 60 bar is produced.
- Cooling the product gas from fluidised bed takes place in a similar way, a gas quench however is not necessary, since the gas leaves the gasifier at 900°C.
- Fluidised bed gasification takes place at 20 bar, which is considered as maximum commercially available.

The following options have been modelled; the main assumptions are given for every option:

- **Option 0 (EF-100 mm):** Dried wood is pulverized to 100 µm particles with 0.08 kW<sub>e</sub>/kW<sub>th</sub> electricity consumption, pressurized in a lock hopper system and pneumatically transported/injected in an entrained flow gasifier.
- **Option A1 (EF-1 mm):** Dried wood is crushed to 1 mm particles (0.01 kW<sub>e</sub>/kW<sub>th</sub> electricity consumption), pressurized with a lock hopper system and transported/injected by a screw feeder in an entrained flow gasifier.
- **Option A2 (EF-1 mm):** idem option A1, but pressurized in a piston feeder.
- **Option B1 (tor-EF):** Dried wood is first converted into torrefied (brittle) wood with the following composition: 1.5% moisture on wet basis and 1.7% ash on dry basis. The chemical composition on dry and ash-free basis is: 51.7% C, 5.9% H, 42.1% O, 0.3% N. The lower heating value (LHV) at 298 K and 1 atmosphere is 18.6 MJ/kg torrefied wood. The torrefied wood is temporarily stored at room temperature before being pulverized (0.01 kW<sub>e</sub>/kW<sub>th</sub> electricity consumption), pressurized in a lock hopper system and pneumatically transported/injected in an entrained flow gasifier. The gases from the torrefaction are combusted and the energy is used to run the torrefaction. The efficiency from dried wood (7% moisture) to cold torrefied wood is 95%.

- **Option B2 (tor-EF):** idem option B1, but piston feeder is used as pressurizing unit.
- **Option C (pyr-EF):** Dried wood is subjected to flash pyrolysis to produce a pyrolysis oil, char and gas. The oil is mixed with the char resulting in slurry with an energy efficiency of 85%. The slurry has the following composition: 19% moisture on wet basis and 2.3% ash on dry basis. The chemical composition on dry and ash-free basis is: 60.5% C, 6.3% H, 32.8% O, 0.3% N. The lower heating value (LHV) at 298 K and 1 atmosphere is 18.2 MJ/kg oil/char slurry. The slurry is stored at room temperature before being pressurized with pumps and injected into an entrained flow gasifier. The electricity consumption for slurry compression is neglected.
- **Option D1 (FB-EF):** Dried wood is gasified at 900°C at 20 bar in a fluidised bed gasifier with steam/oxygen ratio of 1. Wood chips are pressurized using a lock hopper system and are fed by screw feeder. The carbon conversion in the CFB is assumed to be 93%. The heat loss is 1%. The resulting gas/char is fed into an entrained flow gasifier at the same pressure of 20 bar. After conditioning (shift and CO<sub>2</sub>-removal), the gas is compressed to 40 bar.
- **Option D2 (FB-EF):** idem option D1, but piston feeder is used as pressurizing unit.

The following Table shows the calculated efficiencies of the different options mentioned above. In all cases, the reference is the lower heating value of wood with 35% moisture.

Table I1 *Calculated efficiencies [% of lower heating value of wood fuel] for different options to produce syngas (40 bar, H<sub>2</sub>/CO=2) from wood with 35% moisture, see text for details of the options*

option:	0	A1	A2	B1	B2	C	D1	D2
syngas on specification	76	79	81	73	73	69	77	78
overall efficiency <sup>(a)</sup>	59	80	84	74	75	76	84	85

(a) efficiency from wood to biosyngas and electricity; electricity includes production minus consumption for (if applicable) fuel pulverization, piston feeder, CO<sub>2</sub> compression, syngas compression, O<sub>2</sub> compression, and O<sub>2</sub> production (air separation); electricity is calculated as primary energy assuming 40% efficiency

## APPENDIX I PISTON FEEDER TESTS AT TKE

In order to assess and quantify the positive effects of a piston feeder, 20 kg of torrefied park wood has been sent to TKE for tests. The material has been produced at ECN in their batch reactor at 270°C with a residence time of 30 minutes. The particle size was several cm's.

### *TKE test feeder*

The feeder is a hydraulic driven piston plug feeder. It consists of a 125 mm diameter piston, a 75 mm diameter plug piston, a plug section and a friction control section. The maximum feed pressure is 700 bar. The stroke is 600 mm. The principle of the feeder is shown on the drawing below. Figure J2 shows some pictures.

The principle of this feeding technology is that a sealing plug of the material is formed and this sealing plug is constantly fed into the pressurised chamber. The material is pressed into the sealing section where the friction ensures that a plug is formed. In order to maintain constant friction, a friction control device is fitted. It is important to operate the feeder with a sufficient proportion of the friction on the friction device in order to be able to maintain a constant friction in the inlet of the plug. This adjustment is done by adjusting the length of the plug section and the geometry of the plug section. In order to ensure that the plug will stay stable when the piston is pulled back for filling the plug must have sufficient mechanical strength.

The feeder is equipped with a manual possibility for adjusting the stroke length and a manual adjustment of the friction pressure. On an industrial feeder these 2 parameters would be controlled automatically. The feeder feeds into a pressure vessel with a total volume of 107 liter. With the present hydraulic pump it can produce one stroke every 23 seconds. Tests have been carried out with 1 stroke per minute. The gas leak rate is determined by measuring the drop in pressure over time in the pressure vessel. No compensation for the volume of the biomass fed into the vessel is needed, since this is negligible. The friction force is not measured directly but is expressed by the hydraulic pressure in the control cylinders.

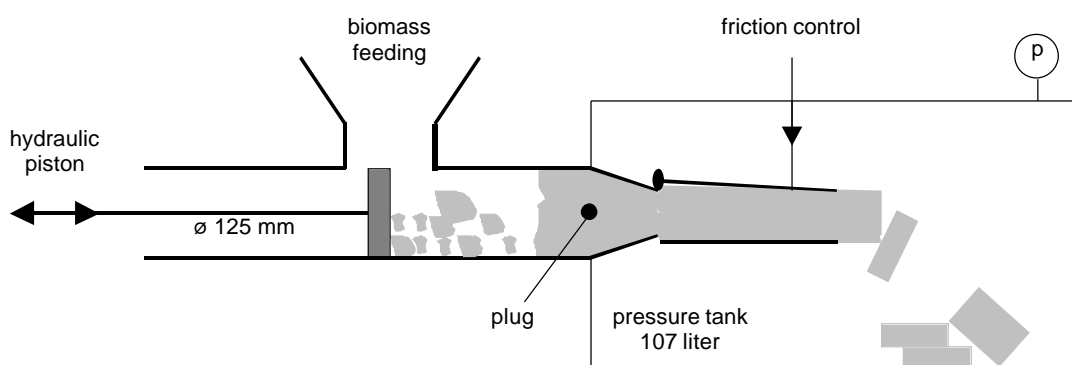


Figure J1 Schematic representation of piston feeder test facility at TKE

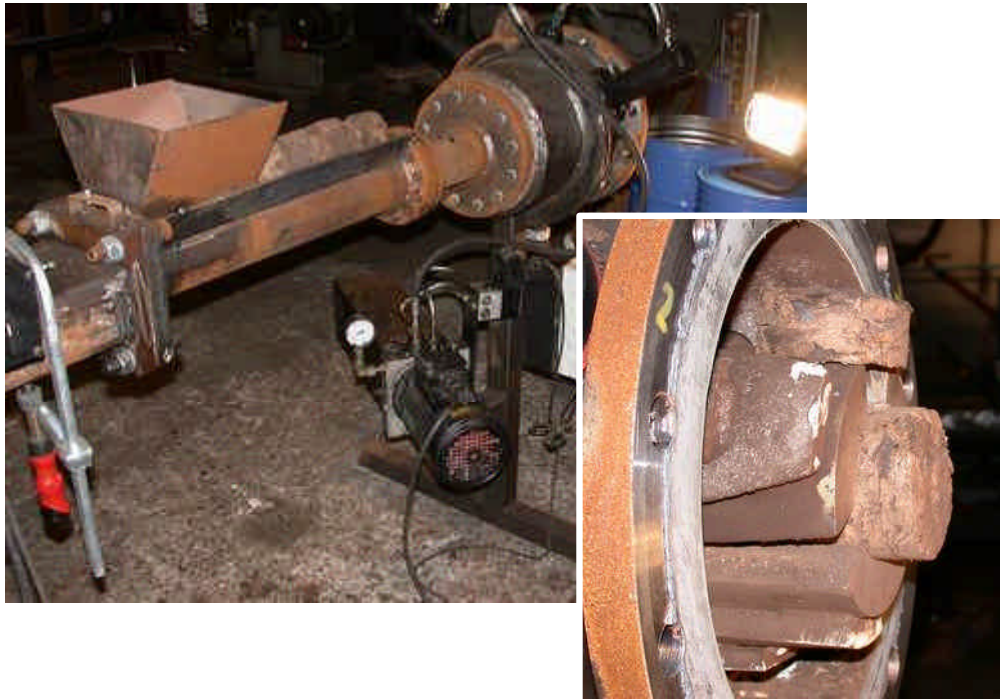


Figure J2 *Piston feeder test facility at TKE (without pressure tank), left: whole system with biomass feeding in front, right: wood plug at outlet of feeder with friction control*

#### *Test results*

Three tests have been done with 0.05, 0.1 and 0.2 kg per stroke. In all cases, the pressure in the pressure tank was set to 20 bar. At 0.1 kg per stroke, the piston pressure increases to 165 bar and reduces to 120 bar upon movement of the plug of 250 mm. The plug density is about  $900 \text{ kg/m}^3$ . The electricity needed to compress amounts to 8 kJ per stroke of 0.1 kg fuel. This is approximately  $0.004 \text{ kW}_e/\text{kW}_{th}$ . The gas leak flow was determined to be  $0.01 \text{ m}_n^3/\text{minute}$ . This means  $100 \text{ m}_n^3/\text{ton}$  biomass. Compared to straw, torrefied wood needs a longer plug to ensure sufficient friction.

#### *Upscaling considerations*

It is expected that plug diameter can be increased to about 300 mm. For straw and bagasse it has been proven that 275 mm is possible. It is assumed that the gas leak flow, expressed as  $\text{m}_n^3/\text{ton}$  fuel, remains constant upon scaling up. The gas leak flow is expected to be  $1\text{-}2 \text{ m}_n^3/\text{ton}$  fuel in a 20 bar system. It is assumed that this increases to  $4 \text{ m}_n^3/\text{ton}$  fuel at 40 bar (this is  $0.1 \text{ m}^3/\text{ton}$ ).

A maximum rate of 1200 strokes per minute can be reached (corresponding to a piston speed of about 0.5 m/s). This corresponds to about 10 ton/hour for a 300 mm diameter piston feeder, or approximately  $50 \text{ MW}_{th}$  lower heating value. The fuel compression will need 220 kW. The complete feeder as shown below will consume about 400 kW, corresponding to  $0.008 \text{ kW}_e/\text{kW}_{th}$ .

In a commercial application, a 3-stage piston feeder will be applied. If operated in such a way that always one piston is sealing, safety can be guaranteed. Figure J3 shows a 3-stage feeder designed for feeding biomass into a pressurized fluidised bed.



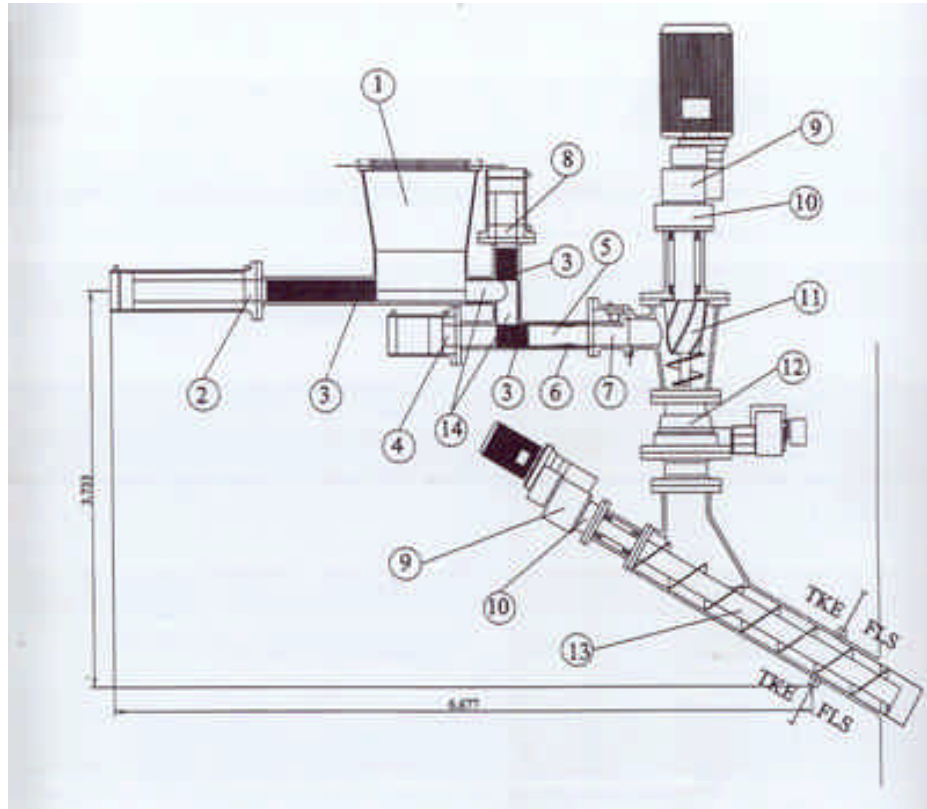


Figure J3 *Three-stage piston feeder of TKE for commercial application, 1: fuel inlet, 2: first piston, 3: replaceable piston front, 4: third piston, 5: primary sealing plug, 6: wear bushing, 7: primary sealing plug control device, 8: second piston, 9: gear, 10: rotary shaft seal, 11: cutter head or disintegrator, 12: safety valve, 13: reactor feed screw, 14: secondary sealing plugs*

## APPENDIX J CONVERSION AND VOLATILE CONTENT

Different fuels have been tested in the LCS at ECN. For description of the LCS, see Appendix C. In order to show the influence of the volatile content on the conversion, some test results are summarised in the following Table and Figure. It can be concluded that a higher volatile content generally results in higher conversion (5th column). Because the overall conversion is partly the result of the fast reaction of volatilisation, the sixth column shows the calculated conversion of the non-volatile fraction of the fuel. Clearly, conversion increases with volatile content. Increased porosity is probably of major importance.

Table K1 *Measured conversion in LCS at 1400-1450°C of fuels with different volatile content, daf: dry and ash-free*

fuel	mean particle size [µm]	residence time [second]	volatiles [wt% daf]	conversion [wt% ash-free]	conversion [wt% ash- and volatile-free]	reference
coal/sewage sludge	ca. 100	0.1	57	68	24	[34]
torrefied beech	90 <sup>a</sup>	0.1	77	88	48	this study
beech	210 <sup>b</sup>	0.1	84	96	75	this study
wood mixture	ca. 200	0.1	85	98	87	this study
wood mixture	ca. 200	0.04	85	96	73	this study
park wood	ca. 150	0.1	85	97	80	[11]

<sup>(a)</sup> fraction between 15 and 340 µm

<sup>(b)</sup> fraction between 35 and 700 µm

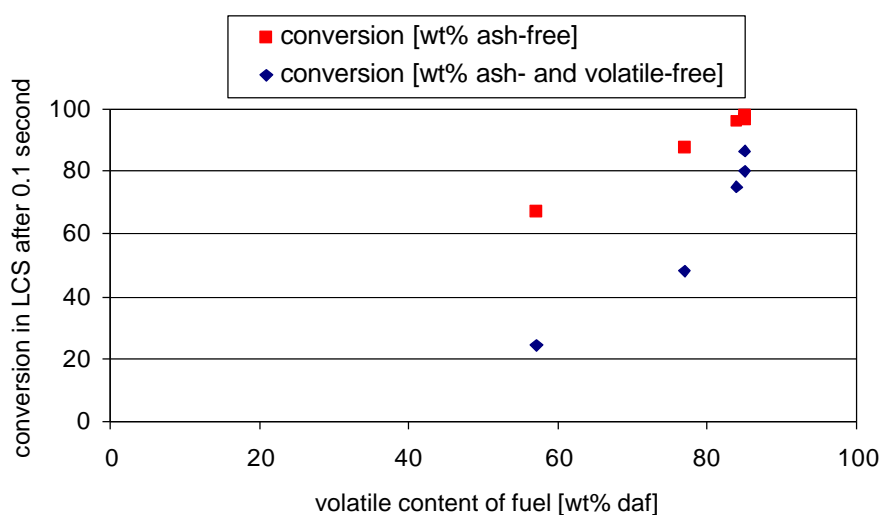


Figure K1 *Measured conversion after 0.1 seconds at 1400°C in the LCS at ECN as function of volatile content of fuel*