

CATALYTIC UPGRADING OF GAS FROM BIOFUELS AND IMPLEMENTATION OF ELECTRICITY PRODUCTION

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Table of Contents

Abstract

1. Objectives of the project
2. Partnership
3. Technical description and results
4. Conclusions
5. Exploitation plans and anticipated benefits

Abstract

A main issue for the development of gasification processes for solid fuels to produce gas for electricity production at small to medium scale is the upgrading of the raw gas to a quality required by engines. A main problem to be solved is to convert the tar formed in the gasifier to useful gas components by an economical, efficient and reliable technique. Older techniques for such systems need separation and periodical cleaning from tar that condenses in ducts between the gasifier and the engine. Other issues that need to be addressed are effects from contaminants both on the performance of the gasifier and upgrading system and on the emissions by the flue gas. Development should preferably aim at technical solutions that will accept a wide range of fuels, with varying contents of chlorine, nitrogen and sulphur.

The results from this project were both the development and implementation of a new technique for tar-decomposition and a large amount of basic information about the behaviour of catalytic materials and their applications that can be used for upgrading of the raw gas by different process concepts.

A complete biomass-to-electricity unit at scale 100 kW_{th} with a new design suitable at small and intermediate scale was thus erected and operated in a test program, using a reversal flow tar converter (RFTC) in a new process concept for decomposition of tar.

Progress was made and large further experience was obtained on how tar contents can be reduced by different catalysts and in different process schemes. Important knowledge and data was revealed concerning the detailed kinetics of tar compounds and about the intrinsic behaviour of dolomite and nickel-based catalysts when used for conversion of tar. New fundamental information on sulphur adsorption on nickel and on effects from chlorine on catalysis by dolomite will be useful in evaluation of process modifications in catalytic hot gas cleaning.

The SPA method for tar analysis and direct mass spectrometry were proven to be very useful tools in the development and testing of gasification processes. The SPA method is currently being discussed to be adapted as an international standard method for tar analysis and it will be included as a candidate for such acceptance in organised work on standardisation, supported by EC and IEA.

1. Objectives of the Project

The overall objective of the project was to broaden the technical and scientific basis which is required for development of electricity and heat producing units for biofuels, in the small to intermediate size range (100 KW_e to 5 MW_e), and to implement a complete biofueled 100 KW_{th} power generating system. The objectives were approached by a combination of basic and applied research and process development.

The objectives of the applied work were:

- to implement systems for gasification of biofuels including gas upgrading to qualities required for electricity production at small scale.
- to implement a complete 100 KW_{th} electricity producing unit which is fuelled with biomass.

The objectives of the basic scientific and technical work were:

- to show the ability of different solid substances to work as catalysts for elimination of tar and nitrogen compounds at atmospheric pressure. The ability for tar elimination at elevated pressure was to be investigated for the most promising catalysts. New catalysts as well as the new application of catalysts developed for other processes will be investigated.
- to obtain kinetic data for known catalysts. Such data are needed for optimal design of catalytic reactors. Nickel catalysts, basic oxides derived from minerals and other promising catalysts are included here.
- to create reactor models for different catalytic reactors, suitable for small scale gasification and based on deep knowledge about reaction kinetics with suitable catalysts.
- to experimentally reveal new information about fundamental chemical and physical processes, such as structural changes, mass transport effects and reaction mechanisms, occurring within particles of at least one promising catalyst. This provides an improved basis for further catalyst development.
- to experimentally reveal new information about poisoning effects by sulphur on nickel catalysts.

2. Partnership

Partner No. 1, TPS Termiska Processer AB was coordinator of the project. The partners and contact persons were:

1. TPS Termiska Processer AB	Mr. B-G Espenäs, Dr. S Frostäng
2. UCM Universidad Complutense Madrid	Prof. J Corella
3. RUG University of Groningen	Dr. G Lammers
4. KTH Kungliga Tekniska Högskolan	Ass. Prof. K Sjöström
5. BTG Biomass Technology Group B.V.	Dr. B van den Beld
6. VTT Technical Research Centre of Finland	Dr. P Simell

3. Technical description and results

Overview of Work Content

The project consisted of four tasks, each consisting of a number of work packages, as listed in Table 1.

Table 1

TASK A	Basic investigations of catalysts and catalysed reactions	Partners
WP-A1	Kinetics for conversion of most refractory tar components, using basic oxide catalysts	1, 3, 6
WP-A2	Interdependence of reaction rates of tar components	1, 3, 6
WP-A3	Reaction mechanism for conversion of most refractory tar components, basic oxide	1, 3, 6
WP-A4	Poisoning effects by chemisorption of sulphur on nickel catalysts	6
WP-A5	Kinetic rate model for conversion of tar using a nickel catalyst	6
WP-A6	Tar characterization	4
WP-A7	Development of new catalysts	1, 6
TASK B	Issues related to different applications of catalysts	
WP-B1	Conversion of tar with dolomite and moderate concentration of chloride	1, 2
WP-B2	Iron-based catalyst for conversion of tar and ammonia	1
WP-B3	Catalyst combinations	2, 4, 5, 6
WP-B4	FCC catalyst	1, 2
WP-B5	Catalyst applications	1, 2, 4, 5, 6
TASK C	Modelling of catalytic reactors and overall kinetics	
WP-C1	Overall kinetics for catalysed tar elimination	1,2,3,4,5,6
WP-C2	Kinetic rate model for ammonia	1, 6
WP-C3	Reactor modelling	1, 2, 3, 5, 6
TASK D	Application of gasifying systems for biofuels	
WP-D1	Gasifying-Gas cleaning units	1, 2, 5
WP-D2	Gas-Electricity unit	5
WP-D3	Economic feasibility	5
WP-D4	Scaling rules	5

Task A. Basic investigations of catalysts and catalysed reactions

WP-A1. Kinetics of catalysed conversion of tar components

This work was done in order to reveal information about the catalytic process of tar conversion with dolomites and lime, which is needed for optimal design of reactors, and for analyses of the performance of such equipment. Knowledge of detailed kinetics is also an important basis for the understanding of the catalytic reaction process.

Detailed kinetics in the conversion of the most refractory tar components was investigated, using basic oxides (dolomite) as catalysts. Reaction rate models, which describe the reaction rate as function of temperature and partial pressures of the main gas components were formulated. Compounds that are typical components in residual tar were used in this investigation. These compounds were benzene, naphthalene, biphenyl, phenol. Different expressions of power law type and Langmuir-Hinshelwood type have been suggested and tested. Power law type expressions have been found to be sufficient at conditions typical for conversion of tar in gas from biomass.

A round test was performed in order to assure the reliability of results on detailed kinetics of tar components. Data on kinetics for conversion of benzene obtained by three partners (TPS, RUG, VTT) were compared. Discrepancies led to discovery of some experimental error, and its causes were corrected. Good agreement was then certified.

A well-designed automated micro reactor set-up, to be used for detailed studies of kinetics of tar compounds was realised by RUG, as part of this work.

WP-A2. Interdependence of reaction rates of tar components

Knowledge about possible interdependence between reaction rates of different tar components is part of the basic information needed to establish a relevant reaction rate model. This was investigated for a dolomite catalyst.

It was shown in work by TPS and VTT that the rate of decomposition of naphthalene is not influenced by the presence of benzene at concentrations up to about 2000 ppm, and in a temperature range of 800-900 °C. Only a minor influence was observed when toluene, ethylene and methane were added simultaneously at concentrations typical for gasified biomass.

WP-A3. Issues related to reaction mechanism of tar conversion catalysed by basic oxides

One issue that was addressed was to explain short-term, reversible changes of activity of dolomite occurring at changes of reaction conditions. It was suggested by TPS that this could be explained by partial coverage of the catalyst surface by a thin (probably monomolecular) layer of a carbonaceous intermediate substance. This hypothesis was verified by dedicated experiments, where naphthalene and benzene were converted by dolomite. Amounts of carbon built up at different contacting times with the hydrocarbon were determined. Good correlation was found between the deactivation

behaviour and time for build up of a steady-state partial coverage of carbon on the surface. It was also found that a larger part of the surface is covered at a low temperature. This effect should contribute to the strong influence from temperature (and large apparent activation energies) found for the overall catalysed reaction of the aromatic compounds.

The effect of higher reaction rate on a “fresh” catalyst surface can be utilised in systems where the catalyst is recirculated at short cycling time, such as a circulating fluidised bed. The magnitude of this effect is about a doubling of the reaction rate (which corresponds to a doubled weight-to-flow ratio).

A second issue was to investigate relationships between surface area, pore structure and reaction rate, in order to find out which factors that determine the catalytic ability of dolomite and lime. Deeper knowledge about these factors will improve the possibilities to find materials of this kind, and possible pretreatment methods giving catalysts with an optimal combination of catalytic and chemical properties. Samples of different calcium-based materials (dolomites and calcined calcium hydroxide) were prepared by different thermal and mechanical treatments at different temperatures. The results are not clear-cut. Results obtained by UCM, where the surface area in macro pores was related to the overall catalytic effect, have indicated that the reaction rate is determined by the area available in macropores ($>500 \text{ \AA}$). Calculations based on porosity data, pore size distribution measured by the BET method and by the mercury intrusion method and accurate data on kinetics of naphthalene did however indicate that the surface area in micropores is almost completely available for the reaction of naphthalene. In such case, differences in catalytic ability (of particles smaller than about 0.5 mm) would be due solely to different activity per unit area, i.e. due to differences in the crystal surface structure. Another explanation that can not be excluded by the data available from this work is restriction to diffusion caused by “bottle-neck pores”.

Reaction-mechanistic studies carried out by VTT were based on comparison of experimental results with benzene and rate equations derived from a number of suggested reaction mechanisms. The best agreement was achieved for a dual-site mechanism, where adsorption to two active sites was assumed to be necessary for C-C bond scission.

It should be mentioned in this context that previous work by TPS, using an isotope technique (deuterium), showed that a significant catalytic effect from dolomite is to abstract hydrogen atoms from all positions in the aromatic ring. This should cause a destabilisation of the aromatic ring, and scission of the ring structure. Some spontaneous scission is obtained already without such destabilisation. Thus, it is very likely that this effect contributes a lot to the reaction mechanism and overall rate of the catalysed decomposition.

WP-A4. Poisoning of nickel catalyst by sulphur

The effect of sulphur on the performance of a nickel catalyst in decomposing hydrocarbons and ammonia in synthetic gasification gas was studied by VTT in atmospheric and pressurised fixed-bed tube reactors. The desorption behaviour of chemisorbed sulphur from the bed materials was monitored by temperature-programmed hydrogenation. A closed-loop gas-recirculation system was used to

measure the isosteric heat of sulphur chemisorption on supported nickel catalyst in hot gas cleaning conditions. The results obtained revealed new information on the behaviour of sulphur in catalytic hot gas cleaning conditions of gasification gas. The new fundamental information of sulphur adsorption at higher temperatures and higher $P_{\text{H}_2\text{S}}/P_{\text{H}_2}$ levels than in previous comparable studies, can be used to evaluate process modifications for improved resistance of sulphur poisoning in catalytic hot gas cleaning.

WP-A5. Kinetic rate model for conversion of tar compounds – Nickel catalyst

Simplified kinetic model equations were developed by VTT. These models are of power-law type and they describe the decomposition of a tar model compound satisfactorily.

WP-A6. Tar characterization

Further development has been done by KTH on the SPA method for tar analysis. This method is convenient to use, since it requires no expensive or complicated equipment for the sampling. Sampling time is short and frequent sampling at 1 sample/min is possible. Components ranging from benzene up to asphaltenes can be analysed. The SPA method was used to a large extent by partners in this project. BTG bought new analysis equipment and introduced SPA method as a regular method in their process analyses.

Tars formed from different fuels (birch, straw, Miscanthus, Salix and reed canary grass) as well samples taken at different conditions and different sampling points were characterised by the SPA. Such data are to be used as input in modelling of the catalytic reactors.

WP-A7. Development of new catalysts

Two different materials based on calcium which have a large surface area compared to other calcium based catalysts (dolomite, lime) were expected to give higher conversion rate. It was suggested by TPS that a low mechanical strength of these materials could be circumvented by inclusion of these materials into a cheap carrier, and that the catalyst produced then might overall be as active as dolomites, but with a largely improved mechanical performance. However, activity tests showed that the catalytic ability of these materials, calculated on the basis of calcium content, did not exceed the ability of the most active dolomites.

A novel catalyst became available for testing by RUG in a late phase of the project. First result is quite promising, giving e.g. better than 99.9 % conversion of benzene at 480 °C and using short residence time (fractions of a second). Kinetic data were determined for in a synthetic fuel gas. It remains to test this catalyst with real tar, and in the presence of typical gas contaminants at long term.

Task B. Assessment of best choice of catalysts for different applications.

WP-B1. Dolomite-Chlorine

The chlorine content of different land-grown biomasses varies over a broad range, up to about 1.5-2% by weight. For example, it is usual that straw has a considerable chlorine content, originating from fertilisers. A large part of the chlorine content is released in gasification, mainly as HCl. The influence from HCl on the catalytic performance of dolomite was studied in this work, both in FB gasification tests performed by UCM and in micro reactor tests performed by TPS.

It was found in the micro reactor tests, using a synthetic gas mixture, that there is no important deactivating effect from the HCl content, as long as this component is present at moderate concentrations. A strong deactivation was established rapidly when the HCl content passed a distinct limit during a stepwise increase of the HCl content. It is a reasonable explanation that this occurs where the equilibrium partial pressure of HCl is passed. It was found however, that the HCl content that was tolerated. It was found that the concentration of HCl, where deactivation occurs was considerably higher than what was expected from equilibrium calculations.

WP-B2. Iron catalyst

Promising results were obtained by TPS in a previous project (AIR2-CT93-1436) using an iron based material as catalyst for conversion of tar. This is a cheap by-product from steel manufacturing, and it has good mechanical strength. In the previous work, its activity for conversion of benzene was found to be orders of magnitudes better than that of dolomite. Results from the experimental work, and from equilibrium calculations indicated that iron carbide was the active catalytic phase, and that this phase possibly could remain in a fuel gas from biomass. Unfortunately, it was found early in the new work that there is a sharp limit within the range of gas compositions of gasified biomass, where the catalyst becomes inactive for conversion of tar.

An expected ability of the iron catalyst to convert NH_3 is still of interest. This was investigated by TPS. The result from that work is described below under WP-C2.

WP-B3. Catalyst combinations

This activity contains screening and assessment of combinations of catalysts.

A large number of catalyst combinations have been tested by UCM in reactors coupled in series, see Table 3 below. Ni catalysts are very active for the conversion of tar. However, coke formation on Ni catalysts can be a problem. It was found that this difficulty can be solved by a first lowering of the tar content, e.g. by use of a low active catalyst before the Ni catalyst bed.

It was suggested by KTH that silica with large surface area could be used to facilitate the conversion of tar by minimising the formation of naphthalene. This was based on experiences from fluidised bed gasification of rice husk, which contains large amounts of silica, and where remarkably low content of naphthalene was obtained. Experiments based on this hypothesis showed that the remaining amount of naphthalene was reduced when first half of a dolomite bed was replaced by the

porous silica. For example, the naphthalene yield was reduced from 1.35 to 0.8 g/kg dry fuel at 850 °C by this procedure.

WP-B4. FCC catalysts

Potential possibilities to use "in-equilibrium" (used) FCC catalysts for tar conversion was tested by UCM in a fluidised bed. The catalytic effect on naphthalene was tested in a micro reactor.

It was shown that the FCC has some positive effect to reduce the tar content, when it is used in the gasifier, and added mixed with the fuel. No significant effect was observed on the conversion of naphthalene in separate experiments performed by TPS.

WP-B5. Catalyst applications

This work package contains an evaluation the suitability of proposed catalysts for different applications, based on the experiences obtained in this project, in the previous AIR project and in other work by the partners

The optimal catalyst is determined by several criteria, such as

- activity for elimination of tar
- activity for elimination of other components, e.g. the NO_x precursor NH₃
- mechanical strength
- resistance to high temperature and thermal shocks
- sensitivity to poisoning (e.g. by sulphur, chlorine)
- lifetime
- costs / economy
- process requirements for separation of lost catalyst

These criteria can lead to different choices, depending on the case of application. Factors such as type of reactor (fixed bed or fluidised bed), pressure, type of fuel and the required gas quality specification determine which is the most suitable catalyst.

Dolomites have up to date been a main choice for catalytic conversion of tar in gasification of biomass. The main advantage of this catalyst is its low cost and non-poisonous properties, which makes it possible to use without dust separation and to drain it together with ash and dust. This is advantageous especially in low pressure processes, where it makes large, costly high temperature filters avoidable. Further, blocking or deteriorating carbon formation on the catalyst is not a problem with dolomite, since the gasification of intermediate carbon formed on catalyst surface is also catalysed.

Dolomites are active for full tar conversion only in their fully calcined form. Typical working temperature is 850-900 °C. A higher temperature is required at pressurised conditions to keep the dolomite in its fully calcined form. For example, a temperature above 920 °C is needed at 10 bar and above 980 °C at 20 bar. Inhibiting

effects from steam and hydrogen on the reaction of aromatic hydrocarbons will also contribute to slow down the rate of conversion at pressurised conditions.

Dolomite is more suited for fluidised bed operation (bubbling or fast FB), rather than fixed bed operation, due to its low mechanical strength. Attrition of dolomite in fluidised bed operation is however of main concern. There are big differences in mechanical strength of dolomites of different origin. Thus, attrition properties are important in quality tests. The rate of reaction of the most stable tar components (e.g. naphthalene) with dolomite is not extremely high at the temperatures that can be achieved in air-blown gasification. Occuring differences in activity between different dolomites are therefore also of importance and should be a part of quality testing. Dolomite is not active for the conversion of ammonia, when the gas contains steam, as in fuel gas from biomass.

Nickel-based catalysts in general are more active than dolomites for the conversion of tar, especially when the gas is free from poisoning components. At “clean” conditions, at least a 200 degrees lower temperature is required for the elimination of naphthalene with a nickel catalyst. The temperature required for nickel catalysts in practical use can be substantially higher. A typical range of working temperature is 700-900 °C with gas produced from biomass. The nickel catalysts are partially deactivated by poisoning from sulphur. This is compensated for, by an increase of temperature. The deactivation by sulphur is increased by elevated pressure. At 20 bars, it is considered that a sufficient tar conversion can be achieved at 900 °C at a space velocity of 2000 h⁻¹. This conclusion has been drawn from tests using different H₂S contents up to 500 ppmv. Nickel catalysts have the advantage of being active for conversion of ammonia to N₂. The mechanical strength of these catalysts make them suited for fixed bed operation. On the other hand, they are not suited for fluidised beds, since attrition will cause unacceptable loss of catalyst. Another suggested application of nickel catalysts is in a monolith structure (which consists of a large number of small parallel, straight channels). Expected advantages of a monolith are better dust tolerance and low pressure drop. It is the opinion of the partner VTT that a monolith reactor might be possible to operate at dust loads up to 10 g/m³_n.

Long lifetime is required for the nickel catalysts, due to the costs for replacement. A lifetime exceeding 3 years has been estimated to be required for nickel monoliths in IGCC operation. Deteriorating effects on nickel catalysts are not sufficiently clear. For example, the effect of chlorine should be investigated. Leakage of nickel by formation of evaporating nickel chloride is a possible problem. Deposition of dust and possible formation and deposition of carbon are effects that need to be tested for nickel monoliths in long term tests. Sulphur lowers the activity for conversion of tar, methane and ammonia. The retarding effect from sulphur is stronger for methane, than it is for tar. This might be taken advantage of, since an almost tar-free fuel gas with a considerable remaining content of methane can be produced. This requires a reactor with a uniform temperature and an optimised residence time.

It has been found by UCM that carbon formation on the nickel catalyst can be avoided if the tar is partially converted in a bed of dolomite before it reaches the nickel catalyst. The tar that could be accepted by nickel catalysts corresponded to a residual content of about 5 g/m³_n. The tendency for carbon formation is probably due not only to the amount of tar, but also to the tar composition. This composition is much dependent on reactions occurring in the gasifier, which are partly non-catalysed and partly catalysed by the char or other materials in the bed. A catalytic

“guard-bed” will of course also cause a change the composition of tar, in addition to the reduction of the amount.

Carbon formation did however not seem to be a problem with the gas containing high tar contents (5-15 g/m³_n), which was processed by the reversal mode of operation used in the RFTC reactor of the BTG gasification system. This was at least the result that was found within the limited time of operation that could be reached within the timeframe of this project.

Other types of catalysts that were tested in this project were assessed to not be suited for the complete conversion of tar in fuel gas, since they had a too low activity for conversion of the most stable components of the tar.

Task C. Modelling of catalytic reactors and overall kinetics

WP-C1. Overall kinetics of catalysed tar elimination

The overall ability to convert tar was determined by TPS, UCM, KTH and BTG for a large number of candidate materials, using fuel gas with real tar produced in laboratory and bench-scale reactors. Experimental work by VTT on the influence from mass transfer limitations was also included. Among these were previously not tested basic oxides (dolomites, limestone), cheap natural materials and commercially available catalysts, such as Ni catalysts, which have been developed for other purposes. In the individual reports from the partners, results on overall kinetics with different catalysts are presented also under the headings of work packages B3, B5, C3 and D1.

WP-C2. Kinetic rate model-NH₃

A large part of the nitrogen content in fuels is converted to NH₃ in gasification. This component should be eliminated from fuel gas since it is a source of NO_x formation when the gas is burnt. Dolomite is not active for conversion of NH₃ to N₂. Thus, it is of interest to find some other catalyst for this purpose. Cheap and non-poisonous catalysts are required for processes where separation of dust from the catalyst is not economically feasible. Process schemes where sophisticated catalysts, e.g. Ni-based, can be used have however been suggested.

A power-law type expression was developed by VTT to describe the conversion of ammonia using a commercially available nickel catalyst.

The iron catalyst described above (WP-B2) was tested by TPS as a candidate for low cost catalysts. A catalytic effect was found and quantified and a simple rate model was formulated. This catalyst might be useful in combination with a tar-converting catalyst in process schemes where a sophisticated catalyst not can be afforded.

WP-C3. Reactor modelling

A reactor model was developed and applied for the scale-up of the RFTC reactor.

Plug flow reactor models for dolomite catalyst were developed and tested by RUG and VTT on the basis of the kinetic models obtained. RUG developed a model for calculation of concentration and temperature profiles at stationary conditions.

A large reactor using operation conditions close to practical reactors was used in the model testing by VTT. Gas partial oxidation was used to heat up this reactor. The reactor model included the effects of mass and heat transfer in addition the chemical reaction.

The detailed kinetic information obtained for single tar compounds, and data obtained by sampling of tar at different positions in catalytic reactors using the SPA method as well as direct mass spectrometry, and experimental procedures with short residence times in the catalytic bed are input to the reactor modelling work.

A model was also developed by UCM for the attrition and elutriation of dolomite in a fluidised bed.

Task D. Application of gasifying systems for biofuels

WP-D1. Gasifying-Gas cleaning units

This is a main work package of the project. It contains the design and construction of new and improved units for the gasification of biofuels and gas upgrading to qualities required for electricity production, and test programs performed both in these equipments.

A pilot-plant was designed and erected by BTG. This system consists of a fluidized-bed gasifier, a reversed flow tar converter (RFTC), a gas cooler, dust filter and a gas engine to demonstrate the technical feasibility of the RFTC under real conditions. A lot of effort was put into the optimisation of the RFTC, since this is a main exploitable result of this project. The control of the RFTC was fully automated. The temperature level in the catalytic part of this system can be controlled automatically by adjusting the secondary air injection. The maximum biomass throughput is 25 kg/hr. Typical tar contents in the producer gas before the tar converter are 5 - 10 g/Nm³. Tar conversions in the RFTC better than 97% have been measured, resulting in tar contents in the cleaned gas of about 50 - 150 mg/Nm³. During start-up the concentrations are somewhat higher, due to lag in time to reach thermal equilibrium and, probably more important, due to time needed to reduce the catalyst (NiO) to Ni.

A large number of different schemes have been tested by UCM in their pilot plants. The capacity of the larger pilot plant of UCM is about 20 kg biomass/h and capacity of the the small system 1 kg/h. Table 2 gives an overview of the different conditions used in tests by UCM.

Table 2

Gasifier beds	Silica sand, Silica sand+Dolomite, Silica sand+FCC catalysts
Feed of solids	Biomass, Biom.+(2-3%) dolomite, Biom.+(5% FCC) catalyst
Chlorine in feed	Biomass with (0.5-3%) chlorine, fed as PVC
Gasifying agents	Air, Steam-Oxygen
Dolomite	In-bed of the Gasifier or in 2 nd Reactor Temperature: 790-835 °C
Commercial Ni catalysts	full size rings, crushed 1-1.5 mm, 7-14 mm Located in 2nd and 3rd bed Temperature: 660-850 °C, mainly at about 800 °C
Catalytic reactors	Full flow: "small", pilot scale
Catalyst used (manufacturers)	BASF, TOPSOE, ICI-Katalco, United Catalysts

The larger system of UCM was built based on experiences from a previous system. Some of the novelties were improved feeder design, increased height and residence time in the freeboard of the gasifier, increased overall height, high efficiency cyclone

after the dolomite FB reactor, improved condensate sampling devices and improved design of the catalytic reactor.

This system contains a full flow catalytic reactor, but also two smaller experimental reactors coupled in series to a side-stream. A large number of catalyst combinations have been tested in these reactors, using silica sand or silica sand mixed with dolomite as bed material in the gasifier. Table 3 gives an overview of the different types of experiments done by UCM on combinations of catalysts. It was found by UCM that optimal performance of dolomite for primary tar conversion is obtained when the bed contains 20-30 % of dolomite. Tar contents achieved by this method in air gasification were about 1 g/Nm³. Very high tar conversion is obtained with nickel catalysts, resulting in residual tar contents down to as low as a few mg/Nm³.

Table 3

Gasifier	Gasifier	1st Gas Cleaning	2nd Gas Cleaning
Gasif. Agent	Bed	catalytic reactor	catalytic reactor
H ₂ O + O ₂	Silica Sand	Dolomite	7 Ni cats
H ₂ O + O ₂	Silica Sand	Silica Sand	1 Ni cat
H ₂ O + O ₂	Silica Sand	DN-34	1 Ni cat
H ₂ O + O ₂	Silica Sand	Dolomite	Silica Sand
H ₂ O + O ₂	Silica Sand	Silica Sand	Dolomite
H ₂ O + O ₂	Silica Sand	1 Ni cat.	1 Ni cat
H ₂ O + O ₂	Silica Sand + Dolomite	Dolomite	1 Ni cat
Air	Silica Sand + Dolomite	Silica Sand	1 Ni cat.
Air	Silica Sand + Dolomite	Dolomite	1 Ni cat.
Air	Silica Sand + Dolomite	1 Ni cat.	1 Ni cat.
Air	Silica Sand + Dolomite	Silica Sand	Silica sand

Some tests in a fluidised bed (capacity 10 kg/h) were also performed by TPS. Dolomite was used as bed material. Quite low tar contents (< 1 g/Nm³) could be obtained at this small scale. These results were in agreement with results from the large number of tests done by UCM. Local concentrations of tar components were measured in these tests by sampling from different positions in the bed. This was achieved by a capillary probe which was directly coupled to a mass spectrometer. It was shown that the conversion of tar is extremely rapid when the primary tars get in contact with the dolomite catalyst immediately after their release from the fuel particles.

WP-D2. Gas-Electricity unit

A VW engine (1200 cc) was connected to the installation at BTG in October 1998. Prior to running on producer gas, this engine was tested fuelled by natural gas. Some modifications were required to enable the use of a gaseous fuel. An AEG electricity generator was coupled to the gas engine.

A number of daily test runs were performed with the complete 'biomass-to-electricity- chain where 'green electricity' was actually produced.

WP-D3. Economic feasibility

An economic analysis of a 2 MW_{el} power plant was given in the previous EC project (AIR-CT93-1436). The investment costs for the RFTC have been updated based on the new information gained in this project. Specific investment costs were estimated for scales of 0.4, 1 and 2 MW_{el}. For the largest scale the specific investment costs are about 165 ECU/kW_{el}. The investment for the RFTC is approximately 10% of the total direct investment cost of a CHP unit. More detailed cost information is currently collected in a demonstration project at a scale of 400 kW_{el}.

WP-D4. Scaling rules

Scaling rules have been formulated for the RFTC and used for preliminary design of larger units up to 1 MW_{el}.

Dimensions concluded for an RFTC in a 1 MW_{el} system are:

Diameter vessel:	1.9 m
Diameter reactor:	1.09 m
Total height:	2.7 m
Height of catalyst:	0.96 m
Height of inert layer:	0.96 m
Amount of catalysts:	0.9 m ³
Amount of inert material:	0.9 m ³

4. Conclusions

A complete biomass-to-electricity unit with a new design suitable for small and intermediate scale was erected and operated in a test program.

Progress was made and large further experience was obtained on how tar contents can be reduced in production of fuel gas at qualities required for electricity production by engines at small and intermediate scale.

The SPA method for tar analysis was proven to be a very useful tool in the development and testing of gasification processes.

Important knowledge and data was revealed concerning the detailed kinetics of tar compounds and about the intrinsic behaviour of dolomite and nickel-based catalysts when used for conversion of tar.

The new fundamental information on sulphur adsorption at high temperatures and high P_{H_2S}/P_{H_2} will be useful in evaluation of process modifications for improved resistance to poisoning by sulphur in catalytic hot gas cleaning.

The results of this project will be used in the development and testing work of gasification processes aiming at electricity production. The results will help in design work of catalytic gas cleaning reactors and they will also be used to explain the test results obtained from larger scale reactors.

5. Exploitation plans and anticipated benefits

Industrial applications

Technologies are being developed for power generation from biofuels by conversion to a fuel gas suited for engines or gas turbines, as routes for utilisation of sustainable energy sources. Techniques that have been drawn to a demonstration phase recently are appropriate for scale larger than about 20 MW_{th}. Different, compact designs with engines, would be more appropriate for smaller scale, giving lower investment cost and better thermal efficiency. Such technologies have not yet been established, due to both economical and technical reasons.

The results achieved in this project will contribute significantly to the progress towards efficient and reliable technologies for small to medium scale processes. The results will also be useful in the further development of systems for larger scale.

The Reversed Flow Tar Converter is developed to work as a stand-alone unit in biomass gasification systems, which can be applied for a large number of different gasification systems and for a wide range of system sizes (see further below).

The results from use of catalytic materials in the gasifier are also encouraging. Two such different lines for further development are possible. One includes catalytic pre-treatment of the raw gas mixture before it enters into a second reactor, where a more sophisticated catalyst is applied. Such process layouts require hot gas filtering which is costly at low pressure. Thus, this would be a solution to (moderately) pressurised processes, which might be applied at medium scale.

The second direction of development includes a cheap catalytic material in the gasifier bed (e.g. dolomite or lime), in combination with other measures needed to ensure sufficient particle separation and low tar content. In spite of the drawbacks and difficulties that can be foreseen or anticipated (such as less efficient tar conversion at large scale, difficulties with certain fuels, loss of catalyst by elutriation and by drain of bottom ash) it is considered that the possibilities to scale up such systems should be investigated further. A successful development in that direction would create an attractive solution for small to medium scale. It would enable a compact design, which is beneficial for efficiency, especially at moderate scale where heat losses are considerable. A compact design should also lead to lower investment costs.

The results from basic research and reactor modelling will be used in the continued development for improvement of gasification techniques. The results on poisoning of

nickel catalysts will be applied in the further development of practical solutions to poisoning problems, such as regeneration schemes and catalysts more tolerant to sulphur. The results on poisoning of dolomite by hydro chloride will be applied in further work to widen the acceptable range of chlorine content in fuels (occurring e.g. agrofuels). The new knowledge about catalytic materials, reaction kinetics, and the reactor models will be used in continued process development for different scales and for different fuels.

Market

Production of electricity from biomass by gasification to a fuel gas for engines is considered to be one of the technologies which can be applied for sustainable energy production. Demonstration of gasification technologies for production of electricity is performed or foreseen in a number of european countries, e.g. in the United Kingdom, the Netherlands, Sweden and Spain, and also in other countries, e.g. within programmes for co-operations between EU-Brazil and EU-China.

Power and heat production at small to medium scale can become a market suited for biomass conversion systems. The supply of biomass feedstocks sets limits the economically feasible size of plants for electricity production, due to the high transportation costs that follows from the low energy density of the fuel. Major obstacles for the commercialisation of these technologies are today the overall economics and lack of proven technology. Tax incentives, which strongly depend on local regulations, are required to achieve economics that are competitive at present market prices of electricity and heat.

The Kyoto objectives, the objectives of EU to reduce greenhouse gas emissions by 8% between the years 2008 and 2012, and the target to double the share of renewables in the EU energy balance from 6% today, to 12% in 2010, are important driving forces that should create a considerable market for technologies for conversion of biofuels to electricity and heat.

Industrial and intellectual property rights

A patent has been applied for by BTG to protect the property rights of the technology for application of the reversed flow technique to conversion of tar.

The results from the basic research are open, and they have already to a large part been disseminated through international journals and conferences. The complete final report is publishable.

Exploitation

Exploitation by the individual partners of the advances made in this project will proceed in a number of different ways. Hence, the industrial partners will progress their respective technologies directly via various consortia, while the academic organisations will offer their enhanced services in support of European industry.

The results of this project will be used both for demonstration of new technique and in further development of gasification processes aiming at electricity and heat production at lowered costs, good reliability and high thermal efficiency. The results will help in design work of catalytic gas cleaning reactors and they will also be used in examination of the performance of experimental and full scale reactors.

Three partners have made the following further specifications on their implementation plans.

TPS

Main part of the results from this project will be used in the further development of gasification processes for application at different scales, to be performed by TPS.

TPS is currently planning a combined R&D and demonstration project for a system where 5 MW_{el} will be produced by an engine. This work will be performed in close co-operation with a European power company. The results from JOR3-CT95-0053 forms an important part of the background information that will be used by that project.

BTG

A demonstration project is of utmost importance for the further development of the system. Recently, a contract has been signed between a number of Dutch parties including a biomass supplier, two construction companies, an electricity distribution company and BTG to perform the pre-engineering of a 400 kW_{el} and a 1 MW_{el} biomass gasification process. The detailed engineering of the system should be ready in the end of March 1999. The intention of the consortium is to demonstrate a biomass gasification system at a scale of 400 kW_{el} in Goor, the Netherlands. This gasification system includes the reverse-flow tar converter, which needs to be scaled up by a factor of 20.

Tentative time table

<i>Period</i>	<i>Activities</i>
1 Jan 1999	End of this project
Dec 98 - March 99	Pre-engineering of 400 kW _{el} and 1 MW _{el} gasification systems
Feb 99 – Jan 00	Catalyst deactivation tests
April 99	Decision on demonstration project (400 kW _{el}) in Goor, the Netherlands

VTT

The results of this project will be mainly used in the development work of catalytic hot gas cleaning reactors. This work has already been started in VTT. The fixed bed reactor model will serve as an example case on which further models, for example for monolith reactors, will be based. The reactor models will be used in reactor design and as well for experimental planning and in the evaluation of the results obtained with larger scale reactors.

The new sulphur adsorption data together with the catalyst activity data obtained will be used in choosing process requirements and nickel catalysts for improved sulphur resistance.

The reactor model and experience gained in the modelling work will be used in the development of the MODEST program package of the company Profmath Oy.