TAR SEPARATION AND CONVERSION USING MICROWAVES (TSC-MW) TO IMPROVE CONVERSION EFFICIENCY INTO ELECTRIC ENERGY FROM PYRO-GASIFICATION

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ABSTRACT: DEVELTAR project was aimed to investigate a device for the removal of tars (cracking) from pyrolysis and gasification gaseous effluents in order to improve their quality as fuel. Tars are condensed and collected by cooling the gaseous effluents than transferred by a wet film of oil to an electricity based technologies with high power and low energy such as microwaves that was extensively examined for this specific purpose. The device developed during the research project was able to use the electrical energy efficiently concentrated towards the disruption of tar molecules, minimizing energy losses through a proper geometrical configuration and setup of process parameters. The main advantage of these technologies is the possibility to concentrate most of the energy on the target molecules avoiding a general increase of the overall energetic level (temperature). The electrical device is not intended for a specific pyrolysis or gasification technology, but as wide-ranging add-on module to be coupled within the gas purification line. The validation at industrial level of the abovementioned technologies, currently known at fundamental research level only, was done at the end of the project.
Keywords: pyrolysis, gasification, tar removal, efficiency, biomass, microwaves.

1 INTRODUCTION

Lignocellulosic biomass can be converted into energy through thermochemical processes such as combustion, pyrolysis and gasification. Nowadays pyrolysis and gasification, and their combination, are of great interest given their high overall conversion rate into energy compared to direct combustion. In this field, tar removal from output gas is still an open issue when energy conversion of such gas through engines or turbines is considered (tar is a general expression for a complex mixture mainly made of heavy hydrocarbons or poly-aromatic hydrocarbons).

Gasification processes is a well proven technology that convert biomass into combustible gasses with an efficiency from 60% to 90%. Gases can be burned to produce heat, to run reciprocating engines for mechanical or electrical power or to make synthetic fuels.

When the fuel gases are used to feed to an engine they must be cleaned and conditioned; tars and particulates concentration may reach 1% in weight and hinder the possibility of power generation from biomass.

The cleanup design strategy for engine feeding are usually based on wet systems that generate remarkable amount of effluents and do not give the possibility to recover heat from producer gas (efficiency loss).

The DEVELTAR project has been implemented by Sea Marconi Technologies S.a.s., a leading company in the field of energy and environment, as project leader. Sea Marconi has been supported by Spike Renewables S.r.l. a partners with relevant competences in the field of interest and a leading engineering and consultancy company in the renewable energy field.

DEVELTAR project (or module) is aimed to investigate a device for the removal of tars, and their subsequent offline cracking, from pyrolysis and gasification gaseous effluents in order to improve their quality as fuel. Tars are condensed and collected by cooling the gaseous effluents, than transferred by a wet film of oil to an electricity based reactor with high power and low energy based on a microwaves system that was extensively examined for this specific purpose.

According to the contact-power theory of gas cleanup (Perry 1973), for a given power consumption, as measured by gas pressure drop or water flow rate, all cleaning devices give substantially the same collection efficiency, and the collection efficiency increases with increasing power. To improve this theory limitation the DEVELTAR module (DM) is intended to reduce power consumption by tar separation and conversion in a small parallel stream. The main foreseen advantage of this technological approach is a relevant improvement in terms of yield of combustible gas, i.e. electrical energy produced downstream, through a low additional energy expenditure, hence an improvement of the net energy balance.

Project goals are addressed to the reduction of contaminants improving the producer gas quality as a fuel and to concentrate energy on the target tar's molecules increasing overall plant efficiency.

The DM is not intended for a specific pyrolysis or gasification technology, but as wide-ranging add-on module to be coupled to the gas cleaning line.

In the following work is reported the results of the DEVELTAR project measurement campaign performed by Sea Marconi Technologies and Spike Renewables with the support of RE-CORD-University of Florence staff. This test campaign is aimed to verify the function of the DM coupled with a typical downdraft gasifier.

2 EXPERIMENTAL SET UP

DM test was carried out coupling the device with the RE-CORD downdraft gasifier; the producer gas coming from the reactor was by-passed from gas cleaning line and directed to DM. The module's performances were assessed by tar sampling and gas analysing at the outlet of tar separation and tar re-gasification.

During these tests the producer gas was burnt in torch after gas sampling and characterization; in fact the aim of the test was to verify the DM performances in real downdraft gasifier operating conditions. Gasifier was operated continuously and at full load for the whole duration of the single tests.

The start-up and transient heating of the gasifier were performed feeding the reactor with charcoal in order to minimize the tar production; after this transition phase it was fed with seasoned, dry and large-particle wood chips (mix of hardwood and softwood). The analysis of used wood chips is reported in Table V.

DM was inserted between the gasification reactor and the gas line. A metallic diaphragm was used to prevent the natural gas circulation and force the producer gas in the DM; from this last-one the tar-free producer gas was re-introduced in the usual gas way through which it reached the torch. A scheme of the above described connection is reported in Figure 1.

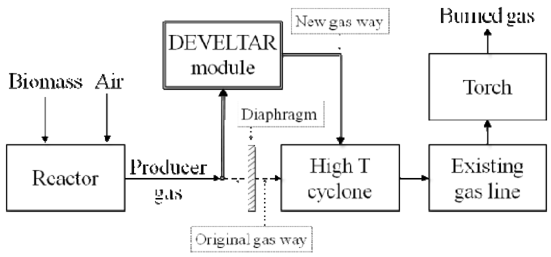

Figure 1: DEVELTAR module connection scheme.

Gas characterization and tar sampling were performed after the DEVELTAR module's condensing unit, in order to verify the removal efficiency and the gas composition. Results of this analysis are reported in section 4.

3 MATERIAL AND METHODS

3.1 RE-CORD gasifier

The utilized gasifier is a downdraft, open top, twin fire reactor characterized by 70 kW of nominal electrical capacity, approximately 70 kg/h of dry biomass inflow and 190 kg/h of producer gas expected outflow.

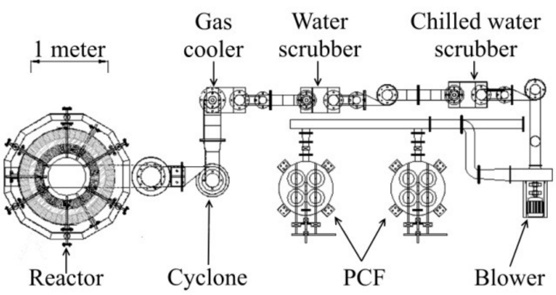

Figure 2: RE-CORD gasifier functional scheme.

3.2 Gas composition

The dry gas composition was measured by a NDIR gas analyzer *ETG Risorse (MCA 100 SYN P)* equipped with an optical sensor for CO, CO_2 and CH_4 measure and two independent sensors for H_2 and O_2 measure. The gas analyser technical specifications are reported in Table I and Table II.

A traps system has been used to connect the gas analyzer to the plant in order to avoid measuring errors because of the methane heavier species presence. Traps were used to remove: (1) particulate and water soluble compound by water bubbling, (2) heavy organics compound by active carbon filter, and (3) humidity by calcium chloride filter.

Table I: Gas analyzer technical specification I

Measure method	Gas Species	Resolution (v/v)	Response time (T10→90)
NDIR	CH_4	1 ppm	<30 s
	CO	10 ppm	<30 s
	CO_2	100 ppm	<30 s
Electro-chemical	O_2	1 ppm	<40 s
	H_2	1 ppm	<30 s

Table II: Gas analyzer technical specification II

Gas Species	Precision	Measuring range	Accuracy
CH_4	3.0% rel.	0-40%	±2% rel.
CO	0.01% abs. or 0.8% rel.	0-10%	±0.02% abs. or ±3% rel.
		10-40%	±5% rel.
CO_2	0.03% abs. or 5% rel.	0-16%	±0.3% abs. or ±3% rel.
		16-50%	±5% rel.
O_2	0.1% abs. or 1.5% rel.	0-25%	±0.1% abs. or ±3% rel.
H_2	3.0% rel.	0-40%	±2% rel.

3.3 Tar sampling

The tar load in the product gas was measured according to a variation of *CEN/TS 15439* procedure, i.e. gas sampling is performed al low temperature (40 °C). In short, after having sampled the gas for 300-600 N litres (dry basis), the gas sampling was stopped and the 2-propanol solutions contained in the six impinger bottles were all mixed in a single Erlenmeyer flask. The 2-propanol solution was then properly treated for both GCMS analysis and gravimetric quantification.

During the sampling, a flow regulator (rotameter) and a volumetric totalizer were used to control the sampled gas flow.

3.4 Laboratory analyses

The tar samples analysis procedure was carried out on the 2-propanol solution and is composed by the following steps:

1. total solution's mass and volume measures;
2. 10 ml sample taking for water content evaluation via Karl-Fisher method;
3. 1 ml sample taking for analysis on light tars via gas-chromatography (as received sample);
4. heavy tars measurement via solvent evaporation (rotavapor) and weighing of the residue (gravimetric tar);
5. gravimetric tar in solvent re-suspension and 1 ml sample taking for qualitative analysis via chromatography (re-suspended sample).

The chromatographic analysis was carried out with GCMS system: GC-2010 (Shimadsu) gas chromatograph with a GCMS-QP2010 Plus (Shimadsu) mass spectrometer and a AOC-20i (Shimadsu) autosampler.

On each sampling solution the GCMS analysis was repeated three times and, when needed, the solutions were properly diluted with fresh solvent. For tar molecule identification and quantification a comparison with the instrument's software library was performed. The gravimetric tar load was instead determined by complete removal of solvent with a rotary evaporator *IKA Labortechnik RV06-LM*.

The water content in the producer gas was measured on a 1 gr sample of the 2-propanol tar sampling solution by Karl Fischer titration system, *848 Titrino Plus (Metrohm)*, according to the ASTM 203-96 standard test method.

4 RESULTS AND DISCUSSION

DEVELTAR module was successfully tested during a 4-day test campaign for over a 24h period without major faults; 1700 kg of biomass were consumed endorsing gasifier full load operation (70kg/h).

The gas analyses after DEVELTAR module are reported on Table III, Table IV and Figure 3:

Table III: Average gas composition during the 3rd day test

Species	Average composition and stability (%vol)		
H2	17.00	±	0.32
CO	15.00	±	0.65
CO2	13.93	±	0.45
O2	0.64	±	0.06
CH4+	1.90	±	0.21
N2 (calc.)	51.53		-

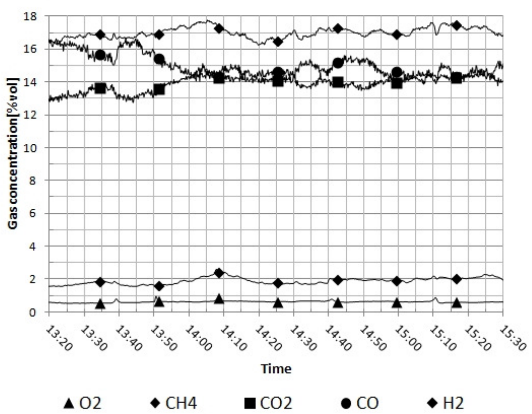

Figure 3: 3rd day test gas composition

Table IV: Average gas composition during the 4th day test

Species	Average composition and stability (%vol)		
H2	15.88	±	0.38
CO	15.94	±	0.54
CO2	13.74	±	0.26
O2	0.30	±	0.03
CH4+	1.19	±	0.08
N2 (calc.)	52.96		-

DEVELTAR module was tested in real operating conditions for 24h with RE-CORD gasifier at full load; producer gas analysis confirms the standard quality expected from a downdraft small scale gasifier.

Tar concentration in cold gas after DEVELTAR module was measured:

- 500 - 534 mg/m3 of GC-detectable compounds (95% benzene)
- 482 – 484 mg/m3 of gravimetric tar (minimal overlap with GC)

Compared to original tar content in producer gas of 1100 mg/m3 the DEVELTAR module efficiency resulted above 55%. The tar concentration after the module is comparable with existing small-scale gasification plants.

The calculated efficiency is highly conservative since benzene (representing 95% of the light fraction) was taken into account, even though the inclusion of benzene in the definition of tars is still controversial and largely depending on the downstream application (ICE, fuel cell, etc.). Moreover also the gravimetric fraction is greatly affected by the presence of an abnormal dust load in the producer gas, due to the absence of a cyclone in the test configuration.

Heat recovery from DEVELTAR module is possible through high-temperature air (avg. ≈150°C); the overall gasification efficiency can therefore be improved by around 7-8%. Through minor system enhancements, efficiency can be further improved up to 10% or more, depending on the output temperature of producer gas.

Collected tars were successfully thermally converted into a gas flow through an offline microwave enhanced partial oxidation with air as oxidizing agent.

Further long-term testing of DEVELTAR module will be performed in conjunction with a proprietary pyro-gasification plant owned by Sea Marconi in Lorraine Region – France.

Table V: Wood chips analysis

Properties		U.M.	Value	Norms
Ultimate analysis				
C	*ad*[b]	*%wt*	48.1	UNI EN 15104
H	*ad*	*%wt*	5.8	UNI EN 15104
N	*ad*	*%wt*	0.1	UNI EN 15104
O[a]	*ad*	*%wt*	39.3	Calculated
Proximate analysis				
volatiles	*ad*	*%wt*	75.2	UNI EN 15148
humidity	*ad*	*%wt*	6.1	UNI EN 14774-3
fixed carbon	*ad*	*%wt*	18.0	Calculated
ash	*ad*	*%wt*	0.8	UNI EN 14775
HHV	*ad*	*$MJ\cdot kg^{-1}$*	18.4	UNI EN 14918
LHV	*dry*	*$MJ\cdot kg^{-1}$*	18.2	Calculated

[a]: by difference
[b]: as determined

5 CONCLUSIONS

DEVELTAR module is not intended for a specific pyrolysis or gasification technology, but as wide-ranging add-on module to be coupled to the gas cleaning line.

Tar separation is a dry gas cleanup that give the opportunity of heat recovery and therefore the possibility to increase efficiency up to 10%.

Micro waves tar re-gasification concentrate most of the energy on the target molecules avoiding a general increase of the overall energetic level (temperature), at the same time preventing the generation of a waste stream to be properly treated or disposed of.

Re-gasified tars may be injected into the main stream of producer gas increasing the LHV and giving a contribution to enhance overall plant efficiency.

6 ACKNOWLEDGEMENTS

The authors of this paper would like to acknowledge RE-CORD-University of Florence staff for their support to this research activities and CCSE (www.ccse.cc) for the financial support.

7 LOGO SPACE