Oxy-steam co-gasification of sewage sludge and woody biomass for bio-methane production: an experimental and numerical approach

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keywords: *sewage sludge, biomass, oxy-steam co-gasification, bio-methane, Aspen Plus*

Abstract. Sewage sludge management is a current problem of extreme concern in Europe. Till now, the most frequent route for sludge management has been incineration or the reuse by shedding on agricultural land both directly or after composting. However, this last method is critically reconsidered lately owing to the increasing contamination of sewage sludge by chemicals, heavy metals, residual organic oils, etc..

In the present study, the performance of a steam-oxy gasification plant has been analyzed. The plant allows the thermo-chemical conversion of sewage sludge, together with woody biomass, into a gas mixture (syngas) composed by basic chemicals (synthetic hydrocarbons), suitable for subsequent industrial production of very high added-value products such as methane.

In particular, a numerical model of methanation plant, composed by a downdraft gasifier, a syngas cleaning system and a Sabatier reactor, has been created within the commercial code Aspen-One and validated with the results obtained from a real scale air gasifier fed with sewage sludge and woody biomass.

First results showed that the steam-oxy co-gasification process has a high energy conversion efficiency that reaches roughly 80% (i.e. produced syngas calorific value vs the dried feedstock calorific value) with an overall methane yield of about 20% (by mass) of the feedstock utilized.

1 Introduction

As reported by Milieu Ltd et al. [1] every year the first 15 state members produce 8,7 million tons of dry sewage sludge, while the additional 12 states members produce 1,2 million tons. These data are increasing as new state members adhere to the directives of the European Community and their relative compliance grows from the initial 50% of collection to the normal rate of the other countries that is over 95%. Thus, sewage sludge management is a current problem of extreme concern in Europe. In particular, the amount of sewage sludge going regularly to landfill must be reduced from 35% of 1995 levels to no significant percentage in the EU-27 by 2020, with an intermediate target of 25% into 2016 (1). As reported by the Italian

Regulatory Authority for Energy, Network and Environment (Det.1/2016-DSID), during 2014 in Italy 25% of the sewage sludge produced, was directly sent to landfill, 38% was destined to direct agricultural use, 46% was used for compost preparation and 6% was incinerated. The remaining 10% had an unknown and, probably illegal, destination [2].

These figures confirm that the most frequent route for sludge management in Italy as in Europe is the reuse by shedding on agricultural land both directly or after composting. This method is regulated by the elderly Sewage Sludge Directive (SSD) (EC Directive, 1986), originally developed to discourage uncontrolled spreading of pollution on agricultural land so promoting a sustainable use of sewage sludge on land [3]. At present the contamination of sewage sludge by

chemicals, medicines, heavy metals, residual organic oils, etc. is continuously growing at a rate even larger than that of their total amount.

The target of lowering pollutants limits in agriculture reuse of sewage sludge has led the European Commission (EC) to critically reconsider SSD in the last years, presumably resulting in the prescription of more stringent limits, and consequently growing costs, in the next years (EC, 2010), in particular for Cd, Hg, Pb and Cr. The complete ban of untreated sludge is expected as well.

According to the described scenario, the present research activity aims at using an innovative oxy-gasification technology to perform the thermo-chemical conversion of sewage sludge together with woody biomass into a gas mixture (called syngas) composed by basic chemicals (artificial hydrocarbons), suitable for subsequent industrial production of very high added-value products such as methanol and methane. Even though gasification is a relatively mature technology, traditional reactors can produce particle and ash free syngas at working temperatures limited to a range between 350 °C and 500 °C. On the contrary, in the proposed reactor, which operates at very high temperatures (up to 1000 °C), the conversion of char can reach high values, such as 90-95%, leaving mercury and other heavy metals and minerals of sewage sludge almost completely in the exhaust ash.

2 Oxy-steam co-gasification approach

Air co-gasification of biomass with organic and waste matrices has been already tested [4-6], at least at experimental level, confirming that it can achieve higher conversion efficiencies and lower pollution when compared to other methods, such as standard combustion (incineration).

In the present research activity, the conventional air co-gasification is replaced by oxy-steam co-gasification technology. This approach allows to obtain a nitrogen-free synthesis gas (syngas), mainly composed by elemental chemical compounds (CO, H₂, CO₂, CH₄ and H₂O), which can be used into the synthesis of various high valuable chemicals like synthetic natural gas (SNG), hydrogen, methanol, hydrocarbons (via Fischer-Tropsch reaction) [7-9]. Steam is utilized in combination with oxygen to mitigate its reactivity which can leads to excessive reaction temperatures.

The proposed sewage sludge treatment through oxy-steam co-gasification with woody biomass represents a new approach with much better results in terms of energy conversion and emissions than conventional sludge incineration. This treatment can be considered competitive even against composting, since this last can be utilized as a soil amendment material but requires particular attention to maintain the concentrations of toxic chemicals in the sludge to acceptable levels. The application of a co-gasification process will minimize the contaminants and only clean by-products (clean condensates and biochar) will result from the gasification [10-14].

Another environmental benefit introduced by the

proposed solution is related to the energetic aspect of the entire oxy-steam co-gasification process. In fact, the energy yield from sludge, through methane gas production during anaerobic digestion or through incineration of dried sludge, is often insufficient to evaporate sludge water content or to power blowers, pumps, or centrifuges required for dewatering. The steam-oxy co-gasification process has a high energy conversion efficiency that can reaches roughly 80% (i.e. 80% of the dried feedstock calorific value can be found in the produced syngas calorific value) and the drying process for sewage sludge and woody biomass (with a moisture content varying from 35% to 45%) does not represent a problem since it requires a maximum of 40% of the produced syngas, hence without compromise the overall convenience of the co-gasification approach.

Last but not least, the oxy-steam co-gasification technology can lead to the installation of small plants widespread on the territory, minimizing the environmental impact and the cost of transportation [15].

3 Description of the experimental gasification plant

The present research activity can be divided into two parts. The first one was dedicated to analyse the behaviour of a real-scale gasification plant where a blend of sewage sludge and woody biomass was co-gasified with air. Data collected during the aforementioned activity were utilised to validate a numerical model, built in Aspen Plus environment, utilised to simulate a bio-methane production plant basically composed by an oxy-steam gasifier and a Sabatier reactor.

The experimental activity has been conducted on a gasification plant equipped with a downdraft gasifier (Imbert type) and a syngas cleaning system. The gasifier has a gross thermal power of about 800 kW_{th} at maximum load, corresponding to a woodchips consumption of roughly 300 kg/h with a moisture content of 10 wt.%. Air was used as a gasification agent and dried woodchips (with moisture content < 15 wt.%) as woody biomass. In the present experimental activity, the syngas cleaning system was not utilized and the produced syngas was burned in a torch after being sampled. Ashes are collected at the bottom of the gasifier and discharged in a sealed tank. Figure 1 shows an image of the gasification plant.

It is worth emphasizing that the gasifier (properly instrumented with air/syngas flow meters, pressure transducers and thermocouples) is equipped with a sophisticated internal gasification air distribution system, named “double-fire” and covered by a patent, which avoids channelling, leads to low Tar production and allows an almost complete oxidation/reduction of the fuel mixture (low char production). Figure 2 shows a sketch of the gasifier with the double-fire air distribution system.

The syngas composition was online monitored by means of an Agilent 3000 micro-gas chromatograph (GC), equipped with two columns (a Molsieve 5A and a PLOT U column) and thermal conductivity detectors,

able to measure the content of hydrogen, oxygen, nitrogen, carbon monoxide, carbon dioxide, methane, ethane, ethylene and acetylene in the product gas. The Lower Heating Value (LHV) of the syngas was calculated from the measured syngas composition.

Sampling and analysis of tar and particles in product gases from the gasifier were performed according to the Technical Specification CEN/TS 15439:2006 [14]. The gravimetric method was employed for the quantitative determination of tar.

Fig. 1. Experimental gasifier.

Fig. 2. Simplified sketch of the gasifier with the “double-fire” air distribution system.

4 Feedstock characterization and experimental results

The characterization of feedstocks used in co-gasification experiments was performed following the methodologies briefly described herein. The moisture content was evaluated according to the European standard test method EN 14774-1. Volatile Matter (VM), Fixed Carbon (FC) and Ash content were determined by thermogravimetric (TG) analysis. A TA Instruments Q-500 thermobalance was employed for TG analysis. Ultimate analysis was carried out with a LECO TruSpec CHN Elemental Analyzer, according to EN 15104 test method. The chlorine and sulphur contents were determined by X-ray fluorescence analysis, according to EN 15309 test method. The oxygen content was evaluated by difference. A LECO AC-500 Calorimeter was used for heating value determination, according to EN 14918 test method.

The results of material characterization are reported in Table 1. As evidenced by the data, the sewage sludge utilised in the present investigation has a very high ash content (69.4 weight % on a dry basis), resulting in a particularly low energy content (4.34 MJ/kg on a dry basis) which avoid its utilisation alone. Furthermore, both the composition and the amount of ash in the sewage sludge feed may contribute to potential ash agglomeration in the reduction zone of the gasifier leading to reactor blockage [16].

The co-gasification of woody biomass and sewage sludge was experimentally investigated in the full scale downdraft gasifier described above. In order to compare gasification performance, gasification tests with pure woodchips were also carried out. Tests were performed operating the gasifier at a reduced load for safety reasons, and a further 10 % reduction in the blower capacity was used in co-gasification experiments.

Preliminary tests were performed to determine the maximum percentage (by mass) of sewage into wood chips which allows a reliable gasification process. This amount was found to be roughly 30%. Properties of the mixtures used in co-gasification tests are shown in Table 2, while the mean values of the main gasifier operating parameters in gasification and co-gasification tests are reported in Table 3, where the equivalence ratio is the ratio between the actual air supplied to the stoichiometric air required for complete fuel combustion. Values ranging from 0.30 to 0.38 were achieved for the equivalence ratio.

The pressure drops in the reactor, the temperatures inside the gasifier and in the syngas cleaning line, the gas flow-rate, and the syngas composition were monitored throughout the tests. Operating problems were not observed, no difficulty in the feeding system, and no obstruction in the gasifier occurred during the entire duration of the experimental campaign, even in co-gasification tests performed with out-of-specification feedstocks. Quite stable trends were observed for all the measured parameters in each test, and mean values could be calculated for stationary conditions.

Due to the reduced load, lower values for syngas flow rate were observed during co-gasification tests

Tab. 1. Proximate analysis, ultimate analysis and heating value of the feedstocks used in the present investigation (VM: Volatile Matter; FC: Fixed Carbon; LHV: Lower Heating Value; ar: as received; daf: dry and ash free).

Feedstock	Proximate Analysis				Ultimate Analysis						LHV
	Moisture (wt %, ar)	VM (wt %, dry)	FC (wt %, dry)	Ash (wt %, dry)	C (wt %, daf)	H (wt %, daf)	N (wt %, daf)	O (wt %, daf)	Cl (wt %, daf)	S (wt %, daf)	LHV (MJ/kg, dry)
W	13.2	83.8	15.4	0.8	50.28	6.14	0.17	43.41	<0.01	<0.01	19.21
SS	21.2	28.0	2.6	69.4	46.47	6.35	3.53	42.12	0.70	0.83	4.34

Tab. 2. Properties of the mixtures used in co-gasification tests (LHV: Lower Heating Value; ar: as received).

Mixture	Moisture (wt %, ar)	Ash (wt %, dry)	C (wt %, dry)	H (wt %, dry)	N (wt %, dry)	O (wt %, dry)	Cl (wt %, dry)	S (wt %, dry)	LHV (MJ/kg, dry)
W – SS	15.6	20.0	39.89	4.93	0.42	34.61	0.06	0.07	15.05

(403-415 Nm³/h) with respect to tests performed with pure woody biomass (493 Nm³/h). Temperatures ranging from 800 °C to 900 °C were measured in the reduction section of the gasifier utilising wood-chips, while for woodchips – sewage sludge co-gasification a mean reaction temperature of 642 °C was recorded. The syngas temperature at the gasifier outlet ranged from 570 °C to 630 °C.

Tab. 3. Operating conditions, experimental results and performance indexes for the gasification of woodchips (W) alone and co-gasification of 30% (by mass) of sewage sludge (SS).

Feedstock	W	W – SS
Fuel feeding rate (kg _{ar} /h)	212	226
Air flow rate (Nm ³ /h)	287	250
Equivalence ratio	0.335	0.351
Gasification temperature (°C)	847	642
Syngas composition (dry)		
H ₂ (vol %)	15.1	16.4
CO (vol %)	23.8	20.6
CO ₂ (vol %)	9.39	11.3
CH ₄ (vol %)	2.28	1.83
C ₂ H ₆ (vol %)	0.05	0.02
C ₂ H ₄ (vol %)	0.58	0.23
C ₂ H ₂ (vol %)	0.06	0.01
N ₂ (vol %)	48.7	49.6
Syngas LHV (MJ/Nm ³) (dry)	5.86	5.18
Syngas flow rate (Nm ³ /h)	493	415
Gas Yield (Nm ³ /kg fuel)	2.32	1.84
Cold Gas Efficiency (%)	76.9	71.9

5 Simulation model of the oxy-steam gasification

5.1 Construction and validation of the model with air as gasifying agent

Using the experimental data reported above, a specific simulation model of the existing gasifier using Aspen Plus has been developed and validated. The geometrical features of the reactor and the real data of the plant equipment such as fan and wet scrubber, have been used to simulate in a reliable way the operative behaviour of the gasification plant.

With particular reference to the existing gasifier, the phases of drying, pyrolysis, combustion, gasification and post combustion within the different zones of the reactor are separately simulated with several blocks (Figure 3). The drying and pyrolysis are carried out using the thermal power from the combustion zone at the first inlet of the gasifying agent. They were simulated using the RYield and SEP blocks of Aspen in order to separate the volatile matter from the solid phase composed by char and ash.

The combustion and gasification processes are detailed simulated using 18 RCSTR blocks where the volatile and solid phases are contemporarily present. For each block, which rigorously models the configuration of continuous stirred tank reactor, the actual size of the part of the gasifier, in terms of volume and phases, is inserted as input. The streams of syngas outlet, post-combustion air and residual char are connected considering the real geometry of the reactor.

The gasification process has been simulated using a pseudo kinetic approach in accordance with the chemical reactions reported in Table 4 [17]. The tars produced from the pyrolysis process have been considered composed only by benzene (C₆H₆). The right fitting of the experimental data has been obtained tuning the coefficients of the reactions as reported in the following. The amount of the gasifying agent that is supplied using the second inlet of the post combustion has been estimated using the experimental data, too. The

overall goodness of the simulation model is demonstrated by Fig. 4, where its results are compared with the experimental ones, both for wood and the mixture between wood and sewage sludge using pure air as gasifying agent.

Tab. 4. Main gasification reactions.

<i>Heterogeneous reactions</i>		
R_1	$C_{(s)} + (\alpha+2)/(2\alpha+2) O_{2(v)} \rightarrow \alpha/(\alpha+1) CO_{(v)} + 1/(\alpha+1) CO_2 + (110.53 \alpha + 393.5)/(\alpha+1) \text{ kJ/mol}$	Char Combustion
R_2	$C_{(s)} + CO_{2(g)} \rightarrow 2 CO_{(g)} - 172 \text{ kJ/mol}$	Boudouard
R_3	$C_{(s)} + H_2O_{(v)} \rightarrow CO_{(g)} + H_{2(g)} - 131 \text{ kJ/mol}$	Water-gas
R_4	$C_{(s)} + 2 H_{2(g)} \rightarrow CH_{4(g)} + 75 \text{ kJ/mol}$	Methanation
<i>Homogeneous reactions</i>		
R_5	$CO_{(g)} + 0.5 O_{2(g)} \rightarrow CO_{2(g)} + 283 \text{ kJ/mol}$	CO combustion
R_6	$H_{2(g)} + 0.5 O_{2(g)} \rightarrow H_2O_{(g)} + 242 \text{ kJ/mol}$	H ₂ combustion
R_7	$CO_{(g)} + H_2O_{(v)} \leftrightarrow CO_{2(g)} + H_{2(g)} + 41 \text{ kJ/mol}$	Water gas shift
R_8	$CH_{4(g)} + H_2O_{(v)} \leftrightarrow CO_{(g)} + 3 H_{2(g)} - 206 \text{ kJ/mol}$	Steam- methane reforming
R_9	$CH_{4(g)} + 1.5 O_{2(g)} \rightarrow CO_{(g)} + 2 H_2O_{(v)} + 517 \text{ kJ/mol}$	Methane partial- oxidation
R_{10}	$C_6H_6 + 7.5 O_2 \rightarrow 6 CO_2 + 3 H_2O + 3169 \text{ kJ/mol}$	Oxidation of benzene

Heterogeneous reactions

$$R_1) \alpha = [CO]/[CO_2] = 2500 \exp(-6249/T)$$

$$r_1 = p_{O_2} [1/k_{film,O_2} + 1/k_{ash,O_2} + 1/(Y^2 k_{r,O_2})]^{-1} 6(1 - \epsilon)/d_{p,0} \text{ (kmol/s m}^3 \text{ bed)}$$

$$k_{ash,O_2} = k_{film,O_2} \epsilon_p^{2.5} Y/(1-Y)$$

$$Y = d_{core} / d_{p,0}$$

$$k_{film,O_2} = 1.24 (T/1800)^{1.75} / (d_{p,0} T)$$

$$R_2) r_2 = 2790 \cdot 10^6 \exp(-22650/T) (p_{CO_2} - k_{r,CO_2}) n_c / V_{react} \text{ (kmol/s m}^3 \text{ bed)}$$

$$k_{r,CO_2} = p_{CO_2}^2 / \exp(20.92 - 20280/T)$$

$$R_3) r_3 = 558 \exp(-22650/T) (p_{H_2O} - k_{r,H_2O}) n_c / V_{react}$$

$$k_{r,H_2O} = p_{H_2} p_{CO} / \exp(17.29 - 16330/T)$$

$$R_4) r_4 = 0.00175 \exp(-7.087 - 8078/T) (p_{H_2} - k_{r,H_2}) n_c / V_{react} \text{ (kmol/s m}^3 \text{ bed)}$$

$$k_{r,H_2} = (p_{CH_4} / \exp(-13.43 + 10100/T))^{0.5} \text{ (m/s)}$$

where:

$k_{film,i}$ = mass transfer coefficient of the i-th component through the gaseous film (m/s)

p_i = partial pressure of i-th component (atm)

d_{core} and $d_{p,0}$ are the diameter of the unreacted core and of the fed chip, respectively (m)

T = temperature (K)

ϵ = bed porosity (-)

ϵ_p = ash layer porosity, supposed equal to 0.75

n_c = kmol C / s

V_{react} = volume of the reactor (m³)

Homogeneous reactions:

$$R_5) r_5 = \epsilon \cdot 1.17 \cdot 10^{10} \exp(-15105/T) C_{CO} C_{O_2} C_{H_2O}^{0.5} \text{ (kmol/ m}^3 \text{ bed s)}$$

$$R_6) r_6 = \epsilon \cdot 8.83 \cdot 10^{-2} \exp(-12005/T) C_{H_2} C_{O_2} \text{ (kmol/ m}^3 \text{ bed s)}$$

$$R_7) r_7 = \epsilon \cdot 4.17 \cdot 10^3 \cdot \exp(-1513/T) (C_{CO} C_{H_2O} - C_{CO_2} C_{H_2} \exp(-3966/T) / 0.0265) \text{ (kmol/ m}^3 \text{ bed s)}$$

$$R_8) r_8 = \epsilon \cdot 15 \cdot 10^8 \exp(-15.03/T) C_{CH_4} C_{H_2O} \text{ (kmol/ m}^3 \text{ bed s)}$$

$$R_9) r_9 = \epsilon \cdot 9.2 \cdot 10^6 \cdot \exp(-9650/T) T \cdot C_{CH_4} C_{O_2} \text{ (kmol/ m}^3 \text{ bed s)}$$

$$R_{10}) r_{10} = \epsilon \cdot 1891.04 \cdot \exp(-12200/T) \cdot T \cdot C_{tar}^{0.5} C_{O_2} \text{ (kmol/ m}^3 \text{ bed s)}$$

where C_i is the molar concentration of the i-th gaseous component in kmol/m³.

Other hypotheses of the simulation model are reported below:

- each block representing a specific part of the gasifier is considered adiabatic and exchanges thermal power only with the adjacent block thanks to the heat conduction, gas and char flow;
- Redlich Kwong Soave is used as Equation of State (EoS) for the simulation of the gas phase;
- the inlet temperature of the gasifying air is 15°C without any preheating.

Fig. 3. Schematic simulation model of the gasifier.

Fig. 4. Comparison between experimental and modelling results of the air gasification of wood and wood+sewage sludge.

5.2 Simulation of the oxy-steam gasification process

Using the validated simulation model, the performances of the oxy-steam gasification with the existing downdraft reactor has been simulated when the mixture wood and sewage sludge is used. The purpose of the analysis is to analyse both the gasifier behaviour when the gasifying agent switches from air to a mixture of pure oxygen and high temperature steam. The same chemical framework described above in Tab.4 is used. The steam for the gasification is supposed to be supplied at 220°C and 13 bar-a and it is mixed with pure oxygen (which is available at 15°C) just upstream the reactor and injected into it at atmospheric pressure. Steam is obtained utilising the heat produced during the methanation process, as better described hereafter.

Figure 5 reports the molar composition of the syngas in function of the equivalent ratio (ER), that is calculated as the ratio between the actual quantity of oxidant and the stoichiometric amount that is required for the complete oxidation, and the mass ratio between the steam mass used in the oxy-steam gasification and the gasified fuel. The hydrogen content is only slightly affected by the operative conditions. Increasing the amount of oxygen, the char conversion increases and, consequently, the concentration of CO and CO₂ is improved, such as the total flow of the syngas.

Fig. 5. Syngas composition (dry basis) with Aspen plus of the oxy-steam gasification of the mixture wood+sewage sludge.

5.3 Simulation of the methane production process

The syngas produced by the oxy-steam gasification process feeds the methanation plant (Fig. 6), which assures the production of a pure methane stream useful for its effective exploitation. The plant is pressurized by a centrifugal compressor at 10.5 bar-a. After that, the production of methane is obtained using five catalytic adiabatic reactors. In the first the CO shift reaction occurs in order to remove the CO and produce H₂ and CO₂, that are the reactants of the following Sabatier reaction:

Then, four catalytic reactors assure the Sabatier reaction:

In order to remove the CO₂ that does not react in the Sabatier reactor, a CO₂ scrubber using KOH solution is present just upstream the last Sabatier reactor. High temperature steam is used to promote the reaction conversion of the water-shift reactor toward H₂ and to mitigate the gas temperature at the outlet of the first Sabatier reactor. The overall process is exothermic assuring that the overall thermal power which can be recovered from the gas is enough to produce the required injected steam from fresh water.

The five reactors have been simulated supposing that the equilibrium is not fully reached and a temperature approach to the equilibrium thermodynamic condition is fixed.

Fig. 6. Layout of the methanation plant.

The performances of the methane production plant are summarized in Fig. 7 and 8. The specific productivity of methane is maximize increasing the O₂ flow-rate and with low steam flow reaching 20 kg per 100 kg of gasified fuel. A critical aspect, that must be carefully managed during the operation of the gasifier,

is the presence of oxygen in the produced syngas, because the oxygen promotes the oxidation of H_2 and CH_4 within the methanation plant with a strong lowering of the methane production.

Another important operative aspect of the methane production is the necessity to use steam in the water shift reactor and in the CO_2 scrubber. The net water consumption of the system (Fig. 8) can reach values higher than 1.5 kg per each kg of gasified fuel. So, higher methane production corresponds to higher water consumption.

Fig. 7. Specific methane production with Aspen Plus using the mixture wood+sewage sludge.

Fig. 8. Specific water consumption with Aspen Plus using the mixture wood+sewage sludge.

6 Conclusions

In this paper the operative feasibility of the methane production from a mixture of wood and sewage sludge using oxy-steam gasification and methanation with Sabatier reactors is assessed utilising a numerical model implemented in Aspen-Plus.

The numerical model has been preliminarily validated with an experimental activity conducted with a full scale gasification plant characterised by a patented air distribution system within the gasifier, called “double fire”.

During the experimental tests, using air as gasifying agent, no operative troubles of gasification occur, obtaining a good quality syngas mixing sewage sludge up to 30% (by mass) with woody biomass.

Using the experimental experience, the performances of the gasifier combined with a methanation plant has been assessed using a simulative approach implemented in Aspen-Plus environment.

The amount of methane which can be produced is in the range 10-20% of the overall mass of the fuel at the inlet conditions of the oxy-steam gasifier. The consumption of fresh water is not negligible and increases if the methane production is promoted.

The possibility to implement the oxy-carbon dioxide gasification could modify the syngas composition so that to be more coherent with the molar ratio of H_2/CO_2 that is required by the Sabatier reaction with the aim to reduce the water consumption.

7 References

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