Contribution to Circular Economy options of mixed agricultural wastes management: Coupling anaerobic digestion with gasification for enhanced energy and material recovery

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Abstract.

Anaerobic digestion (AD) is an established process for the treatment of organic wastes and the production of renewable energy. However, high amounts of digestate produced by AD plants require enhancement for further use. This study investigates a conceptual model for the digestate enhancement by using a downstream gasification. It is based on a 'systemic approach' considering the interactions of every contributing process into the dual system. The digestate was provided by an Italian AD plant, that treats mixed agricultural wastes of pig manure (43%), cow manure (20%), maize and triticale silages (25%), and cereal bran (12%). Digestate air gasification experiments were conducted, in a downdraft fixed-bed reactor, at temperature range from 750 °C to 850 °C, with λ varying from 0.14 to 0.34. Results have shown that gasification of digestate at 850 °C with $\lambda = 0.24$, increased producer gas yield (65.5 wt %), and its LHV (2.88 MJ Nm^{-3}). The gas is classified as medium heating value fuel, suitable to generate electricity of 971 kWhel day^{-1} to enhance the AD plant's economic viability. A carbonaceous material rich in macronutrients (P, K, Ca, Mg) was produced, with $R_{50} = 0.48$, suitable for carbon sequestration. The study offers a resource closed loop approach of converting AD digestate into energy and soil fertilizer. Useful suggestions for policy makers and business can be drawn.

Keywords: Anaerobic digestion, digestate, gasification, industrial symbiosis, circular economy, char, energy.

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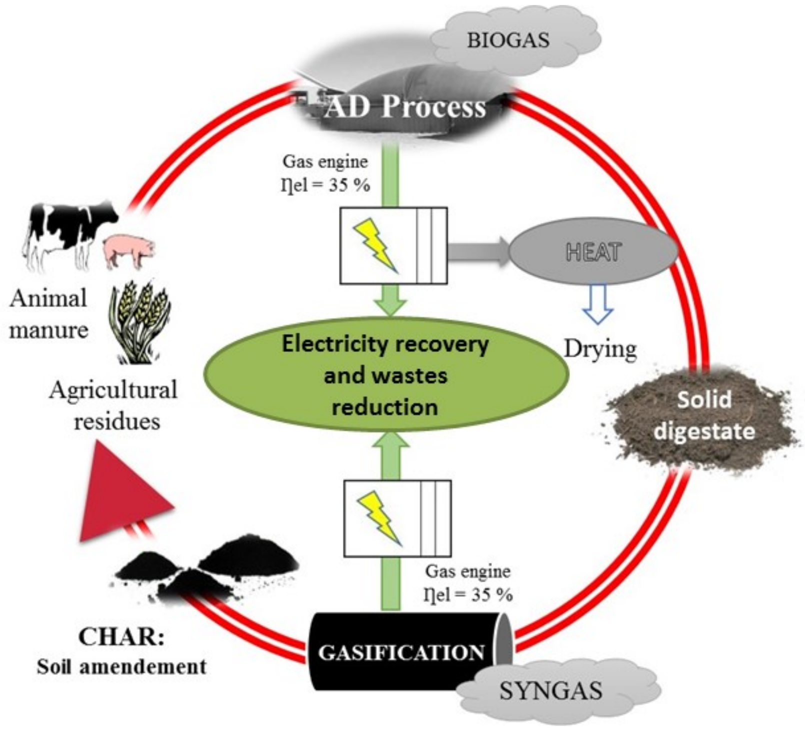
Graphical abstract



Highlights

- A Circular Economy concept of AD and gasification system was investigated.
- Industrial symbiosis of AD with gasification increased electricity production by 12%.
- Solid digestate drying requirements fulfilled by AD derived surplus heat.
- Optimal digestate gasification conditions were: 850 °C and λ=0.24
- Carbonaceous gasification derived char was rich in nutrients.
- Char's R_{50} = 0.48 makes it suitable for carbon sequestration.

Abbreviations

AD Anaerobic Digestion

CHP	Cogeneration Heat and Power
DM	Dry Matter
HRT	Hydrolytic Retention Time
OLR	Organic Loading Rate
λ (ER)	Equivalence ratio
VFAs	Volatile Fatty Acids
VS	Volatile Solids

1 Introduction

The European Commission, Council and Parliament provisionally agreed the Circular Economy Package of measures in wastes. This agreement moves the EU towards a higher level of sustainability in waste management. Anaerobic digestion (AD) is a key process for developing a Circular Economy. By closing the loops on the previously linear processes, AD can tackle waste, energy, sustainable food production and nutrient recycling challenges in a sustainable and circular manner. New ways could strengthen the AD industry as part of developing a Circular Economy.

In many European countries, anaerobic digestion (AD) plays an important role in the agro-industrial sector, displacing emission-intensive waste management strategies such as landfilling (Moretti et al., 2018). AD of agricultural residues is an established process, that not only contributes to the reduction of greenhouse gas emissions from the agricultural sector, but also produces biogas, a biofuel that can be used either to produce heat and electricity, or, after upgrading, to be injected into the natural gas grid (Monlau et al., 2013a, Sambusiti et al., 2013).

AD process seems to be a promising route for the treatment of organic wastes (Santi et al., 2015), but widespread uptake of small scale AD is limited due to economic costs and the safe disposal of digestate (Fuldauer et al., 2018). In addition, AD solves only partially the problem of material and energy recovery, since a significant part of the organic matter (*i.e* polysaccharides, lignin) remains in the so-called "digestate" (Monlau et al., 2015a, Santi et al., 2015), not reaching to the complete recovery of the organic value existing in the wasted matter. The efficiency of the organic matter (OM) conversion through mesophilic (i.e. 35 °C) or thermophilic (i.e. 55 °C) AD is generally in the range of 13–65%, and depends on the type of substrate fed to the digester, as well as on anaerobic reactor parameters, such as the organic loading rate (OLR) and the hydraulic retention time (HRT) (Monlau et al., 2015a). Legislation framework, does not help very much, because although it promotes AD broader implementation across the European continent, clear and solid directions on further valorisation of the obtained digestate, are not provided (Saveyn and Edder, 2014).

Digestate is rich in nitrogen, phosphorous and stabilized carbon. Its conventional valorisation route lies in its utilization as soil amendment and/or as fertilizer, under the condition that it fulfils the restrictions set by European Nitrate Directive (91/676/EEC), especially in the cases of livestock intensive areas (Nitrate Directive, 1997). Due to the accumulation of biogas plants in certain regions, in which intensive livestock farming is encountered, an oversupply of digestate is expected (Kuligowski and Luostarinen, 2011, Lacroix et al., 2014). If these agricultural areas cannot fully process large quantities of digestate, the surplus material must be transported to regions with nutrients deficit, thus increasing the operational cost of a biogas plant. Digestate composition depends on the input materials (feedstock) characteristics and the AD process conditions and needs enhancement for sustainable further uses. Digestate enhancement technological options need to be capable of dealing with a large range of inputs and feedstocks and low-cost, to achieve significant market penetration.

The valorisation of anaerobic digestate via thermal processes (*i.e* combustion, pyrolysis, gasification) is gaining interest (Monlau et al., 2015b, Sheets et al., 2015). Thermo-chemical processes are certainly interesting and complementary to anaerobic digestion. Anaerobic digestion (AD) is best suited to organic, putrescible waste streams, while thermochemical is currently best suited to highly homogenous dry materials (dried fibrous digestate). Research studies have reported the successful use of solid digestate as a fuel for combustion, considering emissions and the overall combustion behaviour (Kratzeisen et al., 2010, Pedrazzi et al., 2015). An anaerobic digestion (AD) process coupled with digestate composting, to produce a soil amendment of good quality was proposed by Cuadros Blázquez et al. (2018). Recently, the involvement of pyrolysis process on the further valorisation of solid digestate for biofuels production (*i.e* bio-oil, pyrolysis gas), and biochar, has attracted a lot of attention (Li et al., 2014, Monlau et al., 2015b, Monlau et al., 2016, Troy et al., 2013). The produced bio-oil and gas from pyrolysis, can fuel CHP systems, whereas the biochar, due to its physico-chemical properties, can be either used complementary on mineral fertilizers, or in soil preservation methodologies (Monlau et al., 2015b, Monlau et al., 2015c). In livestock intensive and isolated agricultural areas, the conjunction of AD plants with thermochemical conversion pathways, can assure a partial reuse of excess heat, which sometimes is lost, for the drying of the solid digestate, after a mechanical liquid/solid separation of the digestate (Monlau et al., 2015a, Monlau et al., 2015b).

From the literature review, it was revealed that only few publications reported the utilization of gasification for the digestate down-stream processing (Kuligowski and Luostarinen, 2011, Lacroix et al., 2014), and fewer studies modelled the potentials of incorporation of gasification process and anaerobic digestion in a dual system (Allesina et al., 2015, Li et al., 2015, Yao et al., 2017). Gasification is a thermal conversion process, which includes the stages of drying, pyrolysis, char gasification and combustion, that converts organic or fossil fuel based carbonaceous materials into a gaseous product, of low to medium heating value (Basu, 2010). This is achieved by material conversion at high temperatures (>700 °C), under a controlled amount of oxygen and/or steam. The gaseous mixture, consisted of hydrogen, carbon monoxide, methane, carbon dioxide and fractions of light hydrocarbons, can be used either as an energy carrier for clean energy or co-fired with other fuels in current power systems (Manara and Zabaniotou, 2014, Zabaniotou et al., 2014). The producer gas can be used to produce combined heat and power, through a CHP unit (Zabaniotou, 2014).

Aiming to improve the sustainability of the existing AD plants, several related biorefinery schemes have been investigated, during the last decades (Fabbri and Torri, 2016, Monlau et al., 2015a, Sheets et al., 2015). In most biorefinery cases, the utilization of the residual polysaccharides and lignin content of the solid-digestate, resulted to an extra-energy recovery and valuable materials production (Monlau et al., 2015a, Sheets et al., 2015).

The literature review made evident that although, AD is a key process for developing a Circular Economy and an important pillar of the European Circular Economy and a part of the European bio-economy improving European resource-efficiency (EBA, 2015), further technological leaps are needed. In this respect, a conceptual dual system of AD and digestate upgrading via downstream gasification process is proposed in this study. The study aims to fill that gap in knowledge and experimental data by investigating the gasification of the dried solid digestate, by-product of agricultural mixed wastes-based biogas plant. The study focuses on the gasification parameters optimisation, an evaluation of the agronomic properties of the char produce and an overall energy balance enhancement of the dual AD/gasification, providing recommendations for its deployment and further commercialisation.

The study contributes to the options of mixed agricultural wastes management, since it promotes the implementation of an environmentally-friendly solution, capable of acting either alternatively or complementary with the traditional agriculture processes, in closed loops. The technical innovations of this study are based on the materials and energetic fluxes exchanges, such as part of the heat produced through CHP system, which is used for solid digestate drying. The deployment of this system could also contribute to the wider implementation of resource efficiency principles, through: (i) maximization of energy recovery (*i.e* biogas, syngas) from agricultural residues, (ii) efficient reduction of wastes, (iii) production of a carbonaceous material for various application, such as for soil preservation and long term carbon sequestration (WRAP, 2012, Zabaniotou et al., 2015).

2 Materials and Methods

2.1 Digestate

The solid digestate was provided by a biogas plant located in Italy, with conventional configuration, including a first digester followed by a post-digester. The digester with the main characteristics of volume ~5,840 m^3, OLR of 120 t FM d^{-1}, HRT of 53 days and temperature at 45°C, was daily fed with a mixture of 43% animal sewage, 20% cow manure, 25% maize and triticale silages and 12% cereal bran. A daily production of 6.7 t DM day^{-1} of solid-digestate was obtained. Prior to any analysis and gasification trials, the sample was oven dried overnight at 105°C. Table 4 summarized the main characteristics of the AD plant.

2.2 Gasification protocol

In this study, gasification was carried out at medium to high temperatures, using air as gasifying agent, in a laboratory scale downdraft fixed-bed gasifier, at ambient pressure (Skoulou et al., 2008). The gasification system consisted of:

(i) stainless-steel reactor (height=500mm, diameter=12.5mm),

(ii) an individually controlled vertical electric furnace, with temperature measurement and modification in three different spots across the tube,

(iii) medium (N_2 or Air) providing system, using a vertical pipe in a downward flow, not preheated before entry (ambient conditions) and

(iv) gas collection system (**Fig. 1**).

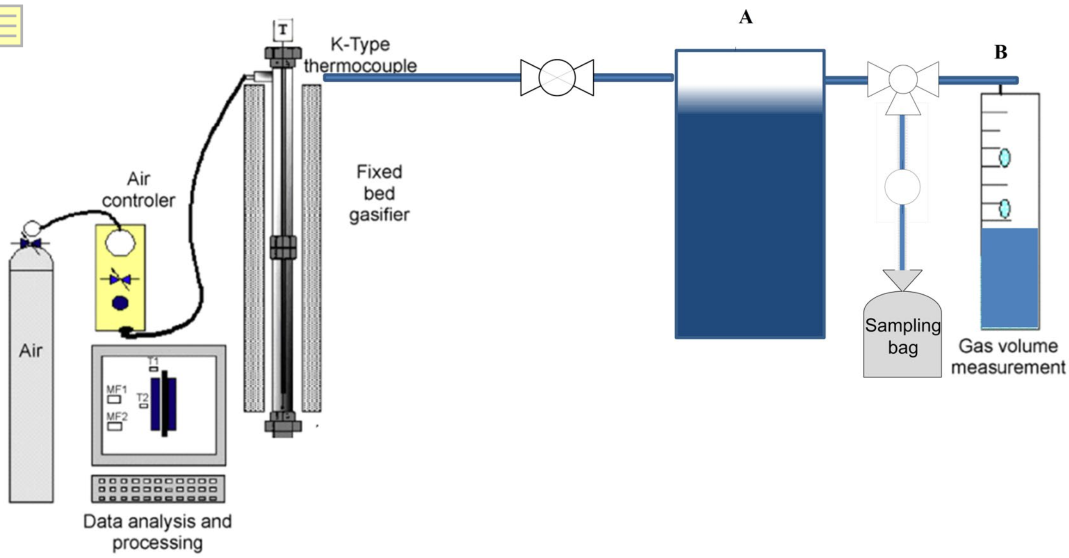


Figure 1. Downdraft fixed bed gasifier.

Gasification experiments were conducted at a temperature range of 750–850°C. Experiments were replicated twice, to achieve repeatability of results, and reproducibility of the process. Prior to each experiment, the reactor was dismantled, and the digestate, in powder form, was fed manually, by batches, from the top of the reactor. The reactor was sealed and nitrogen was purged continuously for 30 min, to remove the included air. The electric furnace was heated up (heating rate=30°C min^{-1}) to the desired temperature. By the time the desired temperature (750, 800 or 850 °C) was reached, the reactor was placed vertically on it. Temperature, measured by a K-type thermocouple (NiCr-Ni), increased and the gas medium was switched from nitrogen to air, when its instant temperature reached the desired one. During gasification, the produced gas through water displacement was collected in bottle A. When gasification ended, the produced volume was measured in bottle B, and the gas, through a gas pump, was transferred into a gas bag for GC analysis. Gas samples were taken after a few minutes for chromatographic analysis and at two additional time intervals.

The produced gas composed of CO, CO_2, H_2, CH_4, and other gaseous hydrocarbons (H_xC_y). The determination of the composition of the producer gas was performed offline, in a gas chromatographer (Model GC 6890N, Agilent Technologies), fitted with two columns, HP-PlotQ and HP-Molsive type (Manara and Zabaniotou, 2013). After the completion of the experiment, the reactor was removed from the furnace, left to cool down, dismantled and the produced ash was collected and weighted, whereas tar was collected on a solvent and its yield was estimated. Operational parameters such as temperature and λ were studied. λ is defined as the ratio of the actual amount of oxidising agent provided for oxidation to the theoretical amount required for complete combustion (Zabaniotou et al., 2014). Based on the elemental analysis of the raw material, the desired λ value for air gasification and the air-flow rate, the required air volume and consequently the air mass was determined.

2.3 Analytical procedure

Total solid (TS), volatile solid (VS) and ash (AS) were analyzed according to the APHA methods for digestate and char (APHA, 2005). Additionally, elemental analysis (CHNS) was also performed using an "Elementar Vario Macro Cube" analyser. The higher heating value (HHV) of the feedstock was calculated using the results from the elemental analysis of the sample and the following equation (Channiwala and Parikh, 2002):

$$HHV\ (MJ\ Kg^{-1}) = 0.3491*C+1.1783*H+0.1005*S-0.1034*O-0.0151*N-0.0211*Ash \quad (1)$$

Digestate and char pH determination was carried out by adding de-ionized water in a mass ratio 1:20. The solution was then hand shaken and allowed to stand for 5 min before measuring pH using a Basic Crison 20® pH meter. *Structural-carbohydrates* from cellulose and hemicelluloses together with "klason lignin" of solid digestate were measured, using a strong acid hydrolysis method, previously described by Monlau et al. (Monlau et al., 2015b). All monosaccharides (*i.e.* glucose, xylose, arabinose) were analysed by HPLC (Agilent® 1260) coupled to refractometric detection. The analysis was carried out with a Hi-PLex H column at 50°C. The eluent corresponded to 5 mM H_2SO_4 under a flow rate of 0.3 mL min^{-1}. A refractive index detector was used to quantify the carbohydrates. The system was calibrated with glucose, xylose and arabinose standards (Sigma−Aldrich®). Thereafter, cellulose and hemicelluloses contents were estimated as follows:

$$Cellulose\ (\%TS) = Glucose\ (\%TS) / 1.11 \quad (2)$$

$$Hemicelluloses\ (\%TS) = [Xylose\ (\%TS) + Arabinose\ (\%TS)] / 1.13 \quad (3)$$

Where:

1.11 is the conversion factor for glucose-based polymers (glucose) to monomers and 1.13 is the conversion factor for xylose-based polymers (arabinose and xylose) to monomers.

Elemental analysis (micro- and macro-elements) was performed by digesting 38.5 mg of sample in 2 mL of HNO_3 (68%) and 5mL of distilled water, in a closed vessel microwave digester (*µondes US49*), for three temperature cycles of 140, 170 and 190°C, respectively. The metals in the solution were analysed by inductively coupling plasma spectrometry-optical emission spectroscopy (ICP-OES Agilent 720).

To evaluate the carbon sequestration potential of both digestate and char, the R_{50} coefficient was calculated. R_{50} corresponds to an index for quantifying char recalcitrance and screening char with respect to their carbon sequestration potential (Harvey et al., 2012). The R_{50} uses the energy required for thermal oxidation of the char (normalized to that needed for the oxidation of graphite) as a measure of recalcitrance. The R_{50} recalcitrance index was calculated according to the following equation:

$$R_{50}Ch = T_{50}Ch / T_{50}\,GRAPH \quad (4)$$

Where:

$T_{50}Ch$ and T_{50} GRAPH are the temperature values corresponding to 50% oxidation/volatilization of chars (Ch) and graphite, respectively.

Values for $R_{50}Ch$ are obtained directly from TG thermograms appropriately corrected for water and ash content. $T_{50}GRAPH$ was assumed to be 886 °C (Harvey et al., 2012). The R_{50} was interpreted considering the following recalcitrance/carbon sequestration classes: Class A ($R_{50} \geq 0.70$), Class B ($0.50 \leq R_{50} < 0.70$), or Class C ($R_{50} < 0.50$).

2.4 Energy analysis

The energetic requirements for drying the solid digestate were estimated based on data provided by the owner of the AD plant. The energetic requirements for drying the solid digestate E_{DD} (kWh_{th} day^{-1}) on a daily basis, was calculated using the modified equation used by Barakat et al. (Barakat et al., 2014).

$$E_{DD} = E_{Heat} + E_{Evaporation} \quad (5)$$

where:

E_{Heat} (kWh_{th} day^{-1}) is the energetic requirement to increase the temperature of water and digestate from 25°C to 105°C and

$E_{Evaporation}$ (kWh_{th} day^{-1}) corresponds to the energetic requirement for water evaporation at 105°C.

E_{Heat} and $E_{Evaporation}$ were calculated according to the equations 6 and 7:

$$E_{Heat} = m \times Cp * [T_{Final} - T_{Initial}] / 3600 \quad (6)$$

m (kg day^{-1}) is the daily mass of water and solid digestate;

Cp is the water specific heat (4.18 kJ $kg^{-1}°C^{-1}$);

$T_{Initial}$ (°C) is the initial temperature of the substrate suspension, assumed as 25°C;

T_{Final} (°C) is the final temperature at 105°C.

$$E_{Evaporation} = [m_{water} * Lv] / 3600 \quad (7)$$

Lv is the latent heat of vaporization equal to 2257 kJ kg^{-1};

m water (kg day^{-1}) is the daily mass of water in the solid digestate.

The lower and higher heating value (MJ m^{-3}) of the producer gas were calculated using the molar fractions of the detected compounds in the produced gas and the following equations (8 and 9) (Li et al., 2004; Lv et al., 2004) :

$$LHV= [30\times v/v\%CO + 25.7\times v/v\%H_2 + 85.4\times v/v\%CH_4 + 151.3\times v/v\% (C_2H_4+C_2H_6)] \times 0.42 \quad (8)$$

$$HHV= [12.75\times v/v\%H_2 + 12.63\times v/v\%CO + 39.82\times v/v\%CH_4 + 63.43\times v/v\% (C_2H_4+C_2H_6)] /100 \quad (9)$$

Furthermore, additional qualitative indicators were introduced to provide a more deliberate characterisation of the obtained gasification gas. These included syngas yield (H_2+CO), and H_2/CO ratio. The H_2/CO ratio is an important parameter for a further utilization of producer gas as a feedstock in the synthesis of chemicals, such as methanol or Fischer Tropsch fuels. H_2/CO ratio desired values were considered those which exceed 2:1, depending on selectivity (Wender, 1996). In general, a gas with high syngas yield and high H_2/CO ratio is a gas with a high calorific value, suitable as fuel and for a wide range of applications.

Finally, to estimate the energetic balance of the entire process, it was assumed that biogas and syngas produced from AD and gasification processes respectively were used to fuel a CHP unit. The energetic efficiency of the conversion system was considered as 35 % for electricity and 50% for heat (Monlau et al., 2013b). Furthermore, it was assumed that heat wastes (exhaust gases and hot cooling water) from the CHP system can be used to cover the thermal needs of the digestate process.

3 Results

Results on gasification producer gas, char yields and their quality are discussed here; yields and quality depend on the digestate characteristics. Energy analysis is also provided.

3.1 Digestate's characteristics

The chemical composition of the AD digestate used in this study, is presented in **Table 1**. The digestate was composed of 17.0 g 100 g^{-1} TS of cellulose, 12.2 g 100 g^{-1} TS of hemicelluloses and 34 g 100 g^{-1} TS of lignin. It contained a high C content of 43 g 100 g^{-1} TS and ash content of 9.5 g 100 g^{-1} TS. The results are in agreement with reported results by Santi (Santi et al., 2015), showing that an important quantity of cell wall polymers remained in the digestate fraction.

Table 1. Solid digestate analysis

Parameters (Units)	**Digestate**
pH	8.6 (±0.04)
VS (%TS)	89.5 (±0.1)
Ash (%TS)	9.5 (±0.1)
C (g 100 g^{-1} TS)	43.0 (±0.07)
H (g 100 g^{-1} TS)	6.2 (±0.03)
N (g 100 g^{-1} TS)	1.3 (±0.03)
S (g 100 g^{-1} TS)	0.14 (±0.05)
O (g 100 g^{-1} TS)[a]	39.5
Cellulose (g 100 g^{-1} TS)	17.0 (± 0.9)
Hemicellulose (g 100 g^{-1} TS)	12.2 (± 1.7)
Lignin (g 100 g^{-1} TS)	34.0 (±1.2)

3.2 Gasification products: effect of temperature and λ on products yield

Digestate was subjected to gasification at 750, 800 and 850°C, using air as gasification agent, at λ values ($\lambda = Air_{experimental}/Air_{theoretical}$) ranging from 0.14 to 0.34. The effect of process parameters (T and λ) on product yields is depicted in **Fig. 2**.

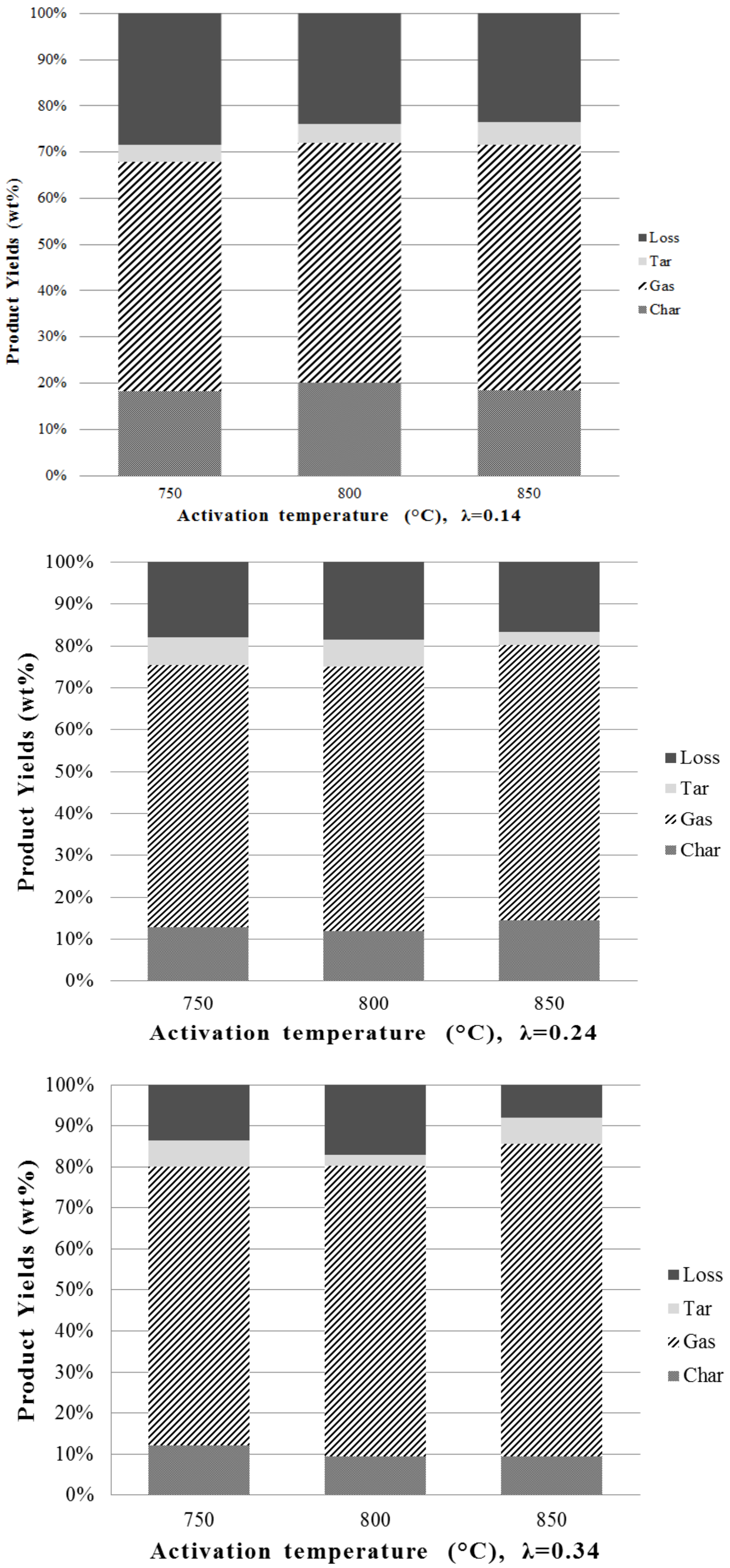


Figure 2. Effect of activation Temperature and λ on a product yields (wt%) and mass balances.

The experimental results highlighted the major effect of temperature on product yields. At 750 and 800°C the char and gas yields remained almost constant at every λ-value studied. A further temperature increase favoured the production of gaseous product, because of the extensive devolatilization of the digestate, thus reducing char yield. Moreover, due to the intense temperature applied (850°C), devolatilization and oxidation of condensed tar also occurred, thus reducing tar yield, resulting in the presence of condensed hydrocarbons in the interior of the reactor. As a result, under higher temperature, higher carbon conversion was achieved (Devi et al., 2003). At the lowest λ values used, high temperature pyrolysis (cracking) occurred rather than gasification (oxidation). An increase of λ resulted in further devolatilisation (volatiles released from the solid phase) and oxidation of the sample, thus decreasing char yield and tar yield (wt%) in favour of gaseous products.

A simplified mechanism describing the evolution of permanent gases during gasification is presented below (Cao et al., 2006; Chen et al., 2013; Fryda et al., 2008; Nipattummakul et al., 2010; Panopoulos et al., 2006):

$$C_{(s)}+O_{2(g)}\rightarrow CO_{2(g)} \quad \text{Complete oxidation} \quad (10)$$

$$C_{(s)}+1/2O_{2(g)}\rightarrow CO_{(g)} \quad \text{Partial oxidation} \quad (11)$$

$$C_{(s)}+CO_{2(g)}\rightarrow 2CO_{(g)} \quad \text{Boudouard reaction} \quad (12)$$

$$CH_{4(g)}+CO_{2(g)}\rightarrow 2CO_{(g)}+2H_{2(g)} \quad \text{Methane dry reforming} \quad (13)$$

$$CH_{4(g)}+2H_2O_{(g)}\rightarrow CO_{(g)}+3H_{2(g)} \quad \text{Steam reforming of methane} \quad (14)$$

$$Tar \rightarrow CH_{4(g)}+ H_{2(g)}+H_2O_{(g)}+C_nH_{m(g)} \quad \text{Tar reforming} \quad (15)$$

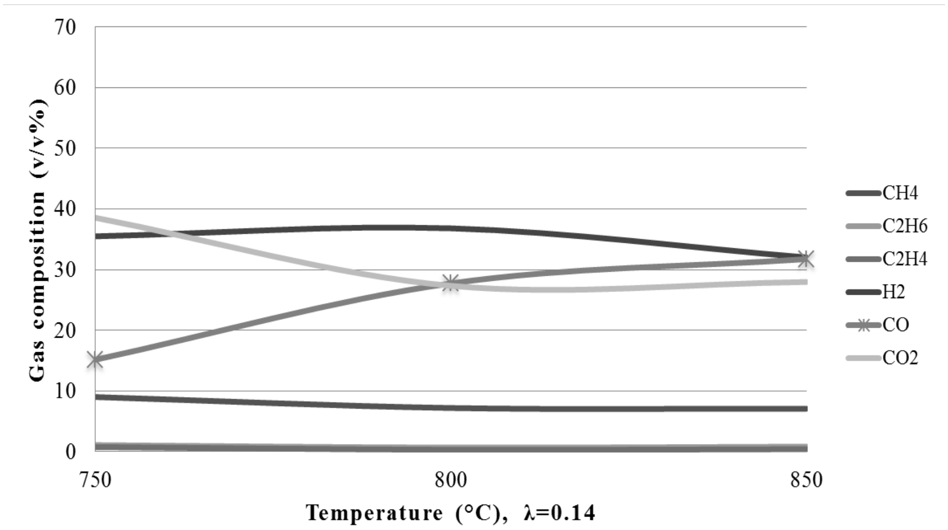
3.3 Gasification products: effect of temperature and λ on producer gas composition

For the gasification experiments, λ varied between 0.2 and 0.3. With a lower λ value, the char was not fully converted into gases, giving rise to higher tar production, resulting in several problems including incomplete gasification and excessive char formation. Gasification at a λ >0.25, resulted mostly in gaseous products. At higher temperatures, further oxidation was resulted. However, a high λ value resulted to an excessive formation of undesired CO_2, reducing the heating value of the producer gas (Basu, 2010). The effect of process parameters on the composition of the gaseous product is depicted in **Fig. 3**.

During gasification at the lower temperature, oxidation reactions (exothermic) took place, resulting to CO_2 and CO. As temperature increased, Boudouard reaction towards the production of CO in expense of CO_2, was favoured. Under elevated temperatures, CH_4 reforming occurred, increasing slightly the generation of CO and H_2. Likewise, under the same conditions, reforming of C_2H_4 and C_2H_6 was also noticed for all λ-values studied.

The quantitative characteristics of the produced gas, as identified by metrics such as syngas yield, LHV, HHV and H_2/CO ratio, are presented in **Table 2**. Producer gas yield increased as temperature increased (reactions 11-15), at all λ values. On the other hand, syngas yield decreased when λ values increased, under the same temperature (reaction 10).

The aforementioned process parameters greatly affected the heating value of the obtained gas. Over the studied temperature range (750 up to 850 °C), the LHV and HHV varied from 1.13 up to 4.25 MJ m^{-3} and from 1.24 to 4.58 MJ m^{-3}, respectively. It seems that a higher temperature favoured H_2 and CO production (reactions 11-15). Both LHV and HHV showed better values with a λ=0.14. This can be attributed to the fact that under these conditions (λ=0.14), the predominant step was high temperature pyrolysis, thus producing a gas of high calorific content.



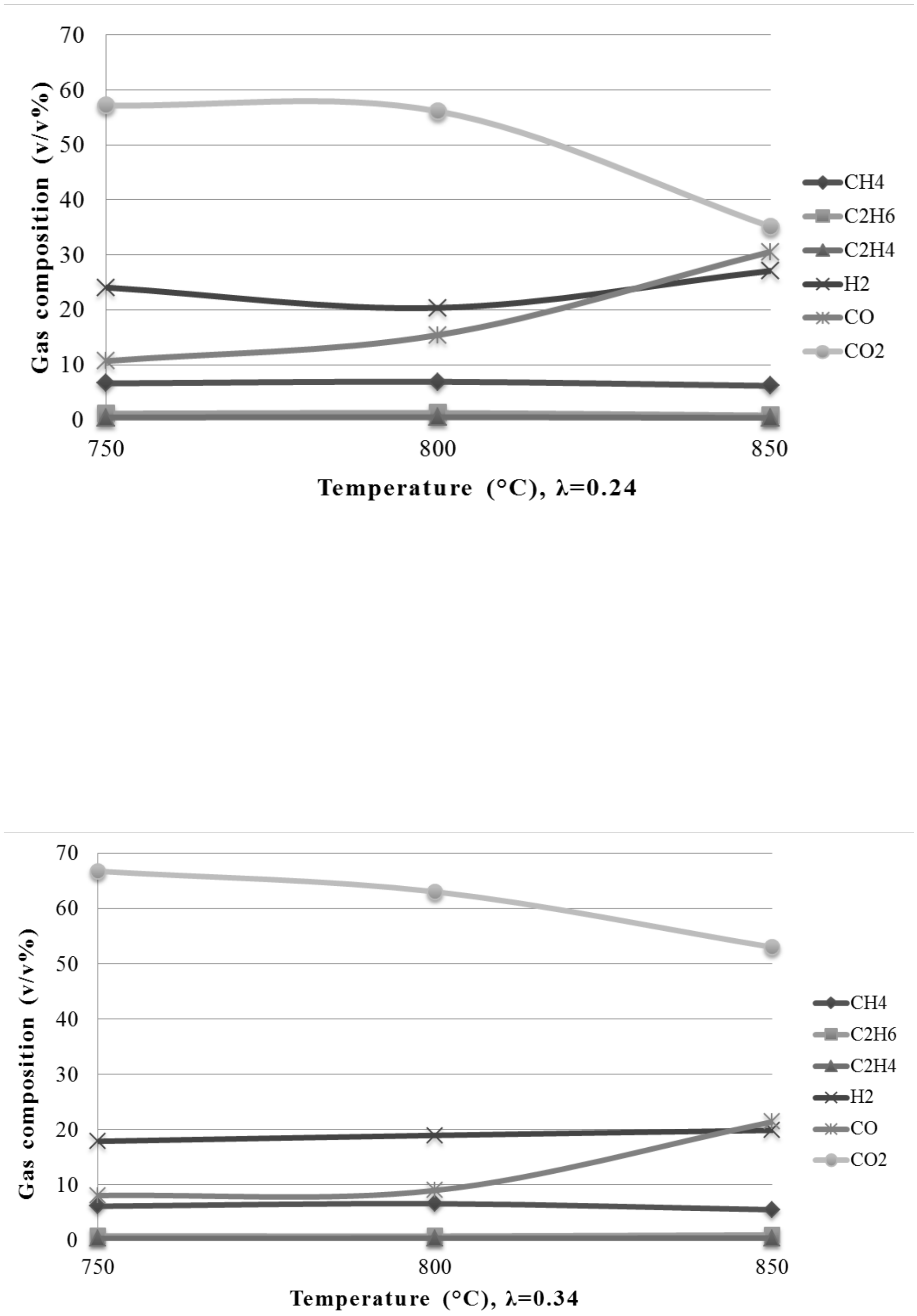


Figure 3. Effect of gasification temperature and λ values on gas composition

Table 2. Producer gas characteristics.

Temperature °C	**750**	**800**	**850**
λ = **0.14**			
syngas yield (%v/v)	17.58	23.73	25.18
H2/CO	2.34	1.32	1.01
LHV (MJ m^{-3})	3.52	3.92	4.25
HHV (MJ m^{-3})	3.87	4.27	4.58
λ = **0.24**			
syngas yield (% v/v)	8.64	9.15	17.27
H_2/CO	2.25	1.36	0.90
LHV (MJ m^{-3})	1.80	1.97	2.88
HHV (MJ m^{-3})	1.98	2.13	3.10
λ = **0.34**			
syngas yield (% v/v)	5.05	5.40	10.25
H_2/CO	2.21	2.20	0.88
LHV (MJ m^{-3})	1.13	1.22	1.88
HHV (MJ m^{-3})	1.24	1.34	2.01

At higher λ, the production of CO_2, through favoured combustion reactions, increased. The presence of CO_2 in the gasification gas is not desirable, since it implies both a dilution effect of the gas heating value and a reduction in the formation of CO, (reactions of production and consumption of CO and CO2 are the water-gas shift or the Boudouard reactions).

The H_2/CO ratio is a critical value for the choice of producer valorisation pathway. This ratio exhibited a maximum value (2.2-2.3) at lower temperature, for all λ values studied. With increasing temperature, CO produced at the expense of CO_2, thus reducing the H_2/CO ratio in values <1, not recommended for synthesis of chemicals, or Fischer Tropsch fuels. Based on the experimental results, λ=0.24 and temperature~850°C were considered as the optimal conditions for a gaseous fuel production to be used for CHP.

3.4 Char characteristics

In order to use gasification solid remained by-product (char) in soil amendment applications, an extensive analysis of chars was performed (Zabaniotou, 2014). During the last decades, biochar has attracted attention as soil amendment for soil properties improvement with valuable nutrients (P, K, Ca, Mg), contributing to carbon sequestration, (Kuligowski and Luostarinen, 2011; Monlau et al., 2016; Srinivasan et al., 2015). Literature data on the agronomic properties of char derived from the gasification of solid digestate, are scarce (Hansen et al., 2015).

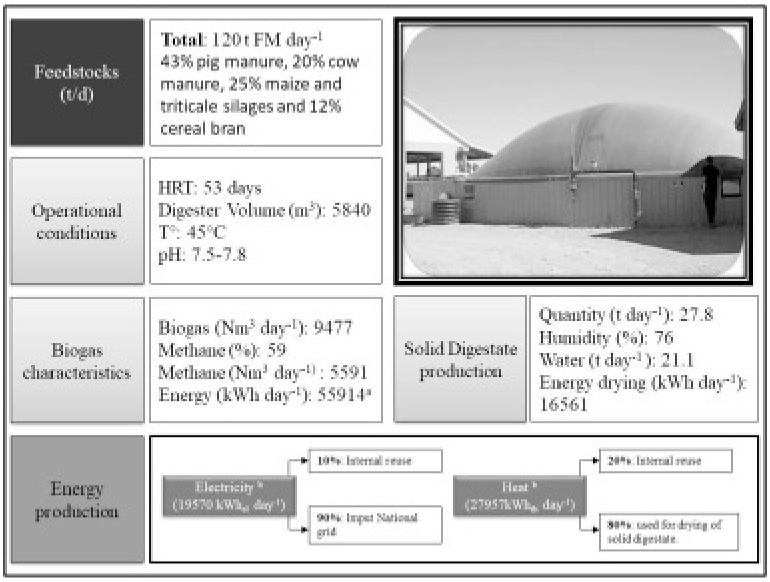
In this study, the digestate showed an alkaline pH (8.6). Digestate alkalinity is commonly observed and it is mainly due to the degradation of VFAs and the production of ammonia during the AD process. Control of pH can improve the addition of basic compounds or carbonates into the digester (Tambone et al., 2010). As **Table 3** depicts, the gasification-char exhibited a high pH value of 11.4, which is in agreement with reported values (Monlau et al., 2016; Opatokun et al., 2017), VS of 61.8% TS and ash content of 38.2 (± 2.6) % TS. The content of C on the gasification-char was found higher by 50% in the dry mass (DM) than the digestate, which is in agreement with the European Biochar Certificate (EBC, 2012).

Table 3. Proximate, ultimate analysis and concentrations of nutrients and heavy metals of the digestate and gasification char (compared with standards).

Compounds	**Digestate (Monlau et al., 2016)**	**Char**	**EBC guidelines**	**IBI standard**
pH_1:25 (H_2O)	8.6 (±0.04)	11.4 (±0.01)		
VS (% TS)	89.5 (± 0.1)	61.8 (± 2.5)		
Ash (% TS)	9.5 (± 0.1)	38.2 (± 2.6)		
R_{50} (%)	34	47.6		
C (g. 100g^{-1} TS)	43.0	66.5	> 50	
H (g. 100g^{-1} TS)	6.2	8.7		
N (g. 100g^{-1} TS)	1.3	1.4		
S (g. 100g^{-1} TS)	0.14	0.196		
Mg (mg kg^{-1} TS)	3689	19446		
Ca (mg kg^{-1} TS)	10814	69139		
Cd (mg kg^{-1} TS)	-	0.15	< 1.5	1.4-39
Cr (mg kg^{-1} TS)	1	786	< 90	64-1200
Cu (mg kg^{-1} TS)	11	94	< 100	63-1500
Fe (mg kg^{-1} TS)	630	8971		
K (mg kg^{-1} TS)	11966	48132		
Al (mg kg^{-1} TS)	356	3418		
Mn (mg kg^{-1} TS)	97	550		
Na (mg kg^{-1} TS)	1886	16285		
Ni (mg kg^{-1} TS)	1	1555	< 50	47-600
P (mg kg^{-1} TS)	5289	17400		

Pb (mg kg^{-1} TS)	-	48	< 120	70-500
Zn (mg kg^{-1} TS)	35	256	< 400	200-7000

Table 4. Characteristics of the agricultural biogas plant. a10 kWh Nm-3 methane; bThe energetic efficiency of the conversion system CHP was considered as 35 % for electricity and 50% for heat.



The distribution of the main macronutrients (N, P, K, S, Ca, Mg) present in both the digestate and its resulting char were presented in **Table 3**. The content of N and S elements remained similar in digestate and char; no char enrichment was noticed due to the fact that most N and S compounds volatilize above 200°C and 375°C (Kookana et al., 2011). A significant increase of the P (17,400 mg kg^{-1}TS), K (48,132 mg kg^{-1}TS), Ca (69,139 mg kg^{-1}TS) and Mg (19,446 mg kg^{-1}TS) macronutrients content was observed on the char. Similar results have been previously reported (Kuligowski and Luostarinen, 2011; Monlau et al., 2016; Opatokun et al., 2017). Kuligowski and Luostarinen (Kuligowski and Luostarinen, 2011), reported that the main macronutrients of gasification char (from solid digestate of AD plant treating manure) were calcium (311 g kg^{-1}), phosphorus (54.4 g kg^{-1}) and potassium (34.7 g kg^{-1}) making the char a good candidate for fertilizer. In parallel, an enrichment of micronutrients (Fe, Cu, Mn, Ni, Zn) was noticed on char. This is in agreement with previous studies, that reported an enrichment of the main macronutrients (P, K, Mg, Ca) and micronutrients on biochar, after pyrolysis of solid anaerobic digestate of food wastes and agricultural wastes respectively (Monlau et al., 2016; Opatokun et al., 2017).

Toxic compounds can be present in the char, preventing its broader use for agronomic purpose, depending on the composition of the precursor material. To assess the possibility to use gasification char as soil amender, it is of prime interest to estimate the potential toxicants, especially heavy metals content, **Table 3**. Except for Cr and Ni, analyses of heavy metals of most samples were under the threshold values recommended by International Biochar Initiative and European Biochar Certificate (EBC, 2012; IBI, 2014). The higher concentration of Ni and Cr compared to threshold values is not worrying because it was due to some experimental limitations: the high content of Ni and Cr were can be attributed to the degradation of K-type thermocouple during the high temperature process or leaching from the gasification reactor made up of stainless steel. Finally, a high concentration of Al (3,418 mg kg^{-1}TS) was also estimated, which is in agreement with values reported by Kataki et al. (Kataki et al., 2017) on char from rice husk gasification.

Further research is needed on the characteristics of the char obtained from gasification of solid anaerobic digestate to assess its implication in soil amendment. It will be interesting to investigate the bioavailability of the main macronutrients presents in gasification char along with growth plant tests, to assess their suitability benefit use for agronomic application Opatokun et al. (Opatokun et al., 2017).

For the estimation of the carbon sequestration potential by char in soils, the recalcitrance index (R_{50}) was calculated from T_{50} values, using the obtained thermogravimetric curves corrected for moisture and ash content. R_{50} values of 0.34 and 0.48 were determined for digestate and char, respectively (**Table 3**). Such results confirmed the recalcitrant structure of gasification char which is more apparent in char than in digestate sample, confirming char's potential in carbon sequestration. The R_{50} was interpreted considering the following recalcitrance/carbon sequestration classes: Class A ($R_{50} \geq 0.70$), Class B ($0.50 \leq R_{50} <0.70$), or Class C ($R_{50} < 0.50$). Considering these R_{50} values, the obtained char from gasification should be ranked in the third class. A lower stability was observed comparing the obtained results with those of biochar from pyrolysis of manure and residues of agricultural crops (Harvey et al., 2012). Nonetheless, the R_{50} values were in the same range (R_{50} =0.40/0.41) with the previously values reported by Monlau et al. (Monlau et al., 2016), on biochar obtained from pyrolysis (at 600°C) of solid anaerobic digestates.

3.5 Dual system's overall energy balance

In **Fig. 4**, the overall energy balances of coupling AD with gasification process, in a basis of on one-day running process is presented. The methodology for the detailed calculations, were described by Monlau et al. (Monlau et al., 2015b). The biogas produced from the AD plant converted into electricity and heat through a CHP unit, for which 10% of electricity and 20% of heat were internally reused for the AD plant operation. A net daily production of 17,613 kWh_{el} and 22,366 kWh_{th} were documented.

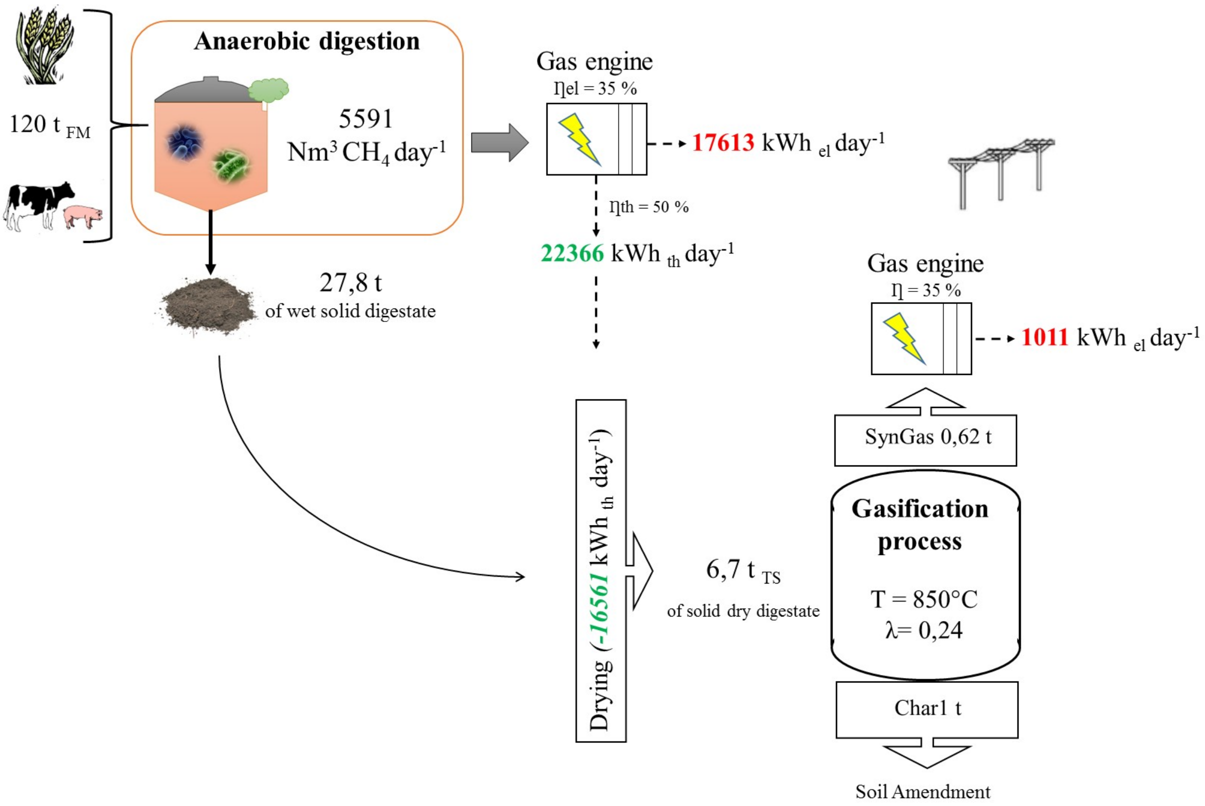


Figure 4. Energy balance of coupling AD with gasification process (base: one-day running process)

It was shown that the daily excess heat produced from the CHP unit (22,366 kWh_{th}) was sufficient for a daily drying of the solid digestate. It was demonstrated that a higher energy recovery could be achieved by coupling AD/gasification process, due to avoiding part of heat losses into the atmosphere. Such results were in agreement with previous results reported by Monlau et al. (Monlau et al., 2015b), demonstrating that waste-heat from the biogas unit was enough to fulfil the drying needs for the solid digestate, through a belt dryer system.

At the gasification process conditions 850°C and λ=0.24, gas yield exhibited its maximum heating value (LHV=2.88 MJ m^{-3}), classified as a medium heating value fuel. Based on the above, the overall energy balance of the dual AD/gasification process was determined by considering: a daiy processing of solid digestate of 6.7 t TS; a daily syngaz production of 0.62 tons equivalent through conversion into a CHP system to 971 kWh_{el} day^{-1}. .

In a similar study on sewage sludge, Lacroix et al. (Lacroix et al., 2014) improved energy recovery by 90%. The produced 971 kWh_{el} per day was a relatively small amount of energy, appropriate for a small scale decentralised gasification system to be used. Such system, as reported by Manara and Zabaniotou. (Manara and Zabaniotou, 2014), was proved to be suitable. The dual system could also effectively reduce any production of secondary wastes. It was calculated that approximately 1 t of final residues (char) are obtained from around 6.9 t of solid digestate treated, thus efficiently contributing to waste minimization.

4. Discussion and recommendations for policy making and businesses development

The study is relevant not only from an academic but also from a policy making and business perspective, particularly in the EU context. The European Commission took several important initiatives in resource efficiency during the years 2011–2015, culminating with the Circular Economy Package in December 2015. Useful managerial suggestions for policy makers and businesses about the implementation of innovative options for agricultural waste management can be drawn, related to gains in energy recovery with the efficient reduction of wastes, elimination of pathogens in digestate and production of a carbonaceous material for soil preservation and long-term carbon sequestration.

The coupled AD/digestate gasification (DG) system proposed in this study shows real promises for future implementation, offering a continual circulation of resources in the long-term rather than offering a temporary solution to the problem of agricultural waste. It provides a source of renewable energy in the form of CHP besides biogas and a soil amendment made from the digestate, which is rich in organic matter and nutrients.

The system can be stand alone or incorporated in a Circular Economy territorial model. The stand-alone dual system offers a management option for the agricultural wastes, mitigating waste-to-landfill, enhancement options for the digestate, and the energy required for the AD plant. It offers a resource closed loop by converting digestate generated by biogas plant into commodity consumed by the plant (energy) and a product to be used in agriculture (char), as biofertilizer, complying with the Circular Economy concept, where waste resulting from a process, AD in the present case, can be used as primary inputs for other process, gasification in the present case, towards achieving environmental, economic, and social advantages (Fig. 5A). In a scheme that involves separate industries in a collective approach to competitive advantage involving exchange of materials, energy and services, many AD plants could be coupled with one central gasification plant for a Circular Economy territorial model. In the case of regionally centralised large-scale gasification unit, digestate from various AD plant can be collected, transported, and be used as feedstocks in the large-scale centralised gasification-based CHP plant (Fig. 5B).

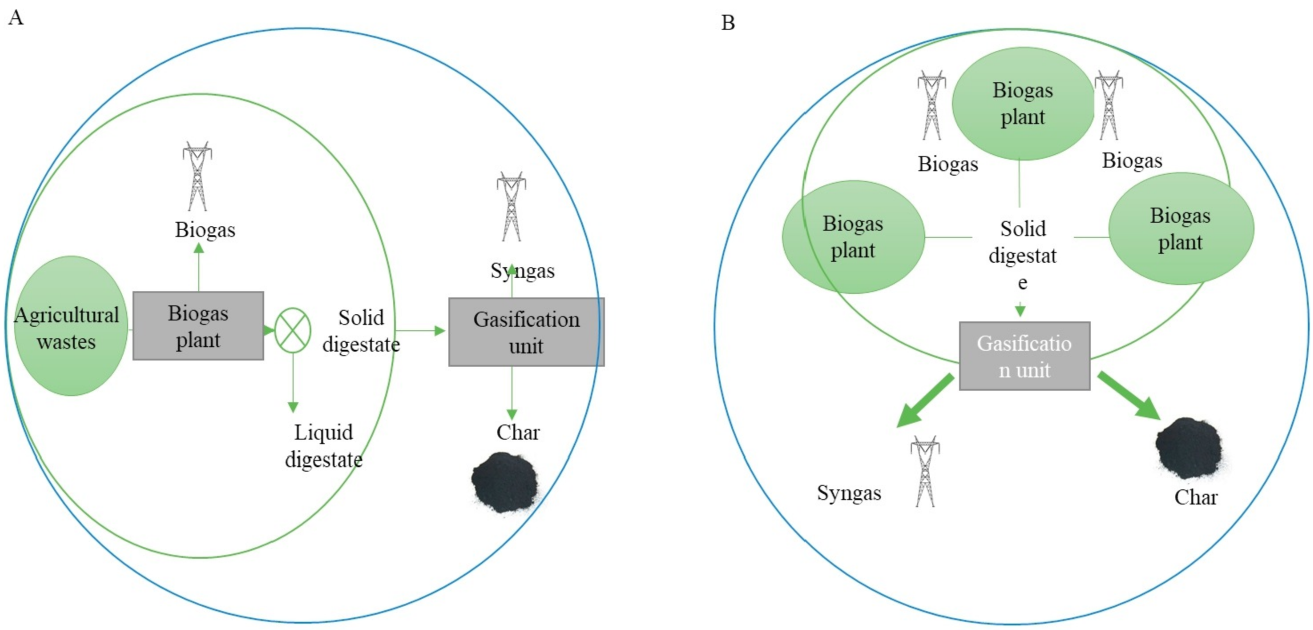
The amount of electric energy and biochar produced in the system depend on the amount of digestate and agricultural waste production rates. Therefore, the efficiency of the application of the approach may be different depending on specific area and case considered: stand-alone dual system (Fig. 5A) or multiple biogas plants in cascade conjunction with a central gasification unit (Fig. 5B). Three main environmental benefits can be obtained:

i) Sustainable management of digestate.
ii) Less amount of energy produced using traditional processes.
iii) Less GHG emissions in atmosphere, due to lower energy production using conventional processes.

In addition, economic advantages can be expected in terms of cost reduction about waste disposal and energy procurement from outside, even if pre-feasibility study is required.

The application of the above system in the 'Circular Economy' across the agri-food sector, will reduce wastes while also making best use of the 'wastes' produced by using economically viable processes and procedures to increase their value. It presents a major opportunity for the development of a Circular Economy (CE) using innovative conjunction of applied technologies and profitable business practices to address the utilization of agricultural wastes, byproducts and co-products.

The proposed system could be a smart sustainable rural energy infrastructure, in rural farming communities, suited to the Circular Economy principle. Dual AD and gasification system with farms supplying crop and slurry feedstock and local industry supplying food waste, can generate electricity and fertilizer which can then be used by the local community (Blades et al., 2017), rather focusing on the social benefits that a transformation from a linear to a Circular Economy would entail.



A. Closing loops: Stand-alone dual system coupling an AD and a Gasification unit in a closing loops concept

B. Circular Economy: Multiple regional biogas plants served by a central gasification unit for in a circular economy model

Figure 5. Conceptual model of: **A)** a standalone dual system of AD with gasification for electricity and biochar recovery; **B)** a symbiotic scheme of various biogas plants with a central gasification unit for electricity and biochar recovery.

5. Conclusions

This study investigated the coupling of a typical anaerobic digestion (AD) with a gasification system in a cascade flow of materials. It showed the dual system's potentiality to increase the renewable energy efficiency and to produce a carbonaceous material for agronomy purposes, towards the transition to an inclusive and Circular Economy. Although the proposed process system might be used in the conventional practice, the closed loop components constitute the differences from the linear model to a circular model of waste management. In this Circular Economy concept, higher energy recovery could be achieved. The coupled AD/digestate gasification system proposed in this study shows real promises for future implementation, offering a continual circulation of resources in the long-term rather than offering a temporary solution to the problem of agricultural waste.

The dual system of AD with air gasification of the digestate produced by a commercial biogas plant was studied.

The optimisation of the digestate gasification was achieved at 850 °C and $\lambda = 0.24$, resulting in a medium heating value gas fuel with LHV of 2.88 MJ Nm^{-3} and $H_2/CO = 2.3$, classified as medium heating value fuel, suitable for CHP production.

From an energetic and economic point of view, results have shown that the heat excess produced from the anaerobic digestion plant is sufficient for a complete drying of the solid digestate. Furthermore after gazification process, it can be generated a surplus of electricity of about 971 kWh_{el} day^{-1}, enhancing thus the economic viability of the AD plant.

In parrallel, the obtained carbonaceous material exhibited pH of 11.4, TS of 89.5%, over 50% more C content compared to the precursor material, and a significant increase of P (17,400 mg kg^{-1}TS), K (48,132 mg kg^{-1}TS), Ca (69,139 mg kg^{-1}TS) and Mg (19,446 mg kg^{-1}TS) nutrients content.

From environmental point of view, three main environmental benefits can be obtained: i) sustainable management of digestate; ii) less amount of energy produced using traditional processes, and iii) less GHG emissions in atmosphere, due to lower energy production using conventional processes iv) by generating a carbonaceous material rich in nutrients and recaciltrant carbon contributing to carbon sequestration in soil. In addition, economic advantages are produced in terms of cost reduction about waste disposal, fertilizers requirment and energy procurement from the grid.

Further research is needed on char's implication on soil amendment applications, mainly on the bioavailability of the main macronutrients present in gasification char, along with growth plants tests, to assess their use in agronomic application.

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