Methanol production from Refuse Derived Fuel: A preliminary analysis on the influence of the RDF composition on process yield

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Abstract

Currently, the production of methanol via high-temperature gasification of Refuse-Derived-Fuel (RDF) can be deemed as an excellent example of circular economy: it represents a promising alternative to Waste-to-Energy (WtE) and environmental impact improvement, thus leading to the reduction of carbon footprint and greenhouse gas emissions. The Waste to methanol (WtM) process can be divided into four main sections, namely RDF gasification, syngas purification, conditioning, and methanol synthesis. Methanol manufacturing needs a suitable syngas composition; therefore, the ultimate goal is to achieve a tailored gasification unit in order to decrease conditioning step efforts. In this work, a steady-state simulation of gasification unit has been developed using Aspen Plus. Considering that RDF is typically characterized by a remarkable composition variability, an extended parametric study, where RDF composition is represented in terms of ash, moisture, and combustible contents of the waste, has been undertaken as a preliminary approach. A synoptic description of results given in terms of ternary diagrams has been chosen to represent the main process parameters. Hence, this exploratory study allows to assess the process features associated with different feeding characteristics and provides preliminary suggestions to recognize process strategies to simplify design step and improve the process.

Abbreviations

Ash&In	Ash + Cl + N + S mass fractions
CHO	Combustible waste mass fraction
GHG	GreenHouse Gas
HHV	Higher Heating Value
LHV	Lower Heating Value
MM	Methanol Module
MSW	Municipal Solid Waste
NG	Natural Gas
RDF	Refuse Derived Fuel
WtE	Waste to Energy
WtM	Waste to Methanol

1. Introduction

In the last decades, political and social efforts have been taken towards replacing fossil fuels with renewable sources, such as biomass, to reduce the environmental impact associated with power generation and production of chemical products. This trend is driven by the low CO_2 emissions provided by renewable resource employment, which contributes positively in the direction of decreasing the greenhouse effect. Hence, the European Parliament subsidized this improvement with Renewable Energy Directive in EU (2009/28/EC) that establishes the year 2020 as a deadline to achieve mandatory targets consistent with 20% share of renewable sources in overall Community energy consumption, and 10% share of biofuels used for transport [1] . On the same directive is promoted the second-generation biomass employment, limiting the food-crops derived biofuel target contribution up to 7% and, on the other hand, counting twice the biofuels derived from waste, residues, non-food cellulosic material or lignocellulosic material. Indeed, considering GHG emission linked to land use, biofuels produced from food crops could even have a higher impact than the fossil reference [2,3]. In this light, both from an economic and an environmental standpoint, producing chemicals from waste appears as a concrete and attracting alternative to replace conventional chemicals and fuels production. Indeed, urea and methanol production from RDF in a sustainable and economic process has been studied [4], and several companies are either already running at the industrial scale or studying at a pilot plant scale processes to produce urea, ammonia or methanol from waste [5,6]. Moreover, using waste as process feedstock just perfectly dovetails Circular Economy concept that promotes a closing loop economic and production model with the aim to migrate to a more balanced economy and society, thus encouraging the most efficient use of products and reuse of residual sources [7]. Specifically, using Municipal Solid Waste or Refuse Derived Fuel (a MSW derived product) provides a possible strategic way to overcome the shortcomings associated with traditional waste management, a problem that is particularly felt in Italy. Traditional waste treatment, such as landfill and incineration, ensures only a partial energy recovery. On the other hand, thermal treatment like gasification provides a higher flexibility and versatility, producing syngas that could be used both for energy and chemicals production. Besides, a recent study demonstrated that chemical production from waste is as better alternative to power generation as regards the impact of emissions and their effects on climate change [8]. In particular, when compared to energy production, methanol production from waste offers other economic and technical advantages. First, methanol is easier to store and distribute than energy, which is subject to fluctuating demand and not troublesome storage, thus setting plant operating conditions free from market trends. Methanol is a more flexible product, owing to its growing importance in different markets, both as raw material for various chemical manufacturers, such as DME, formaldehyde or acid acetic, and as transport fuel (especially since above cited UE directive incentives). The Waste-to-Methanol (WtM) process analyzed in this paper is a four step process: gasification of waste to produce raw syngas, cleaning system of raw syngas, syngas conditioning unit, methanol from syngas synthesis. The gasification unit, that constitutes the main focus of this study is the same employed in Malagrotta Waste-to-energy (WtE) plant. This unit is a high temperature gasifying and direct melting furnace system supplied by OESA s.r.l. The reactor of this system is directly able to gasify and melt the feeding waste, which has a high ash content, thus allowing for the simultaneous production of a slag inert and syngas streams. The gasifier works close to atmospheric pressure and at high temperature, which is maintained thanks to the exothermic combustion reaction. Indeed, high temperature condition, (1100 °C reached at the top of the reactor) and hot-syngas quench technology that freezes the composition lead to avoid dioxin formation [9], thus reducing the environmental impact of the overall plant. The operating conditions of the gasifier are usually appropriate for the production of syngas for combustion and energy generation. It must be considered that, methanol synthesis requires strict ranges for the syngas composition with a high H2 content, in particular, a Methanol Module ($MM=((H_2-CO_2)/(CO_2+CO))$) near to 2 and a ratio $CO_2/(CO+CO_2)$ between 0.2 and 0.5. In a WtM plant, besides, syngas must be devoid of compounds that are inert in methanol synthesis, such as methane, nitrogen or argon. For these reasons, a conditioning unit is required to obtain a syngas composition suitable for methanol production. On the other hand, a tailored design of the gasification unit can result in a syngas with a better composition for methanol synthesis thus reducing the conditioning efforts. In this work, a simulation of the gasification unit with Aspen tool has been developed to evaluate the syngas composition as a function of the feedstock charcteristics and thus methanol yield. Indeed, municipal solid waste is a heterogeneous mixture of several wastes types such as plastic, wood, textiles, paper, organic residue and inert materials pre-treated using mechanic and biological processes to produce Refuse Derived Fuel (RDF) with a mainly variable chemical composition (C to H ratio, C to O ratio, moisture, ash and combustible content) and heating value. Hence, in this paper, we propose a preliminary study considering several feedstock compositions, i.e. with different ash and moisture content. Accordingly, we represent different gasification yield variables through ternary diagrams, adapting to our purpose a well-known approach that was suggested first by Grout [10] for coal characterization and conveniently modified by several authors for gasification studies [11,12]. The obtained results validate with experimental productive data from Malagrotta's plant, can be useful for a more suitable plant design and to identify strategies for improving the syngas quality and methanol yield.

. This work has been structured as follows. An analysis of the reasonable variability of RDF composition has been carried out by considering some experimental data and LHV of available waste, which range from 14 MJ/Kg to 18 MJ/Kg. A simulation Aspen tool, able to model gasification reactor and successive cooling and cleaning syngas equipment, has been developed. A parametric study through sensitive analysis tool has been taken considering as parameters ash and moisture content of RDF. On the basis of the results presented, the effect of a feeding drying treatment has been analyzed. For the sake of completeness, methanol yields - estimated through syngas composition - have been calculated for different waste LHV.

2. Feed characterization

RDF can be mainly characterized by its content in ash, moisture and combustible fraction (CHO), by its elemental chemical composition (weight fraction of C, H, O, Cl, N, and S) and, eventually by its lower heating value (LHV). Here it has been considered RDF with a LHV between 14-18 MJ/Kg. As reported in Table 1, CHO is usually in the range of 50-80% by weight of RDF; its elemental composition is largely variable due to the different nature of materials present in the waste (plastic, paper, textile, wood). A characterization study [13], on a large number of RDF samples, shows that Carbon to Hydrogen ratio is almost constant varying between 6,8 and 8,2, while the Carbon to Oxygen ratio, which has a major impact on RDF LHV, is more variable. The ranges of C, H and O contents in the combustible fraction are represented in Fig.1a, together with the values of the LHV of a RDF with a moisture content of 0.15 and ash content of 0.2. The LHV is evaluated from the empirical relation of higher heating value (HHV), as function of dry basis weight fractions [14]:

$$\text{HHV [MJ/Kg]} = 0.3417\ \text{C} + 1.3221\ \text{H} + 0.1232\ \text{S} - 0.1198\ (\text{O} + \text{N}) - 0.0153\ \text{Ash}$$

A constant content of Cl, N, and S has been considered as reported in Table 1. The plot underlines that C/H ratio has a negligible effect on the LHV value, while C/O ratio equal to 2 can be considered a mean value for a RDF with LHV in the range of 14-18 MJ/Kg. Fig. 1b reports the LHV values of RDF with different CHO, moisture and ash content with C/H=7.5 and C/O=2. In the figure, the region with LHV from 14 to 18 MJ/Kg is highlighted; all point of this region represent a reasonable feeding compositions of RDF interesting for the aim of this work. The plot shows that LHV is mainly dependent on CHO fraction.

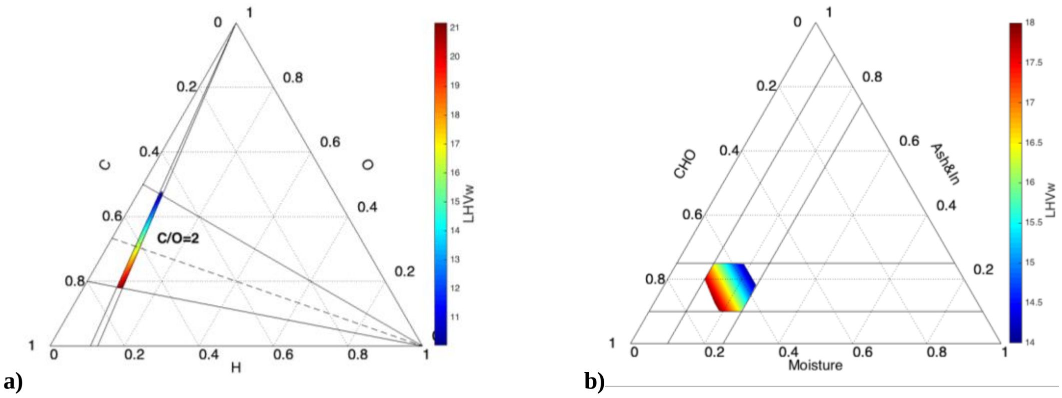

Figure 1 Ternary Diagram for LHV as a function of the composition of waste (a) in terms of elemental composition of combustible fraction (moisture 15% and ash 20% content by weigth) (b) in terms of the ash, moisture and combustible fractions (elemental composition of the combustible fraction C/H=7.5 and C/O=2)

Table 1 - RDF composition (mass fraction)				
CHO	50-80%	C/H=7.5	C	40-55%
			H	5-7.5%
		C/O=2	O	20-27.5%
Ash&In	10-25%	Cl=0.75%		
		S=0.15%		
		N=1%		
		Ash=(Ash&In-Cl-S-N)	SiO_2=35.79%	
			CaO=35.89%	
			Al_2O_3=13.32%	
			Fe_2O_3=15.00%	
MOI	10-25%			

3. Gasification unit

In order to evaluate the composition of syngas obtained from RDF gasification, a model of gasification unit was implemented using Aspen Plus simulation environment. The following assumptions of the modelling have been considered:

- Steady-state process;
- Kinetic-free model: time residence is considered long enough to ensure chemical equilibrium; Since a lot of reactions occur in gasification (Table 2) reaction model are used assuming that components H_2, CO, CO_2, H_2O, H_2S, COS, HCl, N_2, NO_2 can be produced;
- Negligible carbon content in solid slag residue;
- Ash is inert, not participating in chemical reactions;
- Tar and heavy product are not considered as possible equilibrium product at the reactor temperature [15];

Most of these assumptions have been considerate on the basis of experimental data and operative condition supplied by Malagrotta plant.

Table 2 – Gasification Reactions.

Reaction	Reaction name
$C + O_2 \Leftrightarrow CO_2$	Carbon Combustion
$C + CO_2 \Leftrightarrow 2CO$	Boudouard
$C + H_2O \Leftrightarrow CO + H_2$	Steam gasification
$C + 2H_2 \Leftrightarrow CH_4$	Methanation
$CO + H_2O \Leftrightarrow CO_2 + H_2$	Water gas shift
$CO + S \Leftrightarrow COS$	COS formation
$N_2 + 2O_2 \Leftrightarrow 2NO_2$	NO_2 formation
$H_2 + S \Leftrightarrow H_2S$	H_2S formation

3.2. Physical property method

IDEAL thermodynamic method has been used to estimate physical properties of the conventional component. According for low operating pressure [16]. Different methods have been compared and are consistent with each other. The RDF can be introduced into the simulation environment as a *non-conventional component* and HCOALGEN property model has been chosen to evaluate its physics properties. As names suggested, this model was created for modelling coal but is normally used in several references concerning biomass or waste Aspen simulations [17,18]. On the basis of chemical composition and heating value, the model allows to determinate ΔH of formation and heat capacity of the waste. Finally, according to the experimental analysis of slag inert, Ash has been assumed consist of SiO_2, CaO, Al_2O_3 and Fe_2O_3, with a weight fraction composition indicated in Table 1. Ash thermodynamic behaviour has been derived from a method expressed by Mills [19].

3.3. Simulation model

Gasifier has been modelled using four Aspen Plus reactor blocks. RDF, introduced as a non-conventional component, is converted into conventional components in the RYIELD reactor block, to reproduce the decomposition step and to allow the successive steps of Gibbs energy minimization. The yield distribution has been specified directly as a function of feed chemical composition. Then gasification reactions are simulated in three Gibbs reactors (RGIBBS blocks: RG1-RG2-RG3), these reactor blocks return equilibrium composition by minimising Gibbs free energy. The blocks represent the three different zone of the gasifier, depicted in figure 2:

- The melting zone (RG1), where temperature is maintained near to 1600 °C, also helped by methane combustion. The temperature is controlled by manipulating oxygen flow rate;
- The gasification zone (RG2) where gasification reactions continue without further oxidant agent introduction; here the temperature is between 600-800°C;
- On the top, the stabilisation zone (RG3) where other burners introduce a further flow of oxygen and methane in order to reach 1100°C.

The streams paths, modelling the feed subdivision of the reactor, are depicted on the right side of figure 2. External heat streams, correspond to ΔH of RYIELD block, are introduced in RG1 and RG2. In this way suggested in several work dealing with biomass gasification, energy required to break chemical bonds are inclueded into heat balance [20,21]. As the output of the third Gibbs reactor we obtain raw syngas, which is soon cooled in the quench equipment, simulate by flash block. The same type block has been used in order to simulate the successive cleaning gas equipment including acid and alkaline scrubbers. In the simulation a fixed RDF flow rate of 10 ton/h is considered while its composition is varied in the range discussed in section 2.

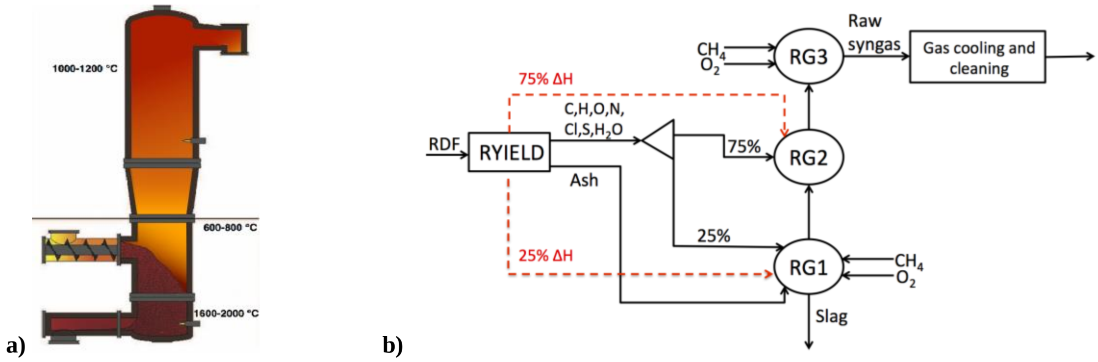

Figure 2 Gasification reactor (a) and scheme of the process modelling in ASPEN PLUS simulation environment (b)

4. Results and Discussions

As well as it has been present in the previous section, a spread of reasonable compositions has been considered as feed of the simulated gasification unit. In particular, RDF some performance parameters, like for LHV, are considered and reported in ternary diagram. In details we report the syngas composition (H_2%, CO%, CO_2%, H_2O%) in Fig.3, while Fig.4 shows the syngas yield (ratio between the syngas mass and the mass of RDF), O_2 consumption referred to combustible waste content and gasification efficiency defined as the ratio between the lower heating value of the produced syngas and the lower heating value of the fuel (RDF and methane) used in the gasifier. These ternary diagrams underline correlation between input and output and lead to evidence waste contents (CHO, Ash&In, Moisture) influences on considered performance parameters.

4.1. Model validation

The results can be compared with productive ranges of Malagrotta gasification unit, correspondent to long enough production period in which RDF feedstock changes its composition. Syngas experimental data present a higher N_2 value than the expected one. This anomaly has been considered caused by the infiltration of N_2 used for inertization of the feeding introduction system. Being N_2 practically inert (NO_2 formation is negligible), we directly enhance N_2% value in simulated syngas and proportionally rescale the other compounds percentages. The confronting ranges are presented in Table 4. A satisfactory agreement between simulation results and Malagrotta's data is obtained, thus validating our assumption and simulation model (see Table 3).

Table 3. Syngas composition range

	Malagrotta	Simulation
H_2%	36-40	36,8-38,4
CO%	37-43	37,2-43,9
CO_2%	8-16	9,0-15,5
H_2O%	5-8	5,4-6,2

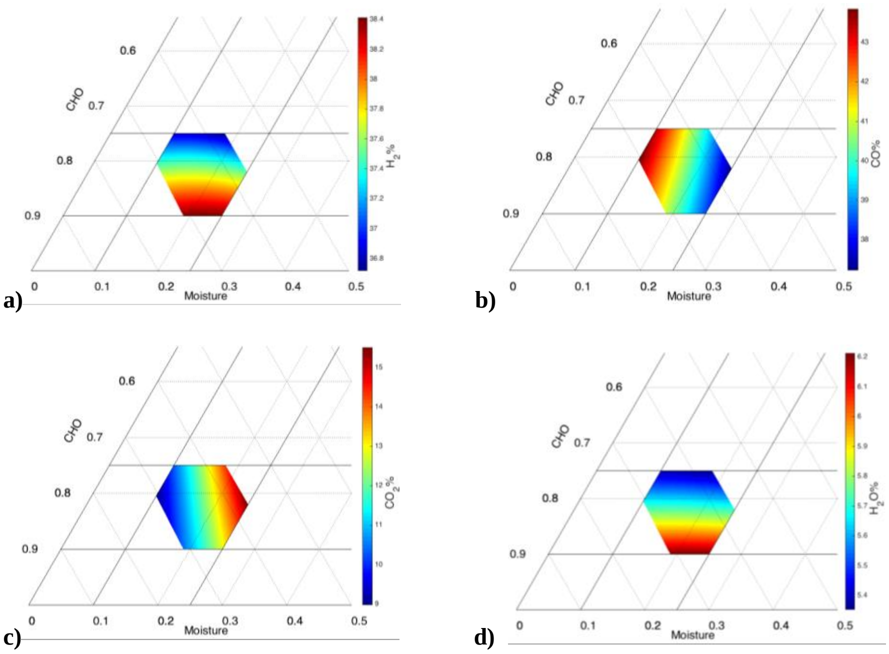

Figure 3 Effect of combustible fraction, moisture and ash contents of RDF on the syngas composition

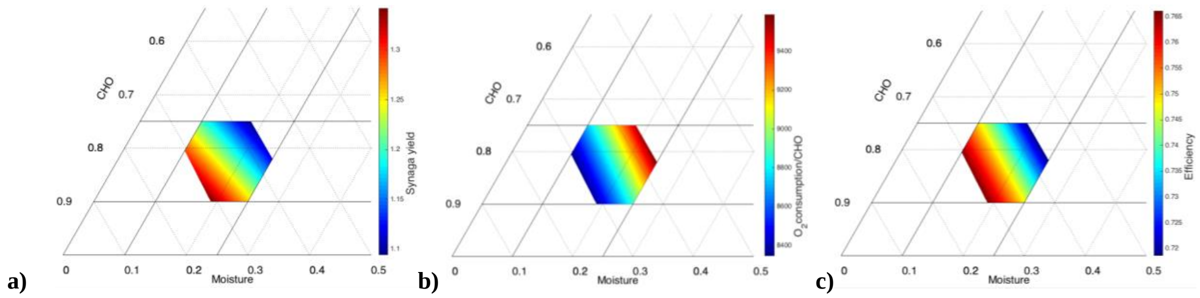

Figure 4 Effect of combustible fraction, moisture and ash contents of RDF on syngas yield, O_2 consumption and energetic efficiency of the gasification

4.2. Effect of Moisture and Ash contents on syngas composition

Effect of ash and moisture on LHV, as previously underlined, is comparable. Indeed LHV is almost linearly dependent on combustible content of waste, as Figure 1 shows. Considering syngas composition and performance parameters the presence of ash or moisture in the waste gives respectively specific contributions, as can be gathered from Figure 3-4. An increase in the ash content has a negative influence on the gasifier performance, with a decrease in the H_2 and CO content and an increase in the CO_2 content of the syngas and an increase in the gasification agent consumption. That behaviour is due to the increment of melting heat that is recovered from strongly exothermic reactions, i.e. H_2 and C combustion, thus as a consequence, the system reclaims a higher oxygen amount and produce more CO_2. Instead, moisture content increase has a double effect on syngas composition. The first one is similar to that of ash content (indeed also an increase in water content results in an increase of the heat requirement, both as sensible and latent heat). On the other hand, moisture is not an inert component as ash, but it takes part in equilibrium reaction of different reactions (steam gasification and water gas shift) in Table 2. In conclusion, we can say that H_2% is mostly sensitive to ash content instead CO%, CO_2% and also oxygen consumption is more sensitive to moisture presence. Efficiency shows similar behaviour to LHV one, being influenced by ash and moisture content with same intensity.

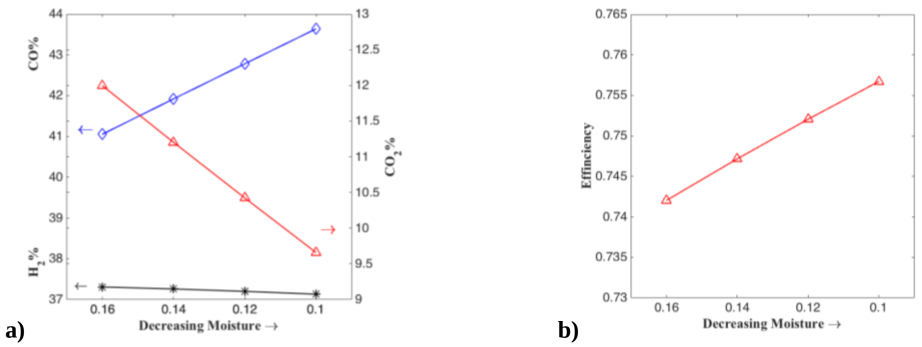

Figure 5 Effect of drying on the syngas composition (a) and on the energetic efficiency (b)

Considering negative effects of moisture content above underlined, we here examined the consequences of a drying treatment of RDF on gasification unit yield. We study one case with initial composition equal to CHO= 0.63, Ash&In= 0.21 and Moisture= 0.16 that is decreased to 0.1. As it is shown in Figure 5 waste drying ensures CO_2% reduction (about 20%) and CO increase (almost 7%). Instead, H_2% maintains its value rather steady. Efficiency is weakly affected increasing about 2%.

4.3. Effect feed variability on methanol production

As previously reported, syngas suitable for methanol production should satisfy the following conditions:

- Methanol module MM=(H_2-CO_2)/(CO+CO_2), near to 2.1;
- CO_2/(CO+CO_2) between 0.2 and 0.5;
- CO_2 content less than 12%.

Therefore, we evaluate MM and the ratio CO_2/(CO+CO_2) for different RDF composition. As reported in Fig.6, MM increases as the ash or moisture content decrease. However, MM is always too much lower than the suitable one. The ratio CO_2/(CO+CO_2) seems to be too low only for the feeding waste with low moisture content. Instead, it never exceeds the upper limit. RDF drying has a weak negative effect on the CO_2/(CO+CO_2) ratio, but it also results in a slight increase in the MM, mainly as a consequence of the CO_2 reduction. Even if a more detailed study about the effects of RDF pre-treatments has to be carried out, the results obtained in this work suggest that is difficult to obtain from gasifier a syngas suitable for methanol production and a syngas conditioning unit has to be included in the process scheme in order to improve syngas characteristics.

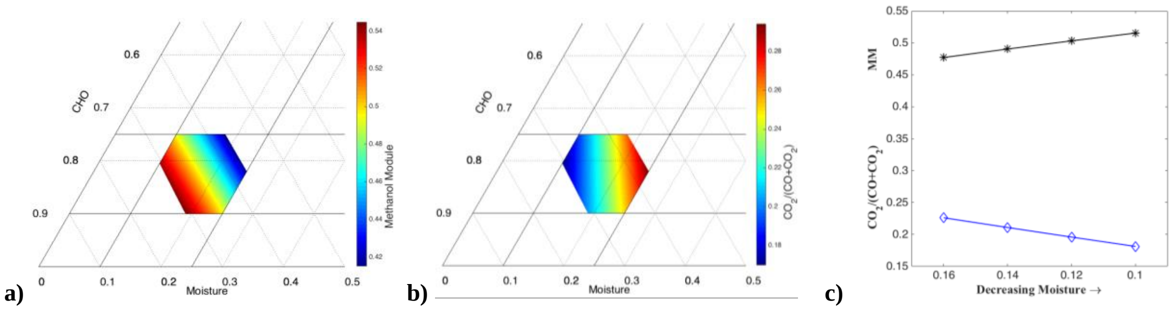

Figure 6 Effect of RDF composition and drying on Methanol Module (a,c) and on the CO_2 content (b,c)

It is worth considering that RDF is usually characterized by its lower heating value, both in legislative and economics terms. Therefore, we attempt to evaluate whether the RDF LHV can be considered as the basic variable for plant design and simulation, even whether there is a wide variability of the waste composition at fixed LHV value. Figs. 7-8 show the syngas composition, the syngas yield, MM and CO_2/(CO+CO_2) ratio as a function of the RDF LHV. Indeed, figures evidence that Methanol Module and Efficiency of gasification unit depends only on the LHV value and are almost independent of waste composition. On the other hand, syngas composition spans quite wide ranges depending on the RDF composition.

Therefore we can conclude that a complete characterization of waste, not only based on its LHV value, is required for a correct design or simulation of the gasifier unit.

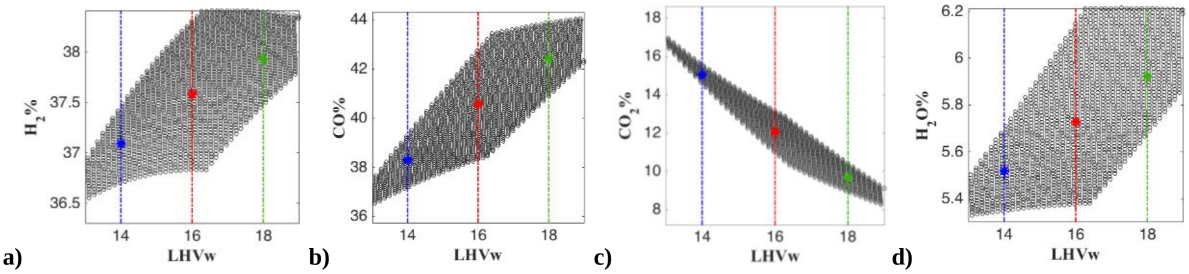

Figure 7 Syngas composition as a function of LHV_{RDF}

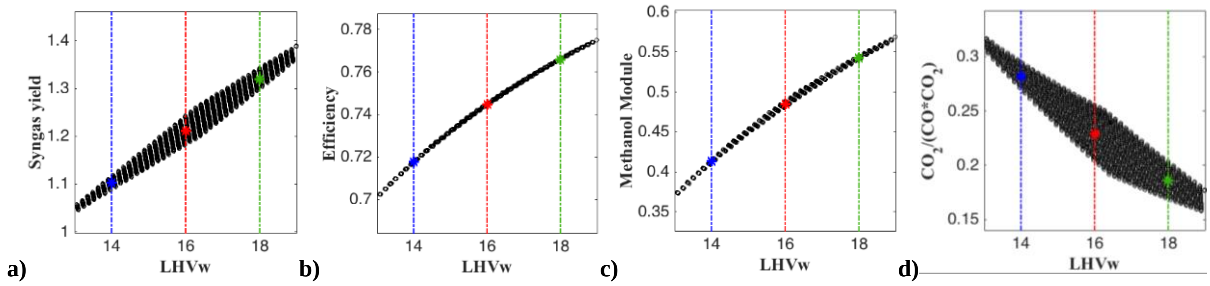

Figure 8 Syngas yield (a) Efficiency (b), Methanol Module (c) and CO2/(CO2+CO) ratio (d) as a function of LHV_{RDF}

To complete our study, we also present some preliminary results of the influence of feedstock on overall methanol production. In particular, we consider three RDF feedstock with 14, 16, 18 MJ/kg respectively and, for each feedstock, a mean syngas composition obtained from the previous simulation and reported in Fig.7. Then average compositions and flow rates are introduced in a simulation model of methanol production from syngas, including both syngas conditioning and methanol synthesis sections. The results of methanol yield are presented in Table 5 in terms of Methanol to waste ratio (kg of MeOH/kg of RDF fed to the gasifier) or efficiency (ratio between the heating value of produced methanol and the heating value of RDF and CH_4 used in the gasifier).

Results show that the higher the RDF LHV the higher both methanol yield and efficiency.

Table 5. Methanol yield

LHV_{Rdf}	**14**	**16**	**18**
H_2%	37,09	37,58	37,93
CO%	38,29	40,57	42,42
CO_2%	15,04	12,06	9,67
H2O%	5,52	5,73	5,92
MM	0,414	0,485	0,542
$\frac{CO_2}{(CO + CO_2)}$	0,282	0,229	0,186
Syngas [Kmol/hr]	562	637	712
CH_3OH [Kmol/hr]	130	157	180
CH_3OH/Rdf	0,416	0,501	0,577
η_{MeOH}	0,540	0,573	0,589

5. Conclusion

In this work, a preliminary analysis aimed to evaluate the feasibility of a Waste-to-Methanol process has been carried out, using ASPEN PLUS as simulation tool. The simulation of the gasification unit suggests that the syngas composition is strongly dependent on the characteristic of RDF used, described in terms of ash, moisture and combustible fraction content, elemental composition or heating value. The range of syngas composition obtained from the simulation is consistent with the composition data of a full-scale gasification unit (Malagrotta, Rome) collected during a long operation period. In any case, it seems that, in the currently employed operating condition of the gasifier, the syngas obtained cannot be directly used for methanol production, but a conditioning step is required. On the other hand, this result also suggests further work aimed to recognize new design criteria, operating conditions or control strategies to improve the quality of the syngas, tailored for methanol production. In addition, it is advisable to compare the simulation results with a wider set of data.

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