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SYNGAS AS A VIABLE SOLUTION FOR LIQUID WASTE CAPITALIZATION

BY

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Abstract. The use of finite resources for obtaining syngas and the accumulation of large amounts of liquid wastes give place to discussions and researches about new technologies and approaches to waste capitalization. Gasification is gaining attention as an efficient process for the conversion of waste. This paper highlights the advantages of using liquid waste as a feedstock for the gasification process. Syngas can be converted to numerous valuable chemical compounds, to olefins by Fischer-Tropsch synthesis in the presence of catalysts, to methanol or to dimethyl ether by direct synthesis and indirect synthesis by methanol dehydration in the same plant. Ammonia is another important chemical compound obtain from syngas. Hydrogen can be separated from hydrogen rich syngas and is a priced fuel of this days. The synthesis gas can be also utilized directly as fuel in the stationary engines of heavy equipment from manufacturing industries and for power plants or as a subsidiary for diesel fuels in transportation.

Keywords: liquid waste, syngas, gasification, fuel, clean energy.

1. Introduction

Two main challenges of this century are represented by the continuous increase in energy demand and the high amount of waste produced by human

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society. If these two problems are treated as separate matters this only deepens the situation, but a simultaneous approach can give a better understanding of the problem and a faster way to the development of viable solutions (Lam *et al*., 2016).

Hazardous liquid waste is less studied than biomass waste or municipal waste, regardless of the difficulties it imposes from manipulation to disposal and environmental pollution (Konur, 2021). This type of waste includes wastewaters, water miscible wastes and oil wastes. Some examples are: medical waste, pharmaceutical waste as out of date parenteral solutions or sirups, cosmetical products, other types of liquid wastes are given by cleaning products, and also petroleum waste from plants, oil lubricants from automotive industry or heavy equipments, cutting oils, antifreeze solution and many others. The majority of this liquid wastes come in mixtures that are hard to recycle and usually they are transported to landfill sites or they are incinerated.

Syngas or synthesis gas is already obtained from an established technology from natural gas (Pei *et al*., 2016) or coal (Lu *et al*., 2020; Masudi *et al*., 2020) in the gasification process. New studies and prototype plants are introducing biomass (Indrawan *et al*., 2020) or a combination of biomass and coal (Wei *et al*., 2021; Bahadar *et al*., 2022) or biomass and natural gas (Nakyai and Saebea, 2019; Qin *et al*., 2021) as feedstock for syngas production. Fewer studies approach the idea of using wastes as plastics (Dogu *et al*., 2021; Ciuffi *et al*., 2020), waste tires (Kaur *et al*., 2021; Jahirul *et al*., 2021), municipal waste (Chanthakett *et al*., 2021) or waste oils (Podlesniy *et al*., 2021) as renewable resources.

Syngas is a mixture of gases, with H_2 and CO as main components. CO_2, light hydrocarbons, CH_4, C_2H_4 can also be found in small amounts (Benalcázar *et al*., 2022). Depending on the feedstock provenience and gasification conditions the syngas can contain traces of heavy metals, sulphur compounds (H_2S, sulphur oxides), nitrogen oxides, and sometimes some toxic compounds like polychlorinated biphenyls (Kuo *et al*., 2021).

This paper reviews the methods of valorification of liquid waste with the new technology of plasma gasification with the production of synthesis gas.

2. Current methods for the transformation of hazardous liquids into synthesis gas

The classical gasification process consists of several stages (Fiore *et al*., 2020):

– drying of the raw material at temperatures below 100°C;

– pyrolysis, the stage in which heavy volatile compounds are released at a temperature of about 240°C and tar and carbon are formed;

– combustion, when water, CO_2 and a large amount of energy are released;

– tar cracking involves molecular breakdown into simpler compounds;
– reduction with the formation of hydrogen and CO in water and CO_2.

In the plasma gasification process, the pyrolysis and combustion steps are accelerated due to high temperatures of the plasma flame, the gases that form inside the reactor, the initial CO_2 produced, acts as a catalyst for the following reactions and also the amount of oxygen introduced in the system is controlled to obtain a partial oxidation. Because of this, the concentration of toxic products (furans, dioxin, polychlorinated biphenyls) is reduced or even avoided at the reactor temperatures. Some chemical compounds, like dioxins can form in the gas cooling phase, depending on the composition of the waste transformed in the process and can be neutralized. In order to prevent the negative effects of these compounds, an important step is to return the contaminated synthesis gas that is obtained in the previous stages of gasification process to the plasma flame as a means of controlling the composition of the synthesis gas and the formation of residual products.

The conditions followed in the gasification process, for optimization and reproducibility, are (Siwal *et al*., 2020):

– gasification temperatures
– operating pressure of the system
– gasifier configuration
– the residence time of the material to be gasified inside the reactor and also the retention time of the gases formed.
– surface speed of gases
– flow rate of the gasification medium
– the use of specific catalysts in some processes

The arrangement of gasifiers significantly affects the quality of the synthesis gas and its chemical composition. Also, the composition of the synthesis gas is directly influenced by the gasifying agent used in the process (Chan *et al*., 2021), so gasification can take place in the presence of the following compounds:

– air
– oxygen
– water in supercritical conditions
– steam
– carbon dioxide

The results of the study by Shahabuddin et al. indicates that the higher the temperature, the higher the yield of synthesis gas, as high temperatures favor the release of volatile substances, endothermic reactions and tar reforming and cracking. For the same reasons, high temperatures also maximize carbon conversion and thermal efficiency of systems. However, the maximum H_2 / CO ratios were observed at intermediate temperatures, when the water-gas exchange reaction is the dominant reaction of the gasification process, supported by an adequate amount of steam. Therefore, the subsequent use of synthesis gases has a direct influence on the conditions to be ensured in the gasification process.

The most important chemical reactions involved in the process of gasification are carbon conversion reactions by partial oxidation (1) and the reactions in the gas phase (2-8).

Combustion reactions:

$$C + \tfrac{1}{2} O_2 \rightarrow CO \tag{1}$$

$$CO + \tfrac{1}{2} O_2 \rightarrow CO_2 \tag{2}$$

$$H_2 + \tfrac{1}{2} O_2 \rightarrow H_2O \tag{3}$$

Other important reactions present in the gasification process:

$$C + H_2O \rightleftharpoons CO + H_2 \text{ Water Gas Reaction} \tag{4}$$

$$C + CO_2 \rightleftharpoons 2CO \text{ Boudouard Reaction} \tag{5}$$

$$C + 2H_2 \rightleftharpoons CH_4 \text{ Methanation Reaction} \tag{6}$$

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \text{ Water Gas Shift Reaction} \tag{7}$$

$$CH_4 + H_2O \rightleftharpoons CO_2 + 3H_2 \text{ Steam Methane Reforming Reaction} \tag{8}$$

Some properties of synthesis gas that influence the quality and determine the valorification methods of the gas, are as follows: H_2 / CO ratio, presence of metal compounds, nitrogen content, methane content, CO_2 content.

Syngas properties that influence the combustion process in internal combustion engines are: flammability limits, laminar velocity of the flame, calorific value, energy efficiency resulting from syngas.

Table 1

The influence of syngas valorification method on gasification parameters

Chemical compounds	**Syngas parameters**		
	H_2:CO ratio	CO_2 content	Sulphur content
Olefins	2	-	-
Methanol	2.0-2.25	2-10% vol (promoter)	H_2S content < 1.6 ppm (catalyst deactivation)
Aldehydes	1	-	-
Dimethyl ether *(DME)*	<1 (direct synthesis)	avoids catalytic overheating; decreases DME yield	-
NH_3	CO elimination (catalyst deactivation)	<1000ppmv	<2ppmv

Table 1 summarizes some of the properties the syngas must have in order to be introduced in the above-mentioned chemical processes. The molar fraction of the H_2 and CO mixture varies from 0.4 to 1.0 for the synthesis gas resulting from gasification. CO-rich synthesis gas describes a different combustion behavior compared to H_2-rich synthesis gas.

Low H_2 synthesis gas is preferred for better internal combustion engine performance, and an H_2 / CO > 1 ratio is often used in chemical syntheses.

In hydrogen plasma gasification, the plasma temperature is very high, over 10,000℃, the synthesis gas produced is of high purity, with high calorific value and is used for cogeneration equipment for energy production.

By controlling the chemical reactions in the reactor, chemical compounds are obtained which will then function as raw materials for various syntheses, and the gases emitted into the atmosphere have a minimal impact on the environment.

According to Wibowo *et al*., in the methanol synthesis the optimal H_2/CO ratio is between 2.1-2.25, the accepted CO_2 content lays in the range of 2-10 vol% for methanol production, than the maximum is calculated in ppmv for ammonia and hydrogen production (Wibowo *et al*., 2021). H_2S content must be below 2 ppmv for methanol and ammonia and the CO concentration below 100 ppmv for ammonia and hydrogen production.

3. Synthesis gas analysis methods

The methods for the analysis of syngas can be classified in 2 groups: in situ analysis and the analysis on samples from the gasification process.

For the in-situ analysis of the gases resulted from the gasification of liquid waste two equipment can give accurate information on quality and composition. First one is the *Testo 350 - Analysis unit for flue gas and emission analyzer*. The analysis unit is equipped with an O_2 sensor and additional gas sensors CO, CO low, NO, NO low, NO_2, SO_2, H_2S, C_xH_y and CO_2. The accuracy of the device is ± 1°C at temperatures between 0 and +1760°C, resolution 0.1°C between 0°C and +1760°C. The device offers the possibility of automatic measurement for a long period of time. Flue gas and thermal process analyzes may be performed. The analysis probe allows the sampling of gases with temperatures up to 1800°C. The second equipment is represented by the *Thermoregulation control system with fuzzy PID control.* Through it, the online monitoring of the plasma gasification process is performed. LabVIEW software, through sensors for temperature, flow, pressure and emission monitoring equipment, controls in real time the reactor module for hydrogen plasma gasification and plasma subassembly for heat treatment, cooling and cleaning of synthesis gases obtained from waste (Cucoș *et al*., 2021). This system also allows the creation of a real-time database with the physico-chemical characteristics of the processed waste, the technological characteristics of the terminal (temperature, flow, pressure, flow, current/voltage) and the chemical composition and nature of the emissions produced.

The syngas samples obtained from the gasification of liquid waste can be analysed using the following methods and equipments:

The combined TG / DTA / DSC *thermogravimetric analysis system* with FTIR is used to identify the compounds resulting from each stage of thermal decomposition of the raw material subjected to the conversion process (Fig. 1a). The thermogravimetric analyzer allows the qualitative analysis of the main products resulting from the thermal degradation process of the analyzed samples, but also quantitative, for at least five compounds for which the calibration gas cylinders are provided. The compounds for quantitative analysis are: carbon dioxide, carbon monoxide, hydrogen, formic aldehyde, methane.

The *gas chromatograph* (Fig. 1a) is an equipment that allows the additional analysis of the non-condensable products from the gas flow resulting from the plasma gasifier and respectively in the measurement points located before and after the gas purification stages. The equipment separates and quantitatively determines the following gases: hydrocarbons C1-C_6, SO_2, H_2S, CO, CO_2, O_2, N_2, NO_x. The integrated system based on gas-chromatographic analysis contains an interface and a specialized software meant to control the installation by operating at least two proportional solenoid valves mounted on the installation pipes, so as to regulate the fluid flows, depending on the concentration of the analyzed components. during research).

Atomic absorption spectrometer is a fully automatic equipment for analyzing the content of heavy metals in the samples to be analyzed and solid products resulting from the processing of raw materials (ash, slag, vitrified materials) using both flame ionization and graphite furnace (Fig. 1b).

(a) (b)

Fig. 1 − Netzsch Perseus STA 449 F3 gas chromatograph coupled gas chromatograph analyzer (a), Analytik Jena Zeenit 700 P atomic absorption spectrometer (b).

The multicomponent gas analyzer (Fig. 2a) is intended for applications of continuous monitoring of the concentration of gases of interest, of the multicomponent gas analyzer type: NO_x, SO_x, VOC (on individual components), CO and CO_2. The equipment allows the determination of trace components of gaseous mixtures related to the raw material (waste), respectively to the reaction products obtained after processing.

Stationary flue gas analyzer (Fig. 2b) is a process analyzer designed for the online analysis of the main compounds in the gas flow resulting from the plasma gasifier and the analyzed samples: CH_4, CO, CO_2, H_2 and O_2. The gas mixture is a complex mixture that can be analyzed using several detectors with specificity for one or more gases as follows: for CO, CO_2 and CH_4 gases the analyzer is equipped with a detector (non-dispersive infrared sensor), for H_2 a detector is used thermal conductivity (TCD); for O_2 a paramagnetic detector is used.

(a) (b)

Fig. 2 − Nicolet IS-50 FTIR Gas Analyzer (a), MRU Stationary Flue Gas Analyzer, Model SWG 200-1 (b).

4. Capitalization of synthesis gas

The synthesis gas has multiple capitalization possibilities (Fig. 3): from chemical feedstock in chemical synthesis, alcohols, olefins, aromatic compounds, ammonia, dimethyl ether, to utilization as fuel in internal combustion engines, either alone or by replacing some of the conventional fuel, then to produce power in gas turbines or generators or power plants and even thermal energy.

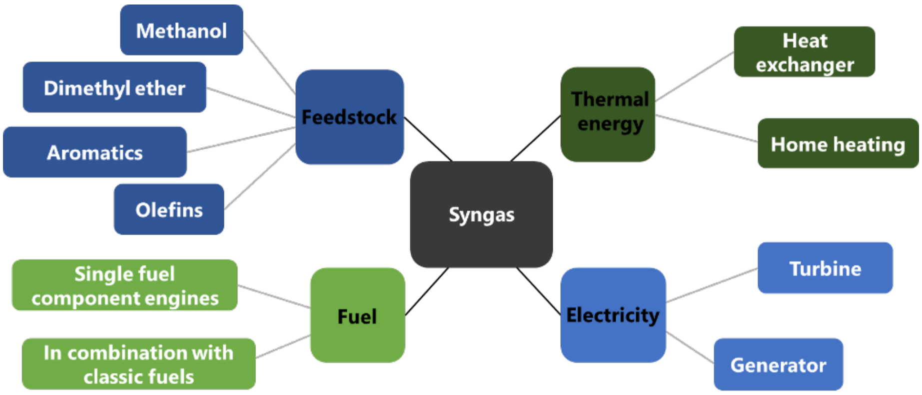

Fig. 3 − Main applications of synthesis gas.

Syngas for the synthesis of olefins

Light olefins, as one of the most important building blocks in the chemical industry, can be produced by Fischer-Tropsch synthesis (FTS) from synthetic gas (Gorshkov *et al*., 2021). High temperature FTS can lead to light paraffins, carbon dioxide, methane and longer chain hydrocarbons. Olefines, which include ethylene, propylene and butylene, are amongst the most important raw materials used in the production of polymers, fibers, solvents and detergents (Yahyazadeh *et al*., 2021).

Ma *et al*. analyzed the addition of sodium to the well-known Fe-Zr catalyst in Fischer-Tropsch synthesis at high temperatures, to obtain olefins from the synthesis gas (Ma *et al*., 2021). The presence of sodium led to a decrease in the H/C ratio on the catalyst surface, which led to increased chain propagation and reduced hydrogenation of light olefins.

Syngas for methanol synthesis

Methanol plays an important role, both for the chemical industry, participating in numerous chemical syntheses (obtaining gasoline, aromatic hydrocarbons, olefins) and for the utility as a fuel in industry and transportation. For the methanol synthesis process, the synthesis gas must first be conditioned to have a certain ratio based on the stoichiometry of the methanol formation reaction of H_2 and CO, equation (9). The optimal ratio calculated by Ribeiro *et al*. (2012) fits in the 2.1 – 2.25 range.

In addition to the synthesis gas ratio, a CO_2 content of about 2 – 10 vol% in the feed gas should also be maintained, it forms spontaneously in the gasification process by interacting with the steam (10), as it acts as a promoter as it is showed in equation (11) (Rauch *et al*., 2014). The sulfur content of the feed gas, usually as H_2S in the synthesis gas, must be kept below 1.6 ppmv to avoid deactivation of the catalyst used in methanol synthesis (Koizumi *et al*., 2004). The same process of methanol synthesis could also be designed to produce dimethyl ether (DME) by dehydrating methanol, equation (12) (Gogate, 2018).

$$CO + 2H_2 \rightleftharpoons CH_3OH \tag{9}$$

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \tag{10}$$

$$CO_2 + 3H_2 \rightleftharpoons CH_3OH + H_2O \tag{11}$$

$$CH_3OH \rightleftharpoons CH_3OCH_3 + H_2O \tag{12}$$

Although studies show that energy efficiency in the conversion of natural gas to methanol (Trimm and Wainwright, 1990) is much higher than other raw materials, the production of methanol, indirectly, from waste, by converting syngas, brings net benefits. On the one hand, this conversion is based on renewable resources, given that used oils or other liquid waste with a high content of hydrocarbons, petroleum waste, are constantly produced in large quantities and a large proportion of existing waste is difficult to be introduced into solvent or component regeneration processes due to increased contamination and high operational costs. On the other hand, the synthesis gas also contributes to the creation of low-cost energy for the steps involved in chemical synthesis.

Syngas for DME synthesis

Dimethyl ether (DME), also called methoxymethane (CH_3OCH_3), is the lowest aliphatic ether, a non-toxic, non-carcinogenic and non-corrosive compound.

DME is considered to be the cleanest fuel for high-efficiency compression ignition and a replacement for diesel fuel, based on the DME self-igniting properties and the high octane number (55 – 60) and low CO, NO_x emissions. It also has physical properties similar to those of liquefied petroleum gas and can therefore be used as an alternative fuel for cooking and heating (Prasad *et al.*, 2008).

The synthesis gas can be converted to dimethyl ether by indirect or direct process. Indirect process, consisting of synthesis gas to methanol and methanol to DME, is depicted in the equations (13)-(15).

$$CO + 2\,H_2 \rightleftharpoons CH_3OH \quad \textit{CO hydrogenation} \tag{13}$$

$$2CH_3OH \rightleftharpoons CH_3OCH_3 + H_2O \quad \textit{CH}_3\textit{OH dehydration} \tag{14}$$

$$H_2O + CO \rightleftharpoons H_2 + CO_2 \quad \textit{WGS} \tag{15}$$

For the direct production of DME, the usual catalyst is a copper-based metal oxide, but a new interesting approach consists in the use of a hybrid catalyst that comprises a solid acid catalyst in addition to the metal oxide.

The direct DME synthesis triggers simultaneously methanol synthesis and in situ dehydration and it is integrated into hybrid catalysts in a single reactor, thus reducing the thermodynamic constraints of methanol synthesis and leading to higher CO conversion and higher selectivity for DME.

The global reaction in the direct synthesis of DME is given by the equation (16).

$$3CO + 3H_2 \rightleftharpoons CH_3OCH_3 + CO_2 \tag{16}$$

Syngas for ammonia synthesis

Ammonia is another important substance for the industry that can be obtained from syngas. Ammonia synthesis is mainly done using the Haber-Bosch process (Fig. 4). Ammonia has extensive uses, from agriculture, as a fertilizer, to the production of chemical compounds, even for the production of pure hydrogen (Lamb *et al.*, 2019). At present, ammonia production used the largest part of the synthesis gas obtained globally, 53% (El-Nagar and Ghanem, 2018).

However, a very small amount of that syngas is obtained from renewable resources, and most of it comes from coal or natural gas processing technologies and this can be improved in the future with the help of the newly described technologies.

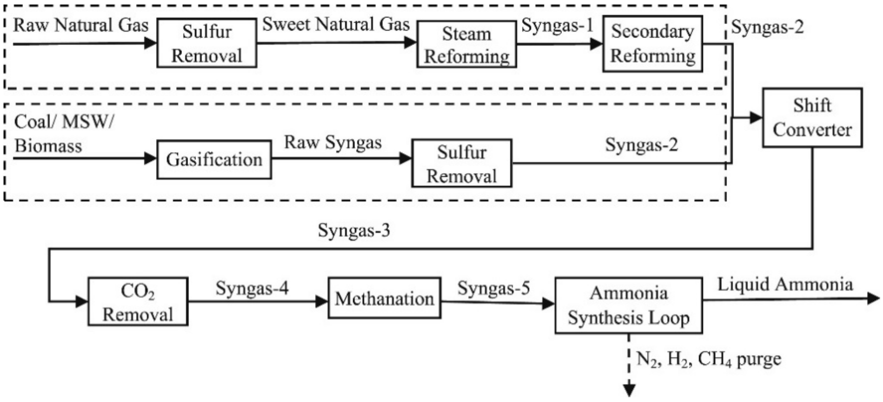

Fig. 4 − Diagram of the ammonia synthesis process (Wibowo *et al.*, 2021).

Syngas for hydrogen production

Hydrogen is considered the best fuel available today because the hydrogen fuel cell has the most efficient conversion of fuel into energy and produces only water as an emission.

Direct hydrogen production from synthetic gas is an interesting process, especially for the advances in supercritical water gasification (SCWG) where a synthesis gas with as high as 50 vol% H_2 is obtain, making it a potential source of hydrogen (Su *et al.*, 2020). Moreover, the SCWG offers the possibility to process wet raw materials, an important issue in waste management.

These processes must meet some specifications of the syngas for use in industrial processes (Ohi *et al.*, 2016): $H_2 \geq 98$ vol%, $CO + CO_2 \leq 10–50$ ppmv, $O_2 \leq 100$ ppmv and inert gases (N_2, Ar, CH_4) <2 vol%. The process of converting natural gas or synthesis gas into hydrogen can be seen in Fig. 5.

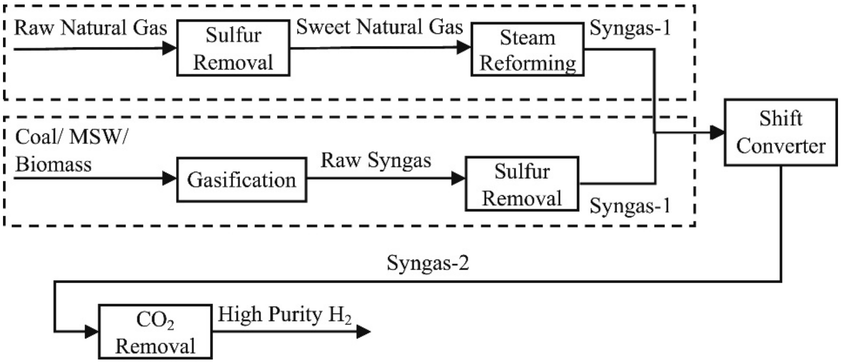

Fig. 5 − Diagram of the hydrogen production process (Wibowo *et al.*, 2021).

Syngas as fuel

The use of synthesis gas in engines can be an advantage in terms of exhaust emissions if the engine's operating conditions are carefully selected. The emissions of pollutants such as sulfur compounds, metals, halogens and ash are generally avoided due to cleaning systems that are able to remove contaminants before being burned in gas engines or turbines. Consequently, the most important emissions are limited to NOx and CO. The amount of CO is also related to the specific composition of the synthesis gas and its CO content, while NOx is related, to the geometry of the engine and its operating parameters.

Different types of synthesis gas can be produced depending on the gasifier type (Umar *et al*., 2021). The synthesis gas derived from the use of steam or oxygen as a gasifying agent has an average heating value of about 10 – 28, while the synthesis gas produced by air gasification is characterized by a heating power of less than 4 – 7 and is generally known as producer gas.

According to the sensitivity analysis of Rosha et al., hydrogen plays a major role in controlling the reactivity of the mixture, while CO has an inhibitory effect, as it increases the ignition delay time by a factor of 2 with the introduction of 50% CO, and a fraction of 10 with a concentration of 90% (Rosha *et al*., 2018).

Conversion of synthesis gas into electricity and heat

Gas turbines and generators are the most popular technologies to generate power from synthetic gas (Santiago *et al*., 2021). Heat exchangers are another option for obtaining heat. Figure 6 shows the efficiency of various methods in terms of generating electricity.

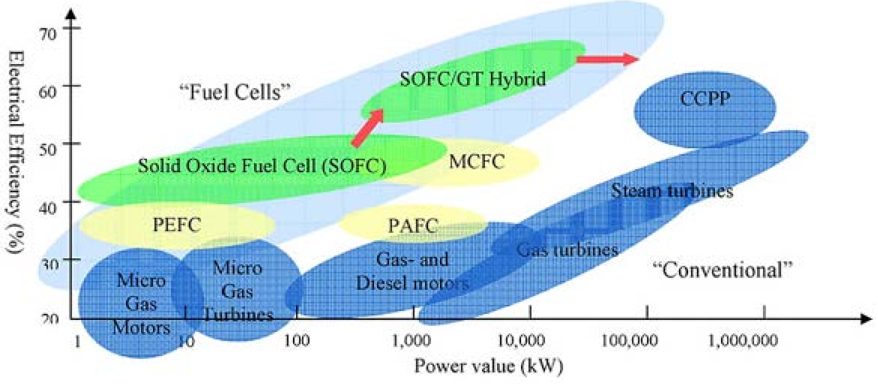

Fig. 6 – Efficiency of several electricity generation routes (Stambouli, 2011).

The electrical efficiency of gas turbines ranges from 20% to 40%, similar to that of conventional systems that use fossil fuel plants, but with a lower power consumption. There are also fuel cell systems with better electrical efficiency at lower power levels, which is giving them a lot of value, but manufacturing issues, limited resources, and high prices for mass production must be researched and resolved.

5. Conclusions

Taking into consideration the varying composition of liquid waste and the numerous syngas qualities that must be achieved, plasma gasification technology provides a viable alternative for converting difficult-to-process liquid wastes.

Advantages of using synthetic gas in internal combustion engines are:

– avoidance of pollutant emissions such as sulfur compounds, halogens, metals and fly ash on behalf of the advanced cleaning systems

– limited NOx and CO emissions

– the use of the exhaust gas recirculation technique, which allows a decrease in peak temperature, which also has an impact on emissions

– minimizing diesel consumption.

In terms of chemical compound synthesis, the synthesis gas also opens up new options. The Fischer-Tropsch method may be used to make light olefins since it is a direct synthesis from the synthesis gas, as opposed to a synthesis from an intermediate like methanol or dimethyl ether, which needs less chemical changes to the synthesis gas. A wide range of wastes can be utilized as input material in the gasification process to produce synthetic gas under these conditions.

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GAZUL DE SINTEZĂ CA SOLUȚIE VIABILĂ PENTRU VALORIFICAREA DEȘEURILOR LICHIDE

(Rezumat)

Utilizarea resurselor epuizabile pentru obținerea gazului de sinteză împreună cu acumularea cantităților foarte mari de deșeuri lichide oferă motivația pentru discuțiile și cercetările legate de noi tehnologii și strategii în ceea ce privește acest tip de deșeuri. Procesul de gazeificare captează din ce în ce mai multă atenție, fiind un proces eficient pentru conversia deșeurilor. Această lucrare subliniază avantajele utilizării deșeurilor lichide ca materie primă în cadrul procesului de gazeificare. Gazul de sinteză poate fi transformat în numeroși compuși chimici valoroși precum olefine, prin sinteze Fischer-Tropsch în prezența catalizatorilor, metanol sau dimetil eter, fie prin sinteză directă, fie prin sinteză indirectă prin deshidratarea metanolului într-un proces continuu. Un alt compus chimic important pentru industria chimică, ce poate fi obținut din singaz este amoniacul. Hidrogenul poate fi separat din singazul bogat în hidrogen și reprezintă un combustibil de viitor. Gazul de sinteză poate fi utilizat, de asemenea, direct drept combustibil în motoare staționare ale unor utilaje grele, în industria energetică sau ca înlocuitor pentru combustibilii derivați de petrol, în transporturi.