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BIOMASS GASIFICATION FOR HYDROGEN PRODUCTION, AN OVERVIEW OF THE PROCESS USING VARIOUS RAW MATERIALS

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Abstract
Climate change, environmental pollution, population growth and overwhelming amounts of waste have led to the need to identify alternative fuels to replace fossil fuels. The gasification process of biomass residues from agriculture and agro-industry represents a suitable source for energy recovery through the production of syngas, including H2, CO and CH4. The gasification processes are carried out in area 500 °C ÷ 1000 °C. Various types of gasifiers were examined, in fixed bed and fluidized bed, following the gasification process and the contribution of specific gasification agents. The oxidation agents used in the biomass gasification process are: air, CO_2, steam, O2. The effect and type of catalysts used were followed from the perspective of the highest possible percentage of hydrogen. This report analyzed the production of syngas, especially hydrogen, through gasification by comparing and analyzing the results of various types of biomass and aimed to bring a globalist approach to the production of hydrogen-rich syngas based on current technologies.

Keywords: agro-residues, gasification, hydrogen, syngas

1. INTRODUCTION

In the last decades, the world energy consumption has grown rapidly (Dudley, 2018). Fossil fuels are used worldwide as the main source of energy. But the irrational and uncontrolled use of fossil fuels produces a series of imbalances such as: environmental (burning fuels produce greenhouse gas emissions – (GHG), energy security (global energy, geopolitical problems) and economic. Greenhouse gases trap heat from the atmosphere and thus the global warming occurs. The global average temperature increased by 1°C. Global temperature increase of more than 1.5 °C leads to the risk of sea level rise, extreme weather events, biodiversity loss and species extinction, food shortages and worsening health for all of humanity. About 75% of greenhouse emissions come from industry: power plants, desalination plants (Rezaei et al., 2017; Naserbegi et al., 2019), and hydrogen production plants that use fossil fuels to provide energy (Aydin and Dincer, 2022; Depren et al., 2022).

It is necessary to develop a sustainable energy system to reduce the use of fossil fuels and reduce environmental pollution. Hydrogen is an inexhaustible source to produce sustainable and green energy because it is abundant on Earth and does not produce greenhouse gas emissions, and for this

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reason it can be a sustainable substitute for fossil fuels (Dawood et al., 2020; Abe et al., 2019; Schlapbach and Züttel 2001; Sinigaglia et al., 2017). The main methods of hydrogen production are: hydrocarbon reforming (Malik et al., 2023; Woo et al., 2023), coal and biomass gas conversion, water splitting by electrolysis, photo-electrolysis (Lee et al., 2019) photobiological production, thermochemical loop water splitting at high temperatures (El-Emam et al., 2020). and hybrid technologies (Temiz and Dincer, 2021; Filippov and Keiko, 2021; Panchenko et al., 2023). Due to these methods of hydrogen production, a series of gaseous impurities appear such as O_2, H_2S, CO_2, N_2, CO, H_2O, CH_4, but also inert gases such as Ar and He, which can be leaked into the air (Sun et al., 2015). The main obstacle in the hydrogen development process is the low density of hydrogen and its storage (Barthélémy et al., 2017).
Biomass, as a source of obtaining hydrogen, represents a very promising potential alternative for pollution-free energy production, being considered the fourth source of energy in the world after oil, coal and natural gas (Missaoui et al., 2017).
Hydrogen it is the most abundant element in the universe, accounting for approximately 75% of all matter, hydrogen is not readily available on Earth in elemental form (Baykara, 2018). But,it can be produced from its compounds found in natural or industrial sources.
Considering the levels of energy consumption in the world and the negative effects of fossil energy sources on the environment, renewable energy sources are ideal for sustainable hydrogen production. Hydrogen can be separated from synthesis gas, (H_2 and CO), obtained from biomass gasification following cleaning reforming and switching processes (Baykara, 2018).
Hydrogen is a clean energy carrier that will play an important role in the coming years to provide an ecological option to meet the growing demand for energy of the world. Intensive research is being done to identify clean routes for the transition to a green hydrogen economy to compete with fossil-based hydrogen production (Arregi et al., 2018). In this case, bio-energy produced from biomass can be directed to generate hydrogen through various thermochemical and biological processes and gasification is the most possible for large-scale hydrogen production (El-Emam and Özcan, 2019).

2. BIOMASS - WASTE WITH HIGH POTENTIAL FOR ENERGY RECOVERY

Biomass can be defined as any organic matter that is available from renewable sources and can be used as a source of energy, such as waste and residues from various fruit or vegetable activities, various types of wood and wood waste, ocean plants, waste animal products and those resulting from the exploitation of animals as well as organic waste from municipalities (Salam et al., 2018).
In general, vegetable biomass is divided into two main categories: woody and non-wood residues. Wood biomass contains wood and forest residues resulting from the processing of forest products, such as needles, cuts, bark, wood sawdust and wood chips.
Non-woody biomass has a lower lignin content than wood biomass, being often classified as waste and can come from a wide range of agricultural processes, animal waste and herbaceous plants such as rice straw (Cao et al., 2018), sewage sludge (Smoliński et al., 2018; Zuo et al., 2022) , palm kernel shell (PKS), cocoa and grape pomace (Teh et al., 2022; González-Vázquez, 2018).
Hemicellulose, cellulose and lignin as main components of wood biomass,Table 1, which together with chicken manure, sawdust as well as different types of municipal or kitchen waste are used as raw material for hydrogen production.

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Table 1. The component elements of various types of biomass (Ren et al., 2019)

Type of biomass	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Other[a] (%)
Softwood	41	24	28	7
Hardwood	39	35	20	7
Wheat straw	40	28	17	15
Pine wood	42.1	17.7	25	-
Birch wood	35.7	25.1	19.3	-
Spruce wood	41.1	20.9	28	-
Sunflower seed hull	18.4	27	-	-
Broiler poultry litter	27	17.8	11.3	20

Biomass selection is important for syngas production during gasification because wood, straw and plant biomass contain more cellulose compared to husks which have a higher lignin content and a high hemicellulose (cellulose) / lignin ratio could produce a high syngas content (Ren, J., et al., 2019).

Kitchen waste can be harmful to the environment if not handled properly, but can be converted into gas as a source for efficient gasification power generation in a updraft gasifier with fluidized bed together with a cyclone separator and a cooler (Cao, W., et al., 2022). It was used an air blower to create flow of air to heated at 400°C in an air preheater.

Biomass with higher moisture content gasified slowly due to the additional heat required to evaporate the water in the biomass before its thermochemical conversion.The dry waste was then ground to a size of 1–1.5 cm to increase its surface area, which helps it to be gasified quickly and evenly.

3. BIOMASS GASIFICATION PROCESS FOR THE DEVELOPMENT OF SYNGAS AND HYDROGEN

3.1.Gasification Process

The gasification process is carried out in the thermal range of 500°C - 1000°C, using air, steam or CO_2 as an oxidizing agent. The biomass is gasified in a pressurized reactor and the generated gas is separated from the inorganic matter and ash. The gas is cooled and separated in the cooling zone below the gasifier (Molino et al., 2016). The water vapor in the fuel gas is condensed and the released heat is used to generate steam. The resulting gas is similar to syngas, consisting in CO, H_2 and CO_2, and can be used to produce synthetic fuels or chemicals through the Fischer-Tropsch process or other synthesis processes. The gasification process is carried out using either fluidized bed reactors or fixed bed reactors. The gasification process is considered one of the best ways to recover energy from biomass by producing syngas, including H_2, CO și CH_4 Figure 1. where a fixed bed reactor with the following components is represented: digester, reactor.condensate, clean cool synthesis gas, gas storage. In gasification processes using fluid bed/circular reactors, a higher air speed is required compared to those using fixed bed reactors.Biomass gasification is an efficient and promising conversion technology. Production of CO_2-free green hydrogen from decarbonised biomass gasification has shown, as an important novelty, a promising potential to deliver high energy conversion (Suryawanshi et al., 2023).

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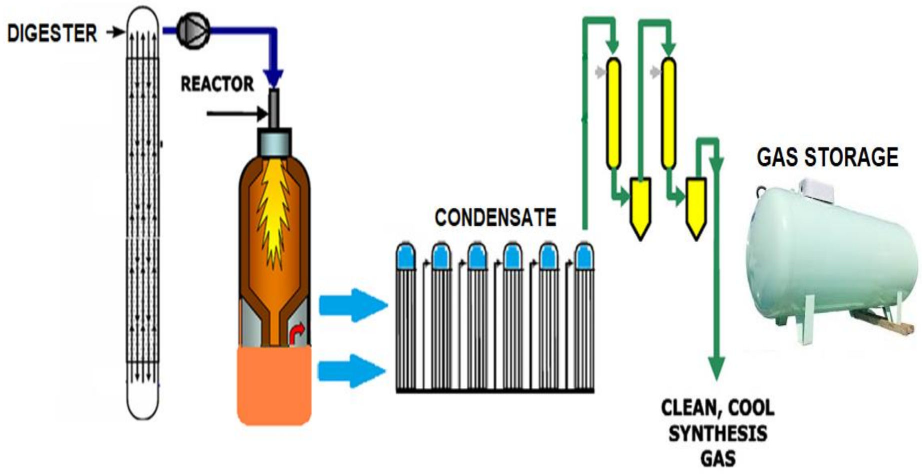

Figure 1. Gasification process (Tezer et al., 2022)

The main types of gasifiers found in literature are gasifiers with a fixed bed, (with upward or downward current), with fluidized bed or (more recently) with plasma, but they can be diversified according to the gasification variables like: oxidation agent, temperature,pressure or transport process (Sansaniwal, S. K., et al., 2017). Fixed bed gasifiers can be classified according to the mode of interaction of either air/oxygen or steam with biomass such as downflow,upflow and crossflow gasifier, Figure 2.

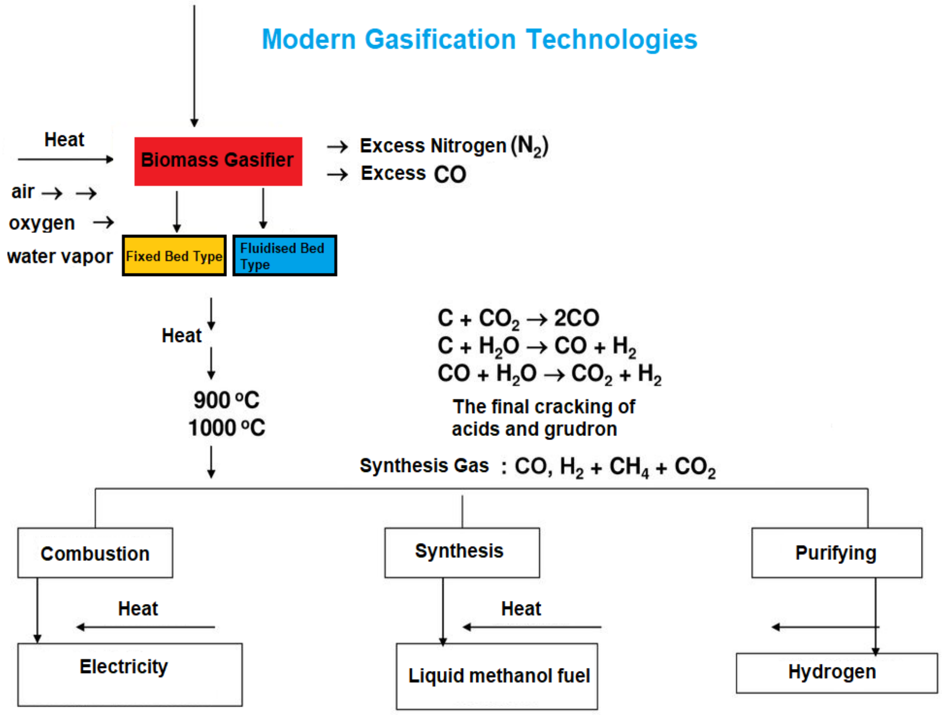

Figure 2. Classification of biomass gasifier (Sansaniwal et al., 2017)

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In the downdraft gasifier, the air flows in a downward direction and interacts with the biomass fuel, which results in co-current movement of waste and gase`s. After pyrolysis and drying the products are forced by oxidation to produce fuel gas. of good quality (Fazil et al., 2022; Vera et al., 2011). In the updraft gasifier, the gasification agent such as air, oxygen and steam are introduced at the bottom to interact with the biomass and fuel gases in the countercurrent direction, and the gas produced in the reduction zone comes out of the reactor at the top (Diboma et al., 2023; Tuan et al., 2023). And in the crossdraft gasifier the biomass enters from the top, the thermochemical reactions take place progressively as the fuel descends into the reactor while the gasification agent enters at high speed from the side near the bottom of the reactor, and the produced gas leaves the reactor. the opposite side.

The fluidized bed gasifier works on the principle that both the fuel and the material in the inert bed behave like a fluid (Motta et al., 2018). One type of fluidized bed gasifier is the bubbling reactor (BFB) which operates at atmospheric pressure and is placed inside an electrically heated furnace. Steam and oxygen are used as gasification agents, which are mixed in the wind box located at the base of the reactor. Sand, silicon or a catalytic bed material, (such as lime, dolomite and olivine) is used as a fluidizing medium to improve the gasification process (Liakakou et al., 2019).

3.2. Syngas production

The main product of biomass gasification is a gaseous mixture consisting of H_2, CO, CO_2 and CH_4. Gasification is a thermochemical process at high temperatures that causes changes in the biomass structure in the presence of a gasification agent, (air, steam, steam and oxygen, air and steam or air enriched with oxygen) resulting in a large amount of gaseous product known as of synthesis gas syngas (Zhang et al., 2023). The varied results of the gasification are due to a reaction sequence:

Biomass decomposition reaction

$CxHyOz + heat \rightarrow Gas + biochar + volatiles(tar) + steam$ (1)

Volatiles secondary cracking reaction

$Volatiles \rightarrow C + CmHn(smaller\ volatiles) + gas$ (2)

Volatiles dry reforming reaction

$CmHn + mCO_2 \rightarrow 2mCO + n_2H_2$ (3)

Volatiles steam reforming reaction

$C_mH_n + mH_2O \rightarrow mCO + (m + \frac{n}{2})H_2$ (4)

Char steam gasification

$C + H_2O \leftrightarrow CO + H_2$ (5)

Methanation reaction

$C + H_2 \rightarrow CH_4$ (6)

Water-gas shift (WGS) reaction

$CO + H_2O \leftrightarrow CO_2 + H_2$ (7)

Boudouard reaction

$C + CO_2 \rightarrow 2CO$ (8)

Steam methane reforming (SMR) reaction

$CH_4 + H_2O \leftrightarrow CO + 3H_2$ (9)

Methane dry reforming (MDR)reaction

$CH_4 + CO_2 \leftrightarrow 2CO + 2H_2$

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In general, biomass contains cellulose, hemicellulose and lignin containing carbon, oxygen and hydrogen, from which syngas is obtained through gasification (Sansaniwal, S. K., et al., 2017).
To obtain syngas, different types of biomass were used: charcoal, wood, sawdust, corn cobs, pine wood cubes, wood shavings, Table 2.

Tabel 2. Composition of gas obtained in gasification with downdrft current from biomass (Sansaniwal et al., 2017)

Biomass	**Gas composition (vol %)**				
	CO	**H_2**	**CH_4**	**CO_2**	**N_2**
Charcoal	28-31	5-10	1-2	1-2	55-60
Wood	17–22	16–20	2–3	10–15	55–50
Sawdust	19.48	18.89	3.96	–	–
Wood chips	26.5	7.0	2.0	–	–
Corn cobs	18.6	16.5	6.4	–	–
Pine wood blocks	25.53	28.93	6.82	–	–

Syngas resulting from the gasification of biomass, is a mixture of carbon monoxide, hydrogen, carbon dioxide and a smaller amount of methane (Cerone and Zimbardi, 2018). The gas composition is varied due to changes in the following factors:composition of feedstock, gasification agents or catalyst selection (Fatema et al., 2022).
Syngas can be used in many industries, such as the chemical industry, the fuel industry and power plants (Chanthakett et al., 2022).
The production of hydrogen from syngas as a result of biomass gasification has many advantages as a secondary source of renewable energy as a clean fuel for vehicles, because it does not release carbon or sulfur emissions during combustion. As a green energy, hydrogen has a great potential to reduce the dependence on fossil fuels and, implicitly, environmental pollution, and thus, the production of hydrogen from renewable sources will reduce this pollution, which is an important factor in global warming.

3.3. Hydrogen production

The following types of biomass were used for the production of hydrogen: chicken manure, kitchen wastes, Table 3.

Tabel 3. Obtaining hydrogen by gasification of various biomass sources

Type of Biomass	**Type of Gasification**	**Operating Conditions**	**H_2 Yield**
Kitchen waste (Liu, J., et al., 2022)	Supercritical water	- 480 °C without catalyst - with Ni/γAl_2O_3. catalyst	563.43 mol / L 673.13 mol/L
Chicken manure (Hussein, M., et al., 2022; Cao, W., et al., 2022)	Steam gasification	T = 1000 °C	0.1 g/min
	Supercritical water	T = 620 °C	22.47 mol / kg

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Most of the biomass gasification reactors evaluated have a hydrogen capacity of 390 MW (corresponding to 100,000 Nm3/hydrogen with purity above 99.95% vol.) with a carbon capture rate of 90%.
The variations of the gasification agents cause variations in the percentage values of the gasification products which are CO,CO_2,CH_4,H_2,carbon and bio-oil. Air gasification is the simplest and most widely used because of the low cost of the gasifying agent and produces a gas with a low calorific value, generally 4 to 7 MJ/Nm^3, while O_2 and steam gasification produces a synthesis gas with a calorific value between 10÷18 MJ/Nm^3. On the other hand, gasification with pure O_2 of biomass is not a feasible process due to the high cost of using only O_2 (Havilah et al., 2022).

3.3.1. Gasification process of kitchen waste

Kitchen waste has a lot of organic matter in its composition, which has a high moisture content and a low energy density. The thermochemical conversion of kitchen waste through gasification represents a future energy production technique. Kitchen waste mainly includes vegetables, fruits, scraps, fruit peels, egg shells and generates more than 30 million tons accounting for 50% to 65% of solid waste. The Food and Agriculture Organization of the United Nations has claimed that more than 1.3 billion tonnes of kitchen waste is generated worldwide every year (Chhandama et al., 2022; Li and Jin, 2015). The composition of kitchen waste is shown in Table 4. which mainly consists of vegetables, fruit peels, cores, peels, meat and meat scraps, broken bones, egg shells and crustaceans (Liu et al., 2023).

Tabel 4 Characteristics of kitchen waste

Vegetables (Yuan et al., 2019)	**Peels, nutshells and cores** (Zhang et al., 2020)	**Leftovers and meat** (Jo et al., 2017)	**Eggshells, bones and** shells (Li et al., 2020)
62.4 wt.%	15.8 wt.%	15.6 wt.%	6.2 wt.%

In the gasification process of kitchen waste was used a stainless-steel reactor heated to 800°C with three heating wires (2500 W each) spiraling around the gasifier. The variation of temperature, air flow rate, moisture content and feed weight in kitchen waste gasification were monitored to optimize the process in terms of CO, CO_2, CH_4, H_2, tar and carbon production. It was observed that when the samples are heated up to a temperature of 800°C, only the mineral substances in the form of ash remain in the gasifier.
The yield percentage of the types of products obtained from gasification of kitchen wastes largely vary depending on the composition of the wastes and the parameters of gasification obtained by them is presented in Figure 3.
In this experiment they obtained a large volume of gaseous products (67%) by gasification of kitchen waste in a fluidized bed reactor. They observed that a high flow of air, as a gasification agent, increases the production of CO_2 and the amount of CH_4 decreases with increasing oxygen.
The conversion of kitchen waste in H_2-rich syngas was also investigated using supercritical water (Liu et al., 2022). They observed that H_2 yield increased significantly, from 150.32 mol /L to 563.43 mol / L with increasing temperature from 360 °C to 480 °C without catalyst and it increased to 673.13 mmol/L with the addition of Ni/Al_2O_3 catalyst.

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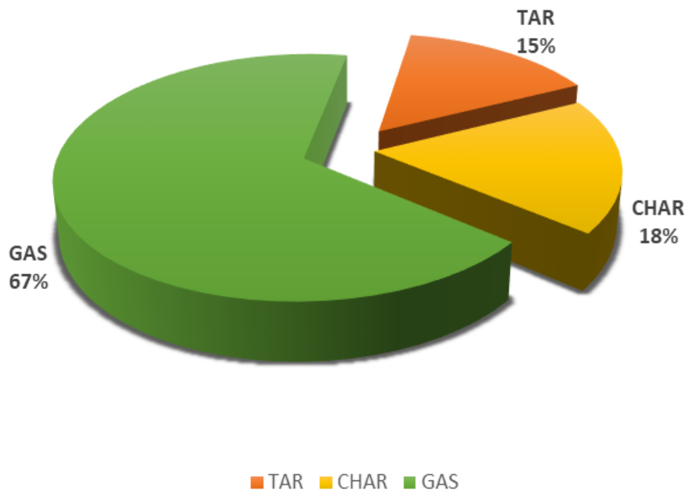

Figure 3. Yield percentage of the gasification products (Fatema et al., 2022)

3.3.2. Gasification process of chicken manure

They used different gasification media and temperatures ranging from 600°C - 1000°C in temperature steps of 100 °C. As in the case of gasification of kitchen waste (Chanthakett, A., et al., 2021), chicken manure used in this study it was also dried of irregular granular form with sizes ranging from 1-3mm. They analyzed the gases produced by gas chromatography. The main components of the obtained gas are H_2, CO, CO_2, CH_4, C_2H_6, C_2H_4, C_2H_2, and other higher series of hydrocarbons. Gasification with CO_2 and steam has been found to provide the highest energy yield as syngas. The evolutionary behavior of hydrogen mass for different gasification media (air, steam, CO_2 as gasifying agents) at different temperatures and observed higher hydrogen flow rates over longer periods of time with the use of steam compared to other gasification agents and concluded that the higher H_2 can be attributed to the reactions.

High yields of H_2-rich syngas were obtained from the steam gasification of chicken manure at 1000°C. On the other hand it was investigated H_2 production from supercritical water gasification (SCWG) of chicken manure in a high heating rate batch reactor at temperatures between 500°C and 620°C and observed that the H_2 yield increased significantly, from 9.27 to 22.47 mol / kg, when the temperature increased (Cao et al., 2021). In order to characterize the SCWG of chicken manure, several indicators including carbon gasification efficiency, (CE), hydrogen gasification efficiency, (HE), gas yield and gas fraction were employed. The definition of these indicators was listed as follows (Cao et al., 2021):

$$CE(\%) = \frac{total\ carbon\ in\ gas\ prodCE\ (\%)=ucts}{total\ carbon\ in\ manure} x100\% \quad (10)$$

$$HE(\%) = \frac{total\ hydrogen\ in\ gaseous\ products}{total\ hydrogen\ of\ manure} x100\% \quad (11)$$

$$Gas\ yield\left(\frac{mol}{kg}\right) = \frac{the\ molar\ number\ of\ each\ gas\ product}{mass\ of\ manure} x100\% \quad (12)$$

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$$Molar\ fraction\ (\%) = \frac{the\ molar\ number\ of\ each\ gas\ product}{total\ molar\ number\ of\ each\ products} x100\% \qquad (13)$$

4. CONCLUSIONS

In various gasification studies, of some of the most abundant types of biomass, kitchen waste, chicken manure, olive pomace, walnut shell, almond shell, sugar cane bagasse and lignocellulosic biomass, by different types of reactors, downflow, upflow, fluidized bed and plasma, the production of syngas, especially hydrogen, was analyzed. The critical parameters needed to improve hydrogen production are the high temperature of the process, 800÷1000°C, but also the use of steam as a gasification agent. Also, catalytic gasification contributed significantly to the increase of hydrogen yield in the experiments that used biomass/waste as raw material. Prior drying and shredding of the raw material to increase the contact surface with the reactant, helps in into a rapid and uniform gasification.

Taking into account the huge potential that biomass gasification has for the production of hydrogen-enriched gases, such studies must be continued and deepened, at least with regard to the catalysts used for the hydrogen enrichment of the obtained products.

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