Research article

# In-depth analysis of the effect of catalysts on plasma technologies for treatment of various wastes

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## ARTICLE INFO

## ABSTRACT

Energy security and waste management are gaining global attention. The modern world is producing a large amount of liquid and solid waste as a result of the increasing population and industrialization. A circular economy encourages the conversion of waste to energy and other value-added products. Waste processing requires a sustainable route for a healthy society and clean environment. One of the emerging solutions for waste treatment is plasma technology. It converts waste into syngas, oil, and char/slag depending on the thermal/non-thermal processes. Most of all the types of carbonaceous wastes can be treated by plasma processes. The addition of a catalyst to the plasma process is a developing field as plasma processes are energy intensive. This paper covers the detailed concept of plasma and catalysis. It comprises various types of plasma (non-thermal and thermal) and catalysts (zeolites, oxides, and salts) which have been used for waste treatment. Catalyst addition improves gas yield and hydrogen selectivity at moderate temperatures. Depending on the properties of the catalyst and type of plasma, comprehensive points are listed for the selection of the right catalyst for a plasma process. This review offers an in-depth analysis of the research in the field of waste-to-energy using plasma-catalytic processes.

## 1. Introduction

Industrialization and urbanization have made day-to-day life easier but have led to severe issues such as energy crises and the large volume of waste generation. The energy demand has increased significantly. Currently, fossil fuel is the main source of non-renewable fuel for energy generation. It is a need of an hour to find other sources of energy to serve the world population. On the other side, there is a large amount of waste generated worldwide on daily basis due to the modern lifestyle of a large population. Approximately 500 kg of waste per person is generated yearly in a developed country (Mahapatra et al., 2022). Hence, the management of such a massive volume of waste has become a major concern worldwide (Sondh et al., 2022). The available waste treatment techniques are landfill, incineration, composting, pyrolysis, and gasification (Yasmin et al., 2022). Sanitary landfills and incineration are the most predominant technologies for waste treatment around the world (Kundu et al., 2023). Approximately 37% and 11% of total wastes are treated by landfills and incineration, respectively (Kaza et al., 2018). Landfills are increasingly used for the treatment of hazardous waste in the countries with emerging economies. In Europe, approximately 40%

of hazardous waste is treated by sanitary landfills (Li et al., 2023). Sanitary landfill is well-structured, flexible and low-cost technology which makes it suitable for waste treatment. On the other hand, in many economically weak countries, open dumping landfills are widely used due to cost-effective and simple option, which leads to severe issues like more and more land requirements, toxic gaseous emissions, water pollution and land contamination (Kumar et al., 2020). The landfill does not support circular economy as well. Incineration is a thermal technology faster than landfill and bio-processes; hence it is widely used for volume and mass reduction of MSW (Kremser et al., 2021). However, along with very high cost, it emits toxic gases which pollute the air severely. Composting is a sustainable option which treats around 6% of the total waste (Kaza et al., 2018), however, it can treat only green organic waste and it is a very slow process compared to thermal processes. Thermal processes include combustion, gasification and pyrolysis. Combustion generates toxic emissions, and gasification also generates a certain amount of dioxin and furans due to the presence of oxygen. In slow pyrolysis and gasification, the formation of gaseous products is less, while the formation of liquid products is more. Looking at the quantity of waste and the limitations of currently available

processes, there must be some sustainable options for waste valorization (Bhatt et al., 2022). Low emission economy aims to alleviate the carbon emission (Devasahayam, 2019). Energy recovery is also important in current scenario of non-renewable energy scarcity. The energy recovery in kg of oil equivalent (kgoe) per ton of MSW from various waste treatment processes are nearly 4.5–9 kgoe/ton from landfills, 36–45 kgoe/ton from incineration, 35–63 kgoe/ton from gasification and 45–50 kgoe/ton from pyrolysis (Sondh et al., 2022). Waste treatment options are selected on the bases of factors such as sustainability, safety, space requirement, decomposition rate, and local regulations. Waste valorization converts waste to material or waste to energy. It helps in reducing large amounts of generated waste and consequences related to open dumping. On the other hand, non-renewable energy sources are depleting with an increase in the population. Energy from non-conventional sources is beneficial for socio-economic growth. Different types of waste are generated with or without the segregation of MSW, such as various plastics (PET, PE, PP, PS), rubber, biomass, paper, textile, RDF. Proximate analysis and ultimate analysis of wastes help in selecting the waste processing technique effectively. In proximate analysis, fixed carbon is associated with delayed degradation of waste, while volatile matter is associated with high conversion of waste to gas at high temperatures or faster degradation (Fetene et al., 2018). Moisture content also affects the quality of syngas. The various categories of waste can be handled differently, depending on their degradability and harmfulness (Kumar et al., 2020). The product gas yield and quality highly depend on the composition and heating value of the waste feed (Fabry et al., 2013). The HHV of these wastes are generally in the range of 15 MJ/kg to 45 MJ/kg. Out of all these waste materials, MSW is heterogeneous in nature and its composition varies with region, season and time. RDF is derived from MSW after sorting the recyclable materials. The heating value of RDF is higher than MSW. Because of these reasons, it is recommended to use RDF than MSW in waste-to-energy technologies.

Thermal technologies like pyrolysis, gasification, or incineration show better and speedy conversion of waste in comparison to non-thermal technologies like landfill and bio-processes. These processes are applicable for fast and effective solutions to deal with the heavy metal particles as well (Liu et al., 2023). However, these thermal processes are not as efficient as plasma processes for energy generation (Li et al., 2016). Amongst these processes, incineration and gasification may create toxic compounds as well (Bhatt et al., 2022). Plasma technology is the advanced thermal technology which seems to be one of the favourable ways to convert wastes to value-added products. The plasma processes mainly include plasma gasification/vitrification and plasma pyrolysis. These processes convert carbonaceous waste to combustible gases and carbon-rich residue. The output depends upon various factors such as the temperature of the process, air supply, catalyst addition, flow rate of feed, etc. Plasma processes are very fast. These processes are suitable for the valorization of non-degradable waste and converting those waste materials to energy. The temperature requirement of the reaction can be lowered by the use of a suitable catalyst (Cai and Du, 2021). The development of a catalyst for the enhanced production of $H_2$ in waste treatment processes is an absolute necessity (Chai et al., 2020). The reaction time can also be reduced by the application of catalysts. Catalysts improve the rate of decomposition of the hydrocarbons. Reaction yields can also be improved with catalysts. Plasma catalysis is widely used for gases, for example, VOC breakdown, conversion of $CO_2$, $NO_x$ conversion, synthesis of $NH_3$, $H_2S$ elimination, etc. (Bogaerts, 2019). Catalytic plasma gasification can improve gas yield and $H_2$ production in comparison to catalytic gasification at as low as 500 °C temperature (Al-Fatesh et al., 2023). Plasma and catalysis both the processes have their advantages individually in gaseous, liquid and solid waste treatment, but the integration of plasma and catalytic processes has not been studied much in detail. Kaza et al. (2018) reported that only less than 1% of total waste is treated by methods other than conventional methods. Hence it is an emerging area for waste treatment,

especially for solid wastes. A considerable study in plasma-catalytic processes for waste valorization is underway. This review paper covers different aspects of the addition of catalyst(s) to plasma gasification/pyrolysis processes thoroughly.

The paper has the following framework for the forthcoming sections. Section 2 includes the details of plasma technologies and their configurations which are available for waste-to-energy conversion. Section 3 describes different catalysts which are used for waste-to-energy processes, i.e. pyrolysis and gasification. It also includes the preparation method for catalysts. Section 4 includes plasma-catalysis, the chemistry involved in the process and the criteria for the choice of catalysts for plasma processes. Section 5 discusses the concluding remarks of this paper and future prospects. This review will benefit the researchers in the field of waste management by providing insight into the plasma-catalytic processes, which are considered state-of-the-art.

## 2. Plasma technologies for waste treatment

Plasma technologies include plasma application in the gasification or pyrolysis processes of waste valorization. These are thermo-chemical processes for the treatment of waste, in which the plasma is used to provide the energy and temperature to the gasification/pyrolysis process. Plasma technologies are compact and rapid in nature.

### 2.1. Overview of plasma

Plasma in waste valorization technologies is in form of an arc which can be generated using electric current (Materazzi and Taylor, 2019). Plasma mainly consists of neutral molecules along with radicals, electrons, ions etc. (Bogaerts and Centi, 2020). The plasma generation can be governed by Paschen law, which is related to the gap between two electrodes to initiate ionization. Yuan et al. (2014) kept a 5 mm gap between two electrodes in RF plasma (Yuan et al., 2014). Plasmas are mainly of two types: non-thermal plasma and thermal plasma. Thermal plasma has higher enthalpy and excitation energy than non-thermal plasma. Thermal plasmas have thermal equilibrium where electron temperature (Te) is equal to the heavy particle temperature (Th) unlike in non-thermal plasmas. Hence, non-thermal plasmas are non-equilibrium plasmas while thermal plasmas are equilibrium or quasi-equilibrium plasmas as shown in Table 1 (Breeze, 2018; Tendero et al., 2006).

Type of the feed, degradation temperature and reactor type are the major factors for the selection of plasma source. Plasma sources can be combined to make a hybrid plasma reactor which may reduce cost (Kaushal and RohitDhaka, 2022). With an increase in temperature, the energy density also increases. Feed in longer contact time with high temperature inclined towards higher gas yield (Huang and Tang, 2007).

As mentioned in Fig. 1, thermal plasma technology offers various advantages over other waste treatment technologies. It offers high energy density, high temperature, fast reaction, lesser emission, high gas yield, high conversion and less start-up time (Cai and Du, 2021; Puliyalil et al., 2018). Ionization in plasma can be occurred by different

**Table 1**
Types of plasma and their properties.

| Plasma | Non-thermal | Thermal | |
| --- | --- | --- | --- |
| | | Low-temperature | High-temperature |
| Ionization | Small portion is ionized | Large portion is ionized | Fully ionized |
| Plasma Density | Lower | Higher | Highest |
| Temperature | Te ≫ Th | Te ≈ Th | Te=Th |
| Equilibrium state | Non-equilibrium | Quasi-equilibrium | Equilibrium |
| Plasma generator type | Corona discharge, gliding arc | Microwave, RF, AC/DC arc | Fusion plasma, solar wind, lightning etc. |
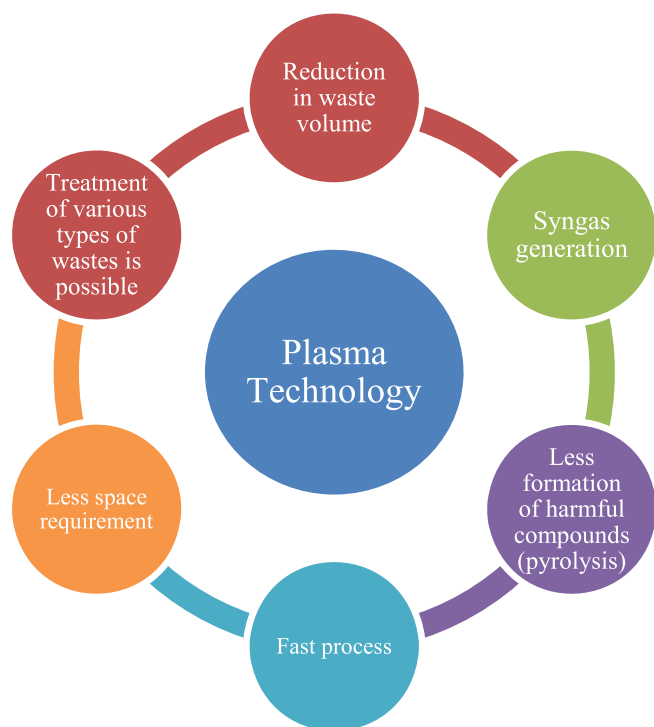
**Fig. 1.** Advantages of plasma technologies.

mechanisms as follows: collision ionization, stepwise ionization, heavy particle collision ionization, ionization by the collision of photons and neutrons and ionization by emission of electrons (Carreon, 2019). Carbonaceous wastes in the form of solid, liquid or gaseous states can be treated using plasma technology (Hassanpour, 2017). Plasma technology can also be combined with other processes because of its flexible nature (Whitehead, 2010).

### 2.2. Plasma technologies

#### 2.2.1. Plasma gasification

Plasma gasification involves the decomposition of material at high temperatures in the presence of air/oxygen. The amount of oxygen is kept limited in this process. Therefore it is known to be an incomplete oxidation process. Plasma gasification converts the waste to product gas and solid residue in form of char or slag. It is used to treat various categories of waste including bio-medical waste because of high temperature and fast process. In plasma gasification, the product gas has considerable calorific value (Agon et al., 2016). The reactions involved in gasification include water-gas shift reaction, Boudouard equilibrium reaction, combustion reaction, methanation reaction, dry-reforming reaction and steam-methane reforming reaction etc. (Byun et al., 2012). Plasma adds an advantage of enhanced $CO_2$ conversion to CO in the process of plasma gasification (Giammaria et al., 2019). In the plasma gasification, almost no tar was present (Tang et al., 2010). Whereas in conventional gasification, tar formation may be in the range of 1 $g/m^3$ to 100 $g/m^3$, which can be curtailed using catalyst, high temperature or plasma application (Devi et al., 2003; Liu et al., 2017; Sikarwar et al., 2016). It is able to reduce the large volume of waste with a high conversion efficiency around 75%–89% mainly depending upon the type of feed, input power and feed flow rate (Bhatt et al., 2022; Gabbar et al., 2020; Tang and Huang, 2010). Plasma gasification is advantageous as it produces more gas, less toxic compounds and minimum leachates than gasification and incineration (Mukherjee et al., 2020). However, due to the presence of oxygen, this process produces oxygen-based pollutants such as furans and dioxins. To overcome this

issue, the oxygen in the process needs to be minimized. Hence plasma pyrolysis should be taken into consideration. It is to be noted that investment and operating costs are high in the case of plasma gasification and plasma pyrolysis due to high energy requirement (Pieta et al., 2018).

#### 2.2.2. Plasma pyrolysis

Plasma pyrolysis is the process of dissociation of molecules at high temperatures without oxygen/air supply. Apart from municipal wastes, plasma pyrolysis may be used to treat hazardous, industrial, medical and e-wastes as well due to high temperatures and lack of oxygen. The gaseous product from plasma pyrolysis mainly consists of $H_2$, CO, $CH_4$ and other light hydrocarbons. The amount of each gaseous component depends on the feed composition along with operating conditions and the type of plasma pyrolysis process. The inorganic fraction is in the form of char or vitrified metal slag. Pyrolysis is influenced by many factors such as reaction temperature, feed flow rate, residence time, heating rate, composition of feed etc. Non-thermal plasma pyrolysis takes place at lower temperatures, generally ranging from 100 °C to 800 °C. The temperature range of thermal plasma pyrolysis is generally from 800 °C to 1500 °C. Thermal plasma is beneficial for the toxic compounds removal. Even though the energy consumption of the plasma pyrolysis process is high due to high power requirements, it reduces the volume of waste with minimum pollutant emissions. This makes the process of plasma pyrolysis a "zero-waste" process (Samal and Blanco, 2022).

### 2.3. Configuration of a plasma process

The plasma gasification/pyrolysis process comprises a plasma reactor, air/gas injector, electric power supply, electrodes, scrubbing and cleaning assembly, and gas outlet. The overview of the setup is shown in Fig. 2. In some cases, the shredder and conveyor are also part of the system for shredding and feeding the solid material to the reactor. The power supply may be DC or AC, which is supplied to the electrodes for the generation of plasma. Oxygen or air is supplied to the reactor system in case of gasification, or else inert gas is supplied for pyrolysis. For steam gasification or steam pyrolysis, an additional arrangement of steam injection is provided. Solid residue can be removed from the reactor directly and gas is sent further for cleaning. Wet scrubbing or condensation is used for the product gas treatment. It is followed by gas filtration and the final product gas is sent for analysis generally using gas chromatography.

The plasma system is mainly governed by voltage, current, carrier gas flow rate, plasma source, type of electrode, plasma temperature, and reactor pressure (Hinde et al., 2020).

### 3. Catalysts used in plasma processes

A substantial amount of energy is required in the thermal cracking of waste, as it is an endothermic process. Such processes require high activation energy. To enhance the rate of reaction and decrease the activation energy of the thermochemical conversion of waste material, a catalyst plays a vital role (Sriningsih et al., 2014). The morphology of the residue char can also be improved using catalysts (Miskolczi et al., 2013). Char from high temperatures may behave as a catalyst (Wang et al., 2017). The catalyst activity can be derived using dispersion and surface area.

Various factors of catalysts and plasma processes, which may affect the waste conversion processes, are shown in Fig. 3. There are two types of catalysts, which are used for the waste conversion: microporous (pore size < 2 nm) and mesoporous (pore size ∼ 2 nm–50 nm) (Li S.C. et al., 2021; Miandad et al., 2019). Microporous catalysts have limitations for large-sized particles, which may not be diffused in pores and coke formation may clog the micropores (Li S.C. et al., 2021). Mesoporous catalysts may overcome the limitations of large-sized particle diffusion and
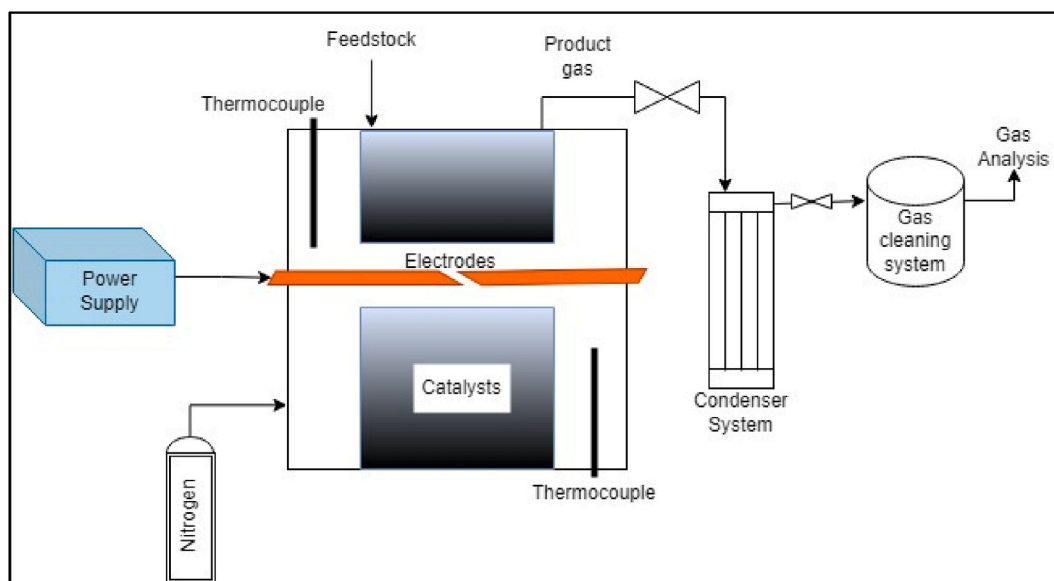
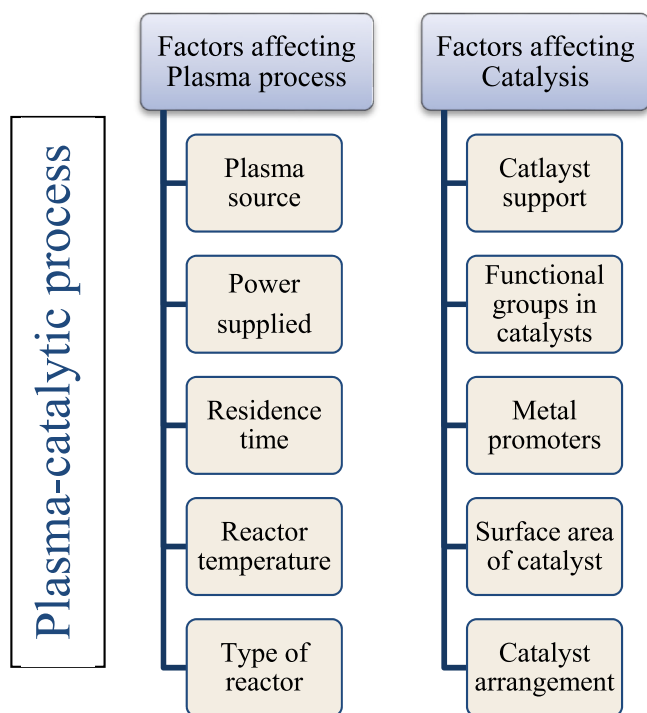**Fig. 2.** Overview of plasma-catalytic process.



**Fig. 3.** Factors affecting plasma-catalytic process.

clogging the catalysts.

Various categories of catalysts and their supports are available for pyrolysis such as zeolites, metallic oxides, and inorganic salts (Cao et al., 2020; Li H. et al., 2021). Ferroelectric and semiconductor materials are also used as catalyst support materials. Different types of support materials have different qualities. For example, metal oxides improve the activity of the catalyst, while ferroelectric materials have a high dielectric constant with a very low BET surface area (Kim et al., 2015). Various types of catalyst support materials and loading metals are discussed in the following sub-sections.

### 3.1. Zeolites

Zeolites are extensively used catalysts for the pyrolysis of waste. They are hydrothermally stable materials with higher shape selectivity. Different types of zeolites have different structures, i.e. zeolite A contains an 8-membered ring whereas zeolite x and zeolite Y contain 12-membered ring (Indira and Abhitha, 2022). ZSM-5, ZSM-35, and ZSM-39 are examples of high silica zeolites. The structural difference affects the composition and yield of the final products of plasma gasification or plasma pyrolysis. It was observed that β-zeolite has higher micropores which led to higher selectivity to $H_2$ (Li et al., 2020). Also, a higher BET surface area decreases the amount of char formation. It was also concluded that zeolites promote the formation of oil rather than gas products, though in the case of non-plasma processes. Metal-impregnated catalysts have a major role in the plastic conversion to fuel. Vasile et al. (2001) observed that HZSM-5 and modified catalyst PZSM-5 using ortho-phosphoric acid showed high selectivity and stability for mixed plastic waste in the pyrolysis process where the temperature was kept only around 480 °C (Vasile et al., 2001). High syngas yield was obtained with a metal-loaded catalyst for high-temperature pyrolysis of municipal plastic waste at 700 °C (Al-Asadi and Miskolczi, 2020). The alkane cracking in zeolites is also subject to Brønsted acid sites and the position of Al in the structure of the catalyst (Li S.C. et al., 2021). It was also observed that lanthanum and cerium favoured hydrogen selectivity and yield with Ni/ZSM-5 (Al-Asadi and Miskolczi, 2020). ZSM-5 has a high Si/Al ratio which makes it a thermally stable catalyst (Pérez-Page et al., 2016). The use of duel catalysts for combining favourable properties may contribute to a long life of catalysts with delayed deactivation (Peng et al., 2022).

### 3.2. Metallic oxides

There are various types of metal oxides available for thermal catalysis. $Al_2O_3$, $TiO_2$, $CeO_2$, and $SiO_2$ are examples of metal oxides. Metal oxides promote coupling reactions (Bogaerts et al., 2020). Aminu et al. (2022) worked on various metal catalysts such as Ni, Fe, Co, and Cu, supported on $Al_2O_3$. Nickel in comparison to Fe, Co, and Cu was observed to be the most favourable metal catalyst for hydrogen production. The addition of steam also favoured the increase in hydrogen yield (Aminu et al., 2020). Nickel-based catalysts were tested at 800 °C to make sure that they can withstand thermal plasma catalytic reactions. As the temperature increases from 500 °C to 900 °C, the gas composition

changes by the increased amount of hydrogen (Kusz et al., 2022). Blanquet and Williams (2021) worked on pyrolysis-plasma/catalysis for the treatment of biomass. They worked on different catalyst support materials (Ni–Al$_2$O$_3$, Ni–Y-zeolite, Ni–TiO$_2$) with various metal promoters (Ce, Co, Mg, Fe, Ni, Cu). It was observed that Ni–Al$_2$O$_3$ resulted in the highest product gas yield and Ce was the best promoter out of all tried for enhanced H$_2$ yield. Ni/Al$_2$O$_3$ also showed better syngas productivity in plasma-catalytic dry reforming of methane (Zeng et al., 2015). Ni/CeO$_2$/Al$_2$O$_3$ catalyst was used in the gasification of polypropylene for the temperature range of 600 °C–900 °C (Wu and Williams, 2008). It was observed that as the temperature increased from 800 °C, there is no oil present in the product and gas yield also increased significantly at 900 °C. Metal oxides are considered to be cost-effective, especially for VOC treatment. Metal oxides with non-thermal plasma favour NOx removal and other gas treatment (Durme et al., 2008).

### 3.3. Inorganic salts

K$_2$CO$_3$, MgCO$_3$, MgCl$_2$, MnCl$_2$, FeCl$_3$, dolomite etc. are generally used for biomass pyrolysis. At high temperature (900 °C), K$_2$CO$_3$ improved the quality and quantity of syngas and good quality char with added micropores. K$_2$CO$_3$ increased the H$_2$ formation and decreased the CO formation. CH$_4$ decreases slightly at high temperature (800–900 °C) and then remained unaffected (Zhou W. et al., 2018). CaCO$_3$ produced the highest gas yield for rice straw pyrolysis followed by MgO and MgCO$_3$. FeCl$_3$ produced 38.5% gas yield in the rice straw pyrolysis process (Cao et al., 2020). But FeCl$_3$ is not suitable for thermal plasma pyrolysis due to its thermal instability. Waste glycerol was treated using a plasma gasification process which contains a heterogeneous salt catalyst (Na$_3$PO$_4$) (Tamošiūnas et al., 2019). It speeds up the transesterification of glycerol. Dolomite is one of the widely used catalysts for waste conversion due to its low cost (Lui et al., 2020). The major

advantage of salts is that they can be mixed directly with the feed (Bulushev and Ross, 2011).

Fig. 4 shows the properties of different catalysts used for gasification/pyrolysis processes, with or without plasma.

It can be seen that zeolites offer a higher BET surface area. The conversion of heavy hydrocarbons to light hydrocarbons is influenced by cracking which is effective when the surface area of the catalyst material is higher (Neyts et al., 2015a,b). Also, it can be observed in Fig. 4, the pore diameters of most catalysts are in nanometres. For better efficiency of the catalyst, smaller diameters are favourable (Neyts et al., 2015a,b). Synergy is observed in the case of selected bimetallic catalysts which increase the product gas yield in comparison to non-metal loaded catalysts (Zhang et al., 2019). Dielectric constant affects catalytic pores and also enhances the local electric field in presence of plasma. The rate of reaction is highly influenced by the electric field which can be improved with polarization on the catalyst pore surface (Zhang et al., 2016). An internal electric field is created due to polarization which lowers the electron energy due to resistance to the overall electric field. Hence high dielectric constant reduces voltage but does not help in catalytic reactions in the plasma environment (Aminu et al., 2022). The electric field is also influenced by roughness due to geometry (i.e. catalyst in plasma reactor) or catalyst porosity (Neyts and Bogaerts, 2014). The selection of a catalyst is a critical step in any plasma-catalytic process. Most of the catalysts that are used for thermal pyrolysis are being used for plasma pyrolysis. Catalyst surface activity defines the adsorption efficacy, which is better in the case of plasma processes due to excitation energy (Whitehead, 2016).

### 3.4. Catalyst preparation

The preparation of supported catalysts can be carried out by the wet impregnation method. This method is known to provide considerable
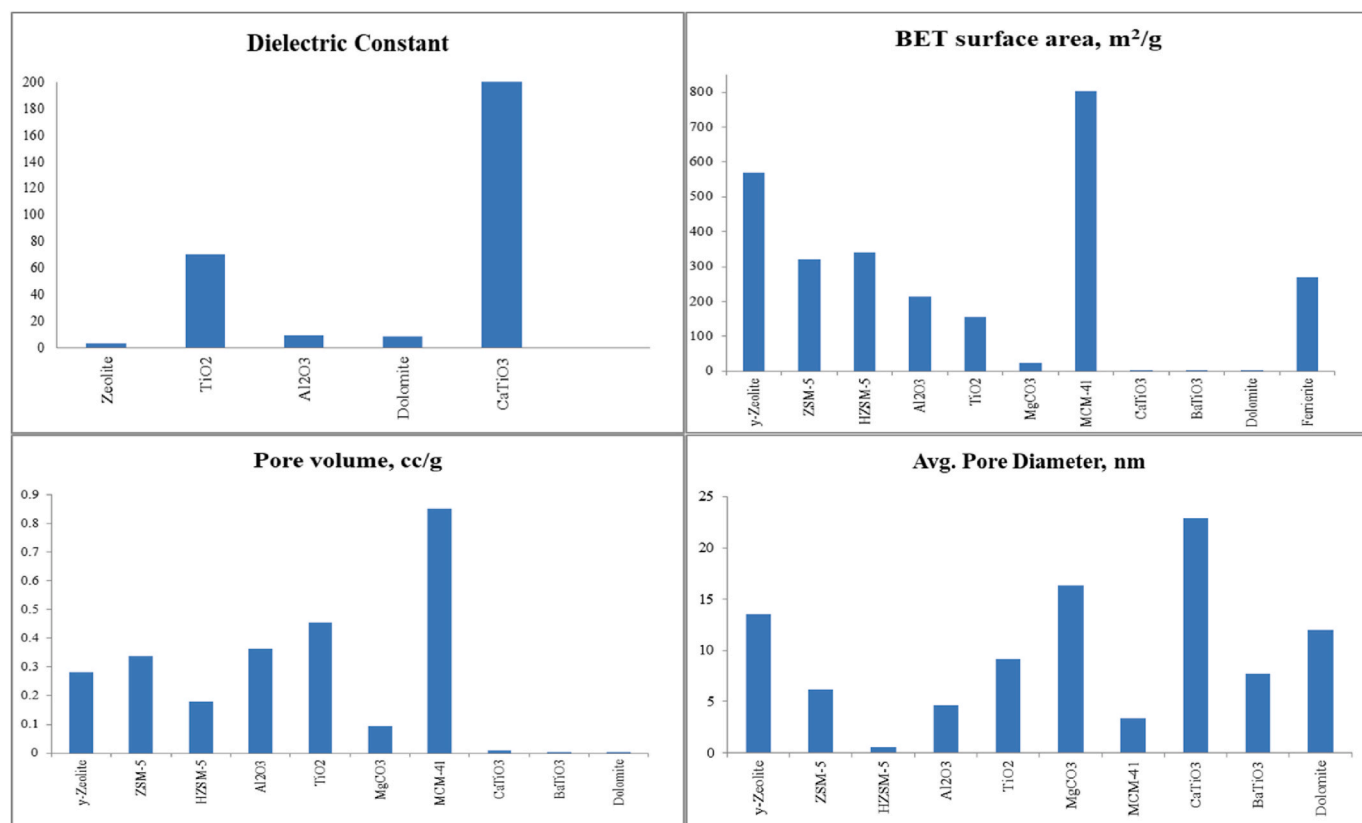


**Fig. 4.** Properties of catalyst support materials (Plotted with the data from: Aminu et al., 2022; Blanquet and Williams, 2021; Hernandez et al., 2007; Kim et al., 2006; Upadhyay et al., 2019; Xiao et al., 2022.).

dispersion of active metal on the catalyst support. Important parameters for the process are calcination temperature and reduction time. For Nickel based catalysts the calcination temperature was kept between 750 °C to 850 °C and the reduction time was between 1 and 3 h (Garcia et al., 1998). Aminu et al. (2022) used this method to prepare a Ni-based catalyst. The support material was added to the nickel nitrate which is dissolved with DI water. The mixture was stirred for half an hour and heated to convert it into the slurry. It is then placed for drying using an oven followed by calcination for 3 h at 750 °C. The calcined catalyst is then ground and sieved for the required size. The reduction was done at 800 °C under a hydrogen atmosphere for 2 h. The heating rate was 20 °C/min for the reduction process. Xiao et al. (2022) used a ready-made ZSM-5 catalyst which was ground for the required size. The material was then sieved and the calcination was carried out for 5 h at 550 °C temperature. Blanquet and Williams (2021) studied the effect of metal promoters on Ni–Al$_2$O$_3$ catalyst. They prepared the catalysts by impregnation using 5 wt% promoter metal to 5 wt% Ni. It was then dissolved in DI water followed by support (Al$_2$O$_3$/TiO$_2$/Y-zeolite) addition. It was then heated up to 95 °C to make it a slurry material and then placed for drying. Calcination took place at 750 °C with a heating rate of 2 °C/min. Crushing and sieving were carried out for converting it to the required size. The reduction process was carried out at 800 °C for 1 h with a heating rate of 20 °C/min (Blanquet and Williams, 2021). Some catalysts are commercially available with the required specification. Acharya et al. (2010) used CaO for biomass steam gasification at temperatures up to 710 °C. The CaO catalyst was heated for 2 h at 950 °C for generation. Liu et al. prepared Ni/γ-Al$_2$O$_3$ by adding γ-Al$_2$O$_3$ to the sol-gel followed by impregnation for 12 h and sent for moisture removal. Calcination was carried out for 3 h at 750 °C. Percentage of Ni was varied from 5% to 20% in precursor solution (Liu et al., 2017). Usually, the catalyst should be used after heating in the furnace for 24 h at or slightly above 100 °C to remove moisture.

## 4. Plasma-catalytic processes

Process of plasma-catalysis is the synergy of catalytic features and plasma surface interactions. It is an emerging discipline in the field of thermochemical waste processing.

### 4.1. Chemistry involved in catalytic and plasma-catalytic processes

The study of chemistry of catalytic processes is important for understanding the mechanism of the plasma-catalytic processes. The rate of degradation of a catalyst with an increase in temperature is to be known for catalyst selection. Physical or chemical adsorption is the primary interaction mechanism in the catalytic processes. Catalyst surface chemistry is another mechanism which is very crucial in determining the conversion, selectivity, and efficiency of the catalytic process (Neyts, 2016). High temperature tends to increase the formation of primary radicals. High energy is required for the conversion of feed to hydrocarbons. It has been observed that a temperature higher than 800 °C is required for higher hydrogen production from plasma pyrolysis (Bhatt et al., 2022). To improve this conversion temperature should be kept above 1000 °C for the non-catalytic process (Sobacchi et al., 2002). Plasma pyrolysis mainly includes the thermal cracking of waste material. Catalytic pyrolysis includes surface mechanisms including cracking as well as isomerization. Catalyst properties such as characteristics of pores and acidity are the governing factors for such mechanisms (Fadillah et al., 2021). The reaction temperature can be lowered by selective catalytic reduction (Male, 2021). The plasma-catalytic process increases H$_2$ and lower hydrocarbon production by increasing the fission of C–C and C–H bonds (Xiao et al., 2022). Low temperature results in unsaturated polymers while high temperature breaks C–C and C–H bonds to syngas with CO$_2$ and lighter hydrocarbons (Bulushev and Ross, 2011). Saturated components undergo dehydrogenation in the presence of metal-based catalysts (Saidi and Zhandnezhad, 2023).

Generally, the polymeric portion of the MSW follows the mechanism of initiation, propagation and termination. Initiation includes scission reactions, propagation follows decomposition reactions and abstractions and termination include second-order reactions (Sipra et al., 2018).

Xiao et al. (2022) presented the synergistic mechanism of catalytic pyrolysis and plasma-catalysis pyrolysis, which is shown in Fig. 5. The left side of Fig. 5 illustrates the mechanism of catalytic pyrolysis and the right side shows plasma-catalytic pyrolysis. It shows that there is no pre-cracking of higher hydrocarbons in catalytic pyrolysis, due to which the heavy molecules block the pore of the catalyst. This is one of the reasons for high coke and wax deposition on the catalyst surface in the process of catalytic pyrolysis. On the other hand, plasma converts higher hydrocarbons to lower hydrocarbons which can disperse into catalyst pores easily. Plasma can also improve the catalyst surface. Plasma pyrolysis of waste material involves thermal degradation in the near absence of air/oxygen. In the thermal catalytic process, the major steps are adsorption, surface reaction and desorption, while in the plasma-catalytic process, bombardment, excited molecules and radicals drive the process (Kim et al., 2016; Neyts et al., 2015). Primary decomposition is followed by secondary reactions which convert to gases and solid residue in form of char or slag (Chhabra et al., 2016). The complex chemical reactions of plasma catalysis are not available in the literature in detail. Dissociation takes place due to radical formation in plasma-based reactions, which produces excited species and prompts the decomposition of feed. The following reaction of thermal cracking is generally considered to be taken place in a plasma pyrolysis reactor:

$$C_nH_mO_l + Plasma\ Heat\ H_{2(g)} + CO_{(g)} + CO_{2(g)} + C_1-C_{4(g)} + Char_{(s)} \qquad (1)$$
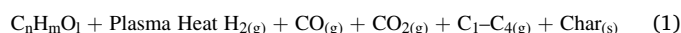
Steam addition may behave more like gasification and it follows a water-gas reaction. The effect of steam in presence of a catalyst is studied by Liu et al. (2018). They observed that for high syngas selectivity, BET surface area and pore volume should be low. Nickel has a lower oxygen affinity than iron. Hence it produces a low amount of oxygenated compounds. Ni also gives more H$_2$ yield than Fe, Co, and Cu (Liu et al., 2018; Zeng et al., 2015). In a plasma environment, the catalyst may be provocative for forward and backward reactions, which is not desirable. Hence the selection of catalyst should be such that it should favour the product reactions and not the undesired products (Bogaerts et al., 2020). Diels-Alder reaction for linear plastic shows less coke formation on the catalyst surface. The high BET surface area of a catalyst promotes the rate of reaction for cracking, which improves product gas yield (Li et al., 2020). Plasma helps to overcome the activation energy barrier by radical formation or excited species (Neyts et al., 2015a,b). Plasma may produce a photocatalytic effect by providing enough energy to excite the electrons.

### 4.2. Treatment of wastes using plasma-catalytic processes

Plasma-catalysis is the combination of a plasma heat source and catalyst in the same process. It helps to get better conversion compared to individual processes of plasma or catalysis (Yan et al., 2022). The interaction between plasma and catalysis creates an additional effect on the process, which is known as synergy in plasma-catalytic processes (Vandenbroucke et al., 2011). Plasma produces high energy electrons in comparison to bond-breaking energy. It can enhance the rate of reaction by producing radicals using high energy (Liu et al., 2016). As thermal plasma involves equilibrium between electrons and gas particles, it has less chemical selectivity (Zhu et al., 2020). Hence, the addition of a catalyst may help in overcoming the issue with higher chemical selectivity. The plasma-catalytic process improves gasification at lower temperature with a lesser quantity of air (Cai and Du, 2021). In case of non-thermal plasma, increase in power leads to higher gas yield (Song et al., 2022). Thermal plasma provides very high heating rates, which may lead to thermal lag (Al-Salem et al., 2017). Plasma-catalytic processes are arranged in three manners as shown in Fig. 6 (Carreon, 2019;
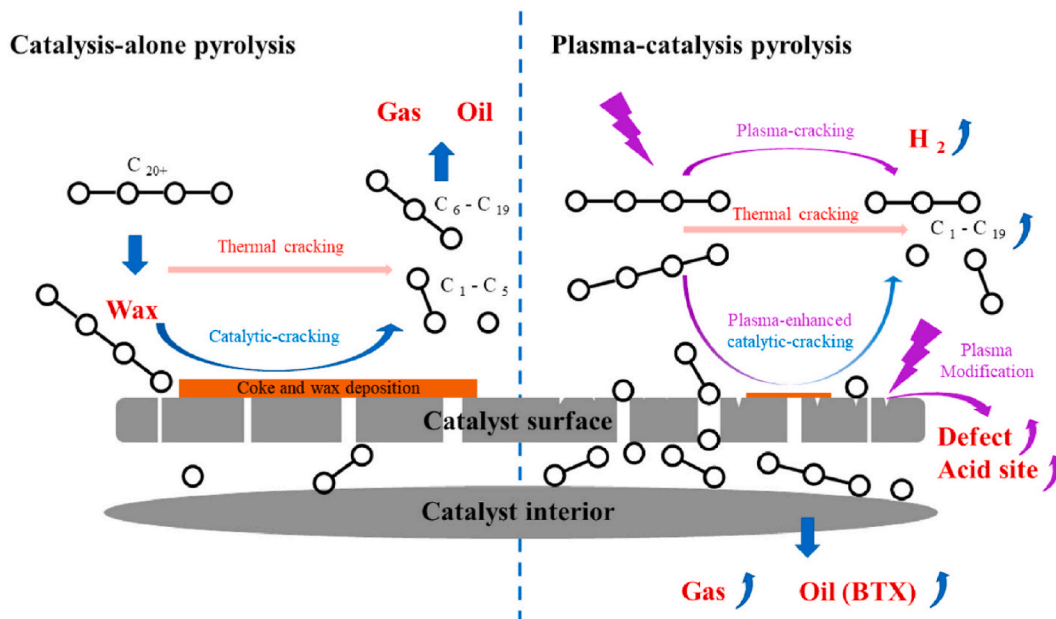
**Fig. 5.** Mechanism of catalytic pyrolysis vs plasma-catalytic pyrolysis (Xiao et al., 2022).
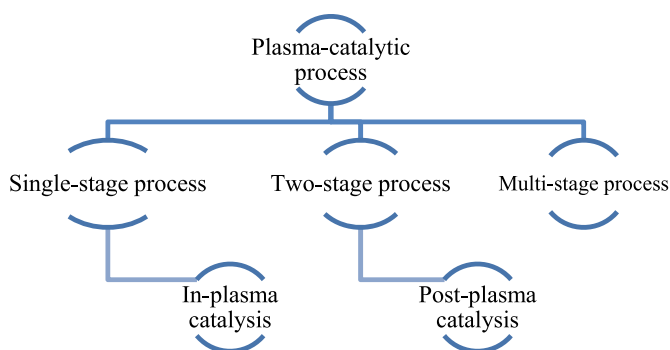


**Fig. 6.** Arrangement of plasma-catalytic process.

Kim et al., 2015). In a single-stage process, the catalyst is placed inside the plasma reactor, which is also known as in-situ catalysis. The arrangement of the catalyst may be placed in layers or coatings (Durme et al., 2008). While in the two-stage process, the catalyst is placed in the catalytic reactor after the plasma discharge, which is called ex-situ catalysis (State et al., 2019). In a multi-stage process, there are more than two types of different catalytic beds placed stage-wise which have a different effects on the decomposition pattern.

During the plasma catalysis process, surface of the catalyst and reactants interact and excitation occurs due to an active catalyst or electrically charged plasma gas (Hinde et al., 2020). The presence of a catalyst may reduce the reaction temperature due to less activation energy requirement and may increase the selectivity of hydrogen in the product gas. Catalysts can decompose the reactants at a lower temperature (Cai and Du, 2021). It also enhances the yield of the product (Mukherjee et al., 2020). The reaction time can also be reduced by the use of catalysts. Catalysts improve the rate of decomposition of the hydrocarbons. The calorific value of the product gas increases at high power plasma with catalysis (Młotek et al., 2019). Due to UV irradiation, there is a photocatalytic effect in the thermal plasma pyrolysis process (Giammaria et al., 2019). Electric properties of catalysts play a major role when applied plasma. The electric field distribution is also affected by a catalyst in the plasma process. Most of the studies presented in Table 2 observed that, low surface area, low porosity and high dielectric constant of the catalyst adversely affects the plasma discharge by

modifying the properties of discharge (Aminu et al., 2022; Blanquet and Williams, 2021). Electric properties of catalysts such as dielectric constant, electric conductivity and surface geometry affect the electric field distribution. Electrical properties accelerate the electric field locally, however the electric field in the void space of the reactor decreases, which may impact overall electric field (Jo et al., 2014). The overall plasma-catalytic process is affected by the term "reduced electric field" (which is electric field divided by gas pressure or the number of neutral particles). Low dielectric constant catalysts have application in reforming reactions and enhanced the hydrogen selectivity. Catalysis has many applications in the field of methane reforming, water-gas shift reactions as well as in gasification/pyrolysis of solid waste.

Table 2 shows various plasma-catalytic processes for the gas yield and product gas composition. It can be seen that most of the work has been done in non-thermal plasma catalysis. The effect of catalysis is more on primary reactions of pyrolysis than secondary reactions. Secondary reactions are more influenced by temperature (Hernandez et al., 2007). The elementary reactions take place in the pores of catalysts and form light hydrocarbons. Secondary reactions like cracking are improved in the presence of plasma which increases the yield of product gas. Therefore, syngas formation is mainly affected by plasma. In microwave plasma pyrolysis also, the high power/high temperature enhances gas yield while oil yield is more when the temperature is below 700 °C (Putra et al., 2022). They also concluded that the addition of a catalyst improves the performance of the microwave plasma process. Microwave plasma pyrolysis with catalyst is faster process to convert the waste to syngas and hydrocarbons (Konstantinov et al., 2022). Hence plasma pyrolysis along with catalyst can be more beneficial than the individual processes. Song et al. (2022) reported that with decrease in feed to catalyst ratio (4:1 to 1:2), oil yield increases (16%–26%). Catalytic non-thermal plasma processes have effectively removed VOCs (Yan et al., 2022). An example of dealing with hazardous gaseous waste using plasma processes is the treatment of chlorofluorocarbon using $TiO_2$ (Whitehead, 2010). CFCs have a strong bond of carbon-halogen which may lead to the formation of dioxin. Therefore thermal plasma aids in the destruction of such gaseous waste. Some catalysts (e.g. $TiO_2$) used with the thermal plasma process, enhance the destruction of CFC. Another pollutant dichloromethane is a VOC which required lower electric power when treated with a plasma-catalytic process than in thermal catalysis or non-catalytic plasma process. It increased the

**Table 2**

Product gases from plasma-catalytic processes.

| Feed Type | Feed | Type of set-up | Catalyst | Carrier gas | Temp., °C | Frequency, Hz | Product gases | Gas yield (%) | Effect of catalyst and Plasma | Ref. |
|---|---|---|---|---|---|---|---|---|---|---|
| Gas | $CO_2$ | DBD plasma reactor | Ni–$CeO_2$/$Al_2O_3$ | – | – | 52000 | $CH_4$ | – | Plasma lowers energy barrier and activate $CO_2$ at low temperature while Ni improves the $CO_2$ conversion | Biset-Peiró et al. (2020) |
| Liquid | VOCs (Benzene, Toluene) | Non-thermal plasma-driven catalyst reactor | $TiO_2$, ϒ-$Al_2O_3$, Zeolites + Metal catalysts | $N_2$, $O_2$ | 100–250 | 500 | CO, $CO_2$ | – | Initial conversion of benzene is influenced by plasma, not catalyst | Kim et al. (2006) |
| Liquid | Toluene | DBD plasma catalytic system | Ni1Al3 | $N_2$, $H_2O$ | 300 | 10000 | $H_2$, CO | – | – | Liu et al. (2018) |
| Liquid | Isooctane | Corona discharge plasma | Chemical catalyst | $N_2$, $O_2$ | 400–800 | 1000 | $H_2$, CO, $CO_2$, $CH_4$ | – | Hydrogen yields are more due to the catalysts than plasma corona discharge | Sobacchi et al. (2002) |
| Solid | Polypropylene | DBD plasma pyrolysis | ZSM-5 zeolite | Ar | 500 | 5000–25000 | $H_2$, $CH_4$, BTX, $C_2$–$C_4$ | 47 | Plasma catalysis increases the gas and oil yield. Plasma enhances $H_2$ formation. | Xiao et al. (2022) |
| Solid | HDPE | Two-stage pyrolysis-plasma/catalysis reactor | Ni/$Al_2O_3$ | $H_2O$ | 500 | 1500 | $H_2$, CO, $CO_2$, $CH_4$, $C_2$–$C_4$ | 19.8 | Catalyst improved overall gas yield than plasma alone process. Plasma and steam enhances $H_2$ formation. | Aminu et al. (2020) |
| Solid | HDPE | DBD discharge plasma pyrolysis | Ni/MCM-41 | $N_2$, $H_2O$ | 500 | 1500 | $H_2$, CO, $CO_2$, $CH_4$, $C_2$–$C_4$ | 33.3 | Various catalysts have different effects on gas yield. Catalysts with high surface area improve the plasma discharge. Surface chemistry of the materials in plasma zone and catalytic zone impact the hydrogen yield. | Aminu et al. (2022) |
| | | | Ni/$BaTiO_3$ | $N_2$, $H_2O$ | 500 | 1500 | $H_2$, CO, $CO_2$, $CH_4$, $C_2$–$C_4$ | 9.5 | | |
| | | | Ni/$Al_2O_3$ | $N_2$, $H_2O$ | 500 | 1500 | $H_2$, CO, $CO_2$, $CH_4$, $C_2$–$C_4$ | 20.3 | | |
| | | | Ni/Y-zeolite. | $N_2$, $H_2O$ | 500 | 1500 | $H_2$, CO, $CO_2$, $CH_4$, $C_2$–$C_4$ | 29.1 | | |
| Solid | LDPE | Microwave-induced pyrolysis | ZSM-5 | $N_2$ | 501 | 2.5 GHz | $H_2$, $CH_4$, $C_2H_6$, $C_2H_4$ | 68.22 | Along with temperature, reactant to catalyst ratio played an important role on the product distribution. | Zhang et al. (2015) |
| Solid | Wood pellets | Two-stage pyrolysis-plasma/catalysis reactor | Ce/Ni–$Al_2O_3$ | $N_2$, $H_2O$ | 600 | 1500 | $H_2$, CO, $CO_2$, $CH_4$, $C_2$–$C_4$ | 15.63 | Ni based support materials improved $H_2$ and CO yields. Plasma improves reactive species in the reactor hence improves selectivity and conversion. Some of the catalysts may enhance the plasma discharge by increasing the electron energy temperature. | Blanquet and Williams (2021) |
| Solid | Bio feedstock (α-cellulose) | DBD plasma gasification + catalysis | Co/ϒ-$Al_2O_3$ | Ar | – | 22000 | $H_2$, CO, $CO_2$, $CH_4$, $C_2$–$C_4$ | – | Gasification and plasma catalysis together doubles the syngas ratio than in gasification only. | Craven et al. (2020) |

energy efficiency of the process by decreasing the temperature requirement for decomposition (Whitehead, 2010). Plasma and catalyst jointly increase the electric field which helps in enhancing the conversion of waste to gases (Aminu et al., 2022). Another way of increasing conversion in some cases is by increasing the discharge length, which ultimately increases the reaction time (Zhou A. et al., 2018). Plasma processes can be affected by the electrical properties of catalysts because of the synergy between plasma and catalyst surface (Kim et al., 2015). Some of the major limitations of the studies carried out on plasma catalytic processes are discussed. The cost of the plasma-catalytic pyrolysis is higher than conventional pyrolysis due to high cost of energy and cost of catalyst. However, it is economical in comparison to catalytic pyrolysis due to large requirement of catlaysts in the later process (Xiao et al., 2022). Another major limitation is scale-up of set-up due to limited plasma discharge zone in the reactor and inadequate approach of

fundamental system and process engineering (Munir et al., 2019). Aminu et al. (2020) observed that catalysts like Ni/$Al_2O_3$ show low activity for catalytic non-thermal plasma pyrolysis, as they require high temperature for the activity. Hence knowledge of appropriate catalyst is primely important for a particular feed and process conditions. Also, Information regarding synergy between catalyst and plasma at very high temperature is not well-known.

### 4.3. Catalyst selection criteria for plasma processes

Catalyst selection criteria for plasma processes are not readily available in the literature. However, various catalysts are reported in the literature, which have been used for the plasma and non-plasma processes for waste treatment at different operating parameters. The literature data may help in selecting the suitable catalyst for a specific feed

and reaction condition. As the temperature increases degradation in the catalytic properties like dispersion and sintering becomes fast. Hence, catalyst selection is critical for high-temperature applications. Various types of catalysts are already discussed in previous section 3 of this paper. Most of the catalysts (i.e. zeolites as well as metal oxides), which have active metal(s) on catalyst support are precursor catalysts which have a substantial effect on the performance of catalytic processes. Such catalysts need to be activated after some cycles, while catalysts in the form of inorganic salts can be directly used in the process. The type of plasma (i.e. non-thermal or thermal) is the key factor in the selection of catalyst as the temperature plays a major impact on the catalyst. Some catalysts cannot withstand very high temperature, on the other hand, some catalysts favour conversion at high temperature. Although it was observed that plasma catalysis does not offer synergistic effects at very high temperature (Neyts and Bogaerts, 2014). Plasma also has a significant effect on the catalyst surface. Plasma gasification with an $O_2$ supply can help in the regeneration of deactivated metal oxide catalysts due to the presence of oxygen. While the same with air supply may form $NO_x$ on the catalysts surface due to the presence of $N_2$. Thermal plasma has high excitation energy which transmits to the surface of the catalyst. On the other side, the ionized portion in non-thermal plasma is small. Due to the same, the catalyst should have a larger surface area to maximize the interaction. Catalysts with a high microporous area are generally favourable for $H_2$ selectivity.

### 4.4. Cost analysis of plasma-catalytic pyrolysis

In plasma-catalytic pyrolysis methods, the cost of energy required is high due to plasma and the catalyst addition. Hence, the economic analysis is required to justify the viability of this method. The cost calculations include mainly the costs of electricity, water, labour and catalyst. It is to be noted that the cost of catalyst may vary widely depending upon the type of catalyst used in the system. The breakdown of cost analysis from literature is as shown in Table 3.

The wide difference in the cost for different processes is due to the different assumptions, plant capacities, region, feed type, catalyst type, no. of labours, payback period and the years of costing reported in various literature (inflation). It is observed that between catalytic processes, operating cost in case of plasma-catalytic pyrolysis is higher than catalytic pyrolysis, while cost of catalyst is higher in the later case.

### 5. Conclusion and future prospective

This paper summarizes the current status of the plasma-catalytic processes. It is a promising technology as it has a lower formation of harmful compounds and higher energy efficiency than the catalytic pyrolysis and plasma-only processes. This process provides the prospective to deal with the challenges faced by catalytic pyrolysis like deactivation of the catalyst and wax formation. Gas yield in this process varies from 9% to 68% at moderate temperatures around 500 °C which is higher compared to catalytic pyrolysis. Various factors affect the hydrogen yield in the plasma-catalytic process such as plasma power, properties of catalyst support material, properties of loaded metal, and surface chemistry of the catalyst as a whole. A synergy between thermal plasma technologies and catalysts are required to be studied for waste-to-energy conversion. Implementation of the plasma-catalytic process for waste management is a challenging task due to the operating cost associated with the process. Very limited research work is carried out in the area of thermal plasma-catalytic processes, which seem to be capable technology for liquid and solid waste treatment. Thermal plasma-catalytic process needs to be studied with an approach to energy saving and hydrogen selectivity. The experimental studies on the properties of active sites are essential for the derivation of core kinetics of the thermal plasma-catalytic process. The synergistic effect of catalyst and plasma also requires to be studied in detail for the appropriate development of catalysts, which can work well with plasma. There is a

**Table 3**
Cost analysis of different waste-to-energy processes.

| Basis: | Plasma catalytic Pyrolysis (100 tpa) | Catalytic Pyrolysis (100 tpa) | Plasma Pyrolysis (1200 tpa) | Gasification (720 tpa) |
|---|---|---|---|---|
| | Xiao et al. (2022) | Xiao et al. (2022) | Tang et al. (2010) | Indrawan et al. (2020) |
| Cost Headings | | | | |
| (1) Capital cost (one time) | Not available | Not available | 300000 | 112500 |
| (2) Variable cost | | | | |
| Feedstock | 6000 | 6000 | Included in operating cost | 24508 |
| Catalyst | 141354 | 1570600 | – | – |
| Operating cost | 189955 | 48330 | 72000 | 47268 |
| Utilities | 9072 | 9072 | 50400 | |
| Total variable cost | 340381 | 1628002 | 122400 | 71777 |
| (3) Fixed cost | | | | |
| Maintenance | – | – | – | 32500 |
| Salaries for manpower | 43516 | 36264 | 2400 | |
| Total fixed cost | 43516 | 36264 | 2400 | 32500 |
| Total processing cost ((2)+(3)) | 383897 | 1664266 | 124800 | 104277 |
| Total processing cost per tonne | 3839 | 16643 | 104 | 145 |

Note: cost is in USD.

vast scope in the field of plasma catalysis in the field of waste valorization as it is a path towards sustainable development.

### CRediT authorship contribution statement

**Kangana P. Bhatt:** Writing – review & editing. **Sanjay Patel:** Conceptualization, Review. **Darshit S. Upadhyay:** Review. **Rajesh N. Patel:** Supervision.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

No data was used for the research described in the article.

### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2023.118335.

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