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Chemical Recycling Pathways for Agricultural Plastics - A Comparative Technology Assessment

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Abstract – Tertiary (chemical) recycling is the process of breaking down plastic polymers into their individual monomers, which are then reconstructed to create new polymers to produce fresh plastic goods. Tertiary recycling includes methods such as cracking and gasification. Chemical recycling techniques, such as catalytic cracking, pyrolysis, microwave-assisted pyrolysis (MAP), plasma pyrolysis, may effectively extract polymers from mixed rubbish and municipal solid waste. This paper provides a comprehensive analysis of four main types of plastics recycling and eight chemical recycling systems, with a specific focus on cracking and gasification methods. The selection of technologies was based on a thorough examination of existing literature, and their evaluation was conducted by considering significant performance characteristics, peer-reviewed scientific publications, information supplied by technology developers, and expert interviews. The primary objective of the research is to conduct a comparative analysis of technologies that use publicly accessible data. The research only examines tertiary (chemical) recycling techniques, excluding other approaches like bio recycling. The findings underscore the potential of chemical recycling to provide a self-sustaining framework for plastics and examine the challenges and benefits linked to different recycling methods.

Keywords – Catalytic Cracking, Chemical Recycling, Microwave-Assisted Pyrolysis, Chemical Recycling Systems, Pyrolysis, Plasma Pyrolysis.

I. INTRODUCTION

Recycling is classified as the third level in the waste hierarchy, as outlined by the European Waste Framework Directive [1]. Waste preparation and prevention for reuse are positioned higher than this. While it is possible to reuse plastic items after processing, this practice is uncommon and not the norm. Primary recycling, often known as the reuse of manufacturing waste such as cuttings, is considered the most effective way for recycling plastic items. The outputs derived from mechanical processing techniques serve as primary resources to produce fresh plastic goods. Nevertheless, mechanical recycling methods (also known as secondary recycling) have constraints when dealing with plastic garbage that is heavily mixed and polluted. Chemical recycling technologies may be used to effectively bridge the gaps in the cycle. Combustion, especially when oxygen enrichment or oxyfuel is used, may also contribute to the separation of CO_2 more effectively. Nevertheless, incineration is classified as "alternative recycling" in ranks fourth and the waste hierarchy, before disposal.

Plastics Chemical recycling is often understood as an industrial procedure that entails breaking down complex organic macromolecules into monomers, simpler organic molecules, or their individual components [2]. These resulting products may be used, either partially or whole, in the production of new plastics. Breakdowns may be categorized as gasification, hydrolysis, pyrolysis, solvolysis, catalytic cracking, or hydrocracking, depending on the specific chemical process or principle involved. Although incineration is a chemical process, it cannot be considered recycling until most incinerators recycle the resulting combustion products, which mostly consist of CO_2 and H_2O.

Chemical recycling processes may be used to produce fuels such as petrol and diesel, a kind of recycling that is both practical and prevalent in some regions, such as India [3]. Chemical recycling necessitates far more stringent technological prerequisites in comparison to mechanical recycling. However, in contrast to mechanical recycling, it offers versatility in the new plastic production and even allows for the plastics production from recycled raw materials with the same level of quality as new goods (“virgin quality”). The subsequent sections will comprehensively investigate both pyrolysis and solvolysis. **Fig. 1** illustrates the whole life cycle of plastics, including the significance of reuse and many recycling methods.

This research aims to provide a comprehensive analysis of different plastic recycling technologies, focusing particularly on chemical recycling methods like cracking and gasification. The selection of technologies was made after a thorough examination of existing literature and expert interviews, ensuring the information's trustworthiness and up-to-date nature. The assessment aims to provide a comparative analysis of the performance characteristics of various technologies in a manner that is easily understandable by the general audience. The focus on various types of plastic waste generated by municipalities is essential, as it represents a significant amount of plastic refuse that need effective recycling methods.

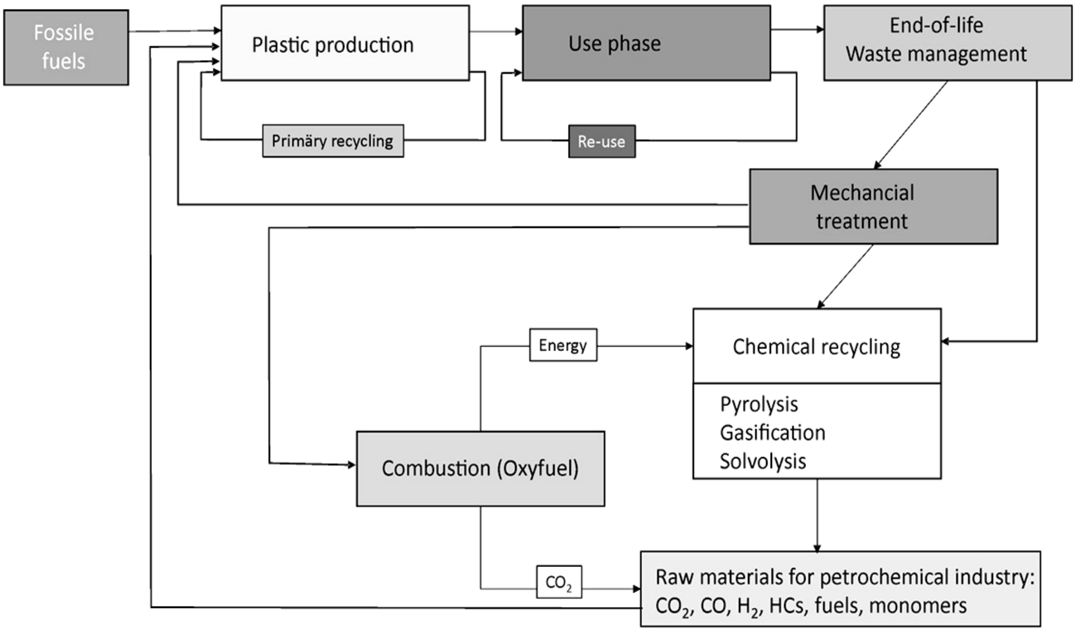

Fig. 1: Chemical Recycling as a Component of a Circular Economy for Plastic Products

The rest of the paper has been organized as follows: Section II presents a discussion of technology identification and evaluations for chemical recycling. Section III reviews the availability technologies for plastic recycling and their characteristics. This section reviews key concepts of conventional pyrolysis (thermal cracking), and catalytic cracking. Section IV presents a summary of the findings obtained in this article.

II. TECHNOLOGY IDENTIFICATION AND EVALUATION FOR CHEMICAL RECYCLING

Our review considers four main types of plastics recycling and grouped two forms of chemical recycling systems, as shown in **Fig. 2**. The selection of technologies was first made based on a thorough examination of existing literature. Chemolysis was not included in the study due to its limited applicability for treating homogeneous plastic trash, while we are specifically addressing heterogeneous municipal plastic garbage. Therefore, our research specifically concentrates on the methods of cracking and gasification. The technologies are evaluated based on critical performance characteristics. The rating depends on peer-reviewed technical articles and data obtained from developers of technology through their official webpages. The expert interviews enhance the results.

Every method is assessed and contrasted according to three crucial parameters: extent of polymer degradation, susceptibility to feedstock contamination, and process temperature. Due to large-scale projects large limited number now in operation, there is a scarcity of actual technical data available. This poses a difficulty in comparing the technologies. The primary objective of our article is to conduct a comparative analysis of technologies using data that is accessible to the public.

Increased temperature facilitates enhanced polymer degradation and improved material purity of the treated substance. According to Jacobs and Swink [4], this offers a greater opportunity to recover materials and increases the range of goods that may be created, resulting in more flexibility for the portfolio. The temperature has a crucial role in the treatment of mixed waste plastic. Polymers with varying chemical compositions have distinct melting points and need somewhat varied conditions for their decomposition into monomers. The recommended process temperature is appropriate for the element with the greatest melting point. However, this may result in overheating and deterioration of components with lower melting points. Elevated operating temperatures lead to increased expenses and need more energy input.

The sensitivity of a technology to the quality of the waste increases as the process temperature decreases [5]. Chemical recycling necessitates more meticulous waste separation in the presence of low temperatures. The availability of sophisticated separation methods now means that further waste separation is no longer seen as a hindrance to the widespread adoption of chemical recycling systems that operate at lower process temperatures. Nevertheless, this might lead to supplementary expenses apart from the expenditures associated with the recovery facility, and will need more attention to logistical matters.

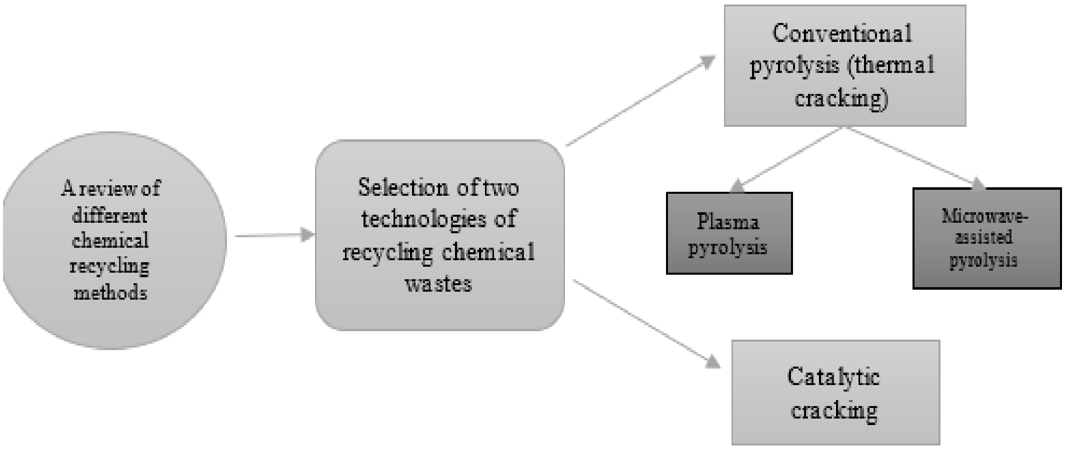

Fig. 2: Determination of appropriate chemical methods for recycling home garbage

The degree of susceptibility of various technologies to waste contamination directly influences the level of logistical efforts required [6]. Greater technological sensitivity necessitates more meticulous waste separation. Process temperature may be raised to decrease the sensitivity of a technology. If high polymer degradation process can be accomplished, then the mechanism is appropriate for novel polymers manufacturing. This may decrease the need for new polymers and help in achieving a closed-loop system for plastics. A more comprehensive analysis of plastic input reduces the need for additional processing stages prior to the production of new plastics. Evaluating the probable technology contribution to reaching circular economy aims requires careful consideration of the probable breakdown of polymers.

Table 1: presents a model for evaluating the TRL (Technology Readiness Level) of plastic recycling systems

Technology Designers	Comment	TRL
Large-size enterprises	In commercial & In development function	8–9
SMEs	In commercial operation & Successfully sold	8–9
SMEs	In development	4–7
Research and development firms		1–5

The Technology Readiness Level (TRL) evaluation is conducted by comparing the kind and scale of the organization responsible for producing the technology, as shown in **Table 1**. Technologies that are merely at the academic or research stage have a low TRL. Upper level TRL stands for Technology Readiness Level and is used to categorize small and medium-sized firms that have either prototype or fully developed technology that can be effectively expanded. Technologies created by major corporations with substantial research and development capabilities and the ability to take advantage of economies of scale have the greatest Technology Readiness Level. The latter group is the more likely to achieve success in the immediate future. The TRL stages are allocated to initiators based on the TRL scale provided by Olechowski, Eppinger, and Joglekar [7].

III. AVAILABLE TECHNOLOGIES FOR PLASTIC RECYCLING AND THEIR CHARACTERISTICS

Plastics recycling may be classified into four main types, as shown in **Fig. 3**.

- Primary recycling, also known as closed loop recycling, refers to the mechanical reprocessing of single, pure plastic materials to produce a same quality product.
- Secondary recycling refers to a process of mechanical recycling that results in a decrease in the quality of the recycled material.
- Tertiary recycling refers to the process of feedstock recycling or chemical recycling, when polymers are broken down into monomers and the recycled material is recovered with high quality.
- Quaternary recycling refers to the process of incinerating garbage in order to recover energy.

Municipal solid waste (MSW) is not compatible with primary recycling due to its inability to handle contaminated mixed plastics. Sorting in secondary recycling leads to significant material losses. Tertiary recycling enables the restoration of material quality, facilitates the manufacturing of raw materials, promotes the reuse of plastics, and aligns with the present goal of achieving a closed-loop system for plastics. Plastic trash has significant potential for the generation of chemicals and fuels. Ultimately, the quaternary process reduces the longevity of the material. While it is used in several places, it is considered an intermediate measure since it does not completely adhere to the developing ideas of a circular economy.

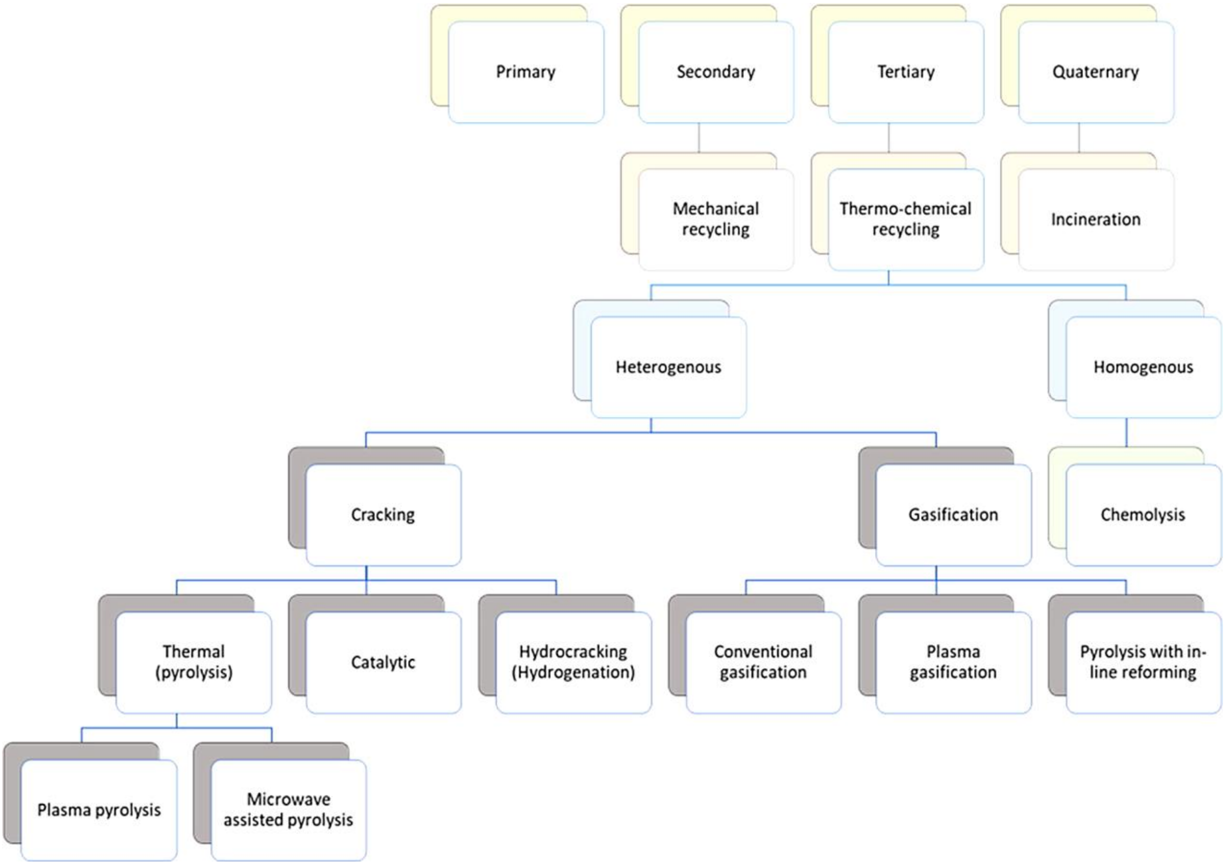

Fig. 3: Recycling Pathways for Solid Plastic Trash

Chemical recycling systems specifically designed for processing plastics in household garbage streams are shown in the color gray. This research specifically examines the tertiary (chemical) recycling technologies, namely cracking and gasification technologies, which are seen as promising solutions for recovering plastics from domestic garbage, as shown in **Fig. 3**. Chemical recycling refers to the procedures that include the chemical decomposition of a plastic polymer into monomers, which are then rearranged to form new polymers for the manufacturing of fresh plastic materials. This study only focuses on a specific sort of recycling, excluding other forms such as bio recycling.

Conventional Pyrolysis (Thermal Cracking)

Conventional pyrolysis [8] is an appropriate approach for processing waste polymers that are challenging to break down into their constituent parts, like multilayered plastic packaging. These are now being burned. The process occurs in the absence of oxygen, and at elevated temperatures, ranging from moderate to high. The primary challenges lie in the intricate nature of the reactions involved and the substantial energy demand associated with the process. The method may be arduous when the polymers are combined with impurities. Pyrolysis has limited liberality to the polyvinyl chloride (PVC) content found in the feedstock. Subsequently, the pyrolysis oil might undergo chlorination, resulting in the formation of chemicals that hinder its use.

Thermal cracking, or pyrolysis [9], involves polymer heating in an inert atmosphere to break the bonds between macromolecules, resulting in the formation of various low molecular weight hydrocarbons. These hydrocarbons include a liquid fraction containing aromatics, paraffins, naphtenes, and olefins, , as well as a solid residue and a volatile fraction. Different cracking processes occur based on the features of the polymer chain, such as the presence of bond dissociation energy, defects, aromatic degree, hetero-arms, and halogens.

In the case of thermal cracking, the polymer is heated in a non-reactive environment with the help of catalysts, which enables the process to occur at lower temperatures. This results in energy and cost savings compared to thermal cracking. Reactions occur in reactors of reduced dimensions and shorter residence durations when catalyzed cracking is used, as a result of increased reaction rates. By carefully choosing the optimal reaction conditions and catalysts, it is possible to enhance the selectivity and productivity of the reaction products. The catalytic cracking limitations arise from the heterogeneous nature of the plastic stream intended for recycling. The catalyst may be rendered ineffective by the certain hetero-atoms presence, like nitrogen or sulphur, which act as catalyst poisons. Furthermore, the presence of chlorine may lead to the formation of hazardous and corrosive chemicals, necessitating the need for separation or dechlorination.

The experiment described by Hossain [10] used a gas metering system to control the flow rates of gas to the desired levels. This was combined with a high-pressure batch reactor to carry out the thermal cracking of petroleum residual oil. **Fig. 4** displays the schematic design of the experimental setup. The system was fed with nitrogen and hydrogen gasses from gas cylinders using pressure regulators, maintaining a consistent pressure. The two streams were merged using a three-way valve. The discharge from the valve was directed towards the reactor. The whole system's connecting pipe consisted of stainless-steel tubing with a diameter of 6.35 mm, which was connected using stainless steel fittings.

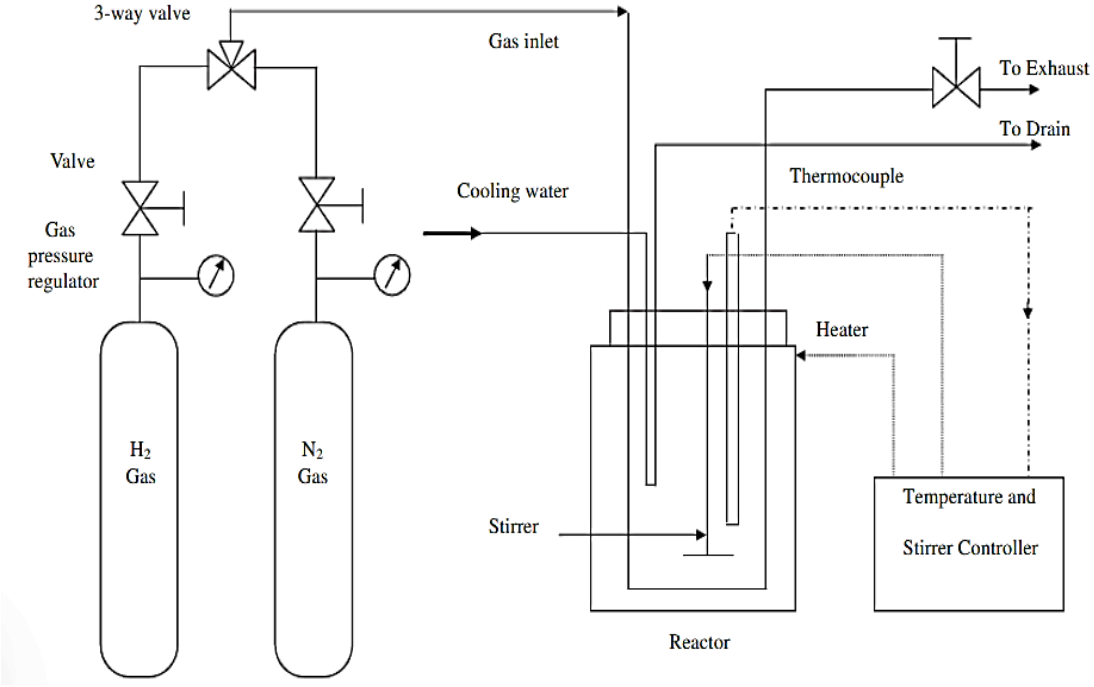

Fig. 4: Experimental Arrangement Used to Thermally Fracture Petroleum Residual Oil

The 300 ml reactor with high pressure capabilities was provided by M/S Parrinstrument business, USA. It can withstand pressures of up to 14 MPa and reach a maximum temperature of 500 °C. The apparatus is furnished with a gas entry valve that is linked to a dip tube, which stretches down to a location close to the base of the bomb cylinder. The configuration enables the injection of gas under the liquid sample's surface. A gas release valve is connected to decrease the pressure of the bomb in the event of overcharging, as well as to remove any gas from the device. A stainless-steel bourdon tube pressure gauge, calibrated from 0 to 14 MPa, is installed on the bomb head along with the gas release valve. A J-type thermocouple is inserted into a thermowell to measure the temperature of the reaction. The internal stirrer has a dependable drive thanks to the motor drive assemblage and magnetic drive correction. The rotational speed of the stirrer may be regulated up to 2500 revolutions per minute (rpm) using a direct current (DC) variable speed controller.

Plasma Pyrolysis

Traditional pyrolysis is combined with the thermochemical characteristics of plasma inorder to convert plastic waste to syngas by plasma pyrolysis. Very high levels of temperatures, varying from 1730 to 9730 °C is reached, causing the decomposition of waste polymers into monomers [11]. The procedure is very efficient, with a duration ranging from 0.01 to 0.5 seconds, contingent upon the temperature of the process and the nature of the waste. The primary constituents of the syngas generated are carbon monoxide (CO), hydrogen (H2), and minute amounts of higher hydrocarbons.

Comparing plasma pyrolysis to conventional pyrolysis has several advantages, including the complete breakdown of polymers. Due to its low tar percentage and high heating value, the produced gas is ideal for producing hydrogen or turbine power. A variety of waste plastic materials may be processed using plasma pyrolysis, which also offers a significant deal of possibilities for producing chemicals or gaseous fuel. The high temperature used in this process inhibits the generation of free chlorine from HCl, a common problem in other chemical recycling techniques, and efficiently breaks down dangerous compounds in syngas.

A system for processing gaseous, liquid, and dispersed hazardous, chemical, and pharmaceutical waste is built around the PR-50 Plasma Pyrolysis Reactor [12]. The system has a maximum power consumption of 100 kilowatts. The byproducts, such as synthesis gas and powder, obtained from the process may be used in alternative applications. The electric energy produced by this process may be used for a multitude of reasons. Approximately 33% of this amount of energy is sufficient to operate the reactor. In order to guarantee the safe functioning of a reactor, we construct a robust and tightly sealed enclosure using several layers of stainless steel. Additionally, we include a water jacket to effectively prevent any pollutants from escaping into the environment. The plasma pyrolysis reactor is equipped with three plasma torches that generate a unique vortex to guarantee high-quality pyrolysis of waste materials. **Fig. 5** illustrates the fundamental configuration of the plasma pyrolysis reactor.

Technology for thermal plasma is a well-recognized technique used in synthesis of substance and metallurgical processing. Currently, the technology`s primary use is focused on the disposal of dangerous waste rather than its recycling, mostly owing to economic and regulatory considerations. The investigation of waste plastics plasma pyrolysis has only been conducted at a laboratory scale. Before the technology can be commercially accessible, there are still other technological obstacles that need to be resolved in order to fulfill waste management criteria. The obstacles vary based on the particular plasma technique. Cai and Du [13] suggest that there is significant interest in identifying waste treatment appropriate plasma technology and propose that spout-fluid bed reactor technology might be a promising option.

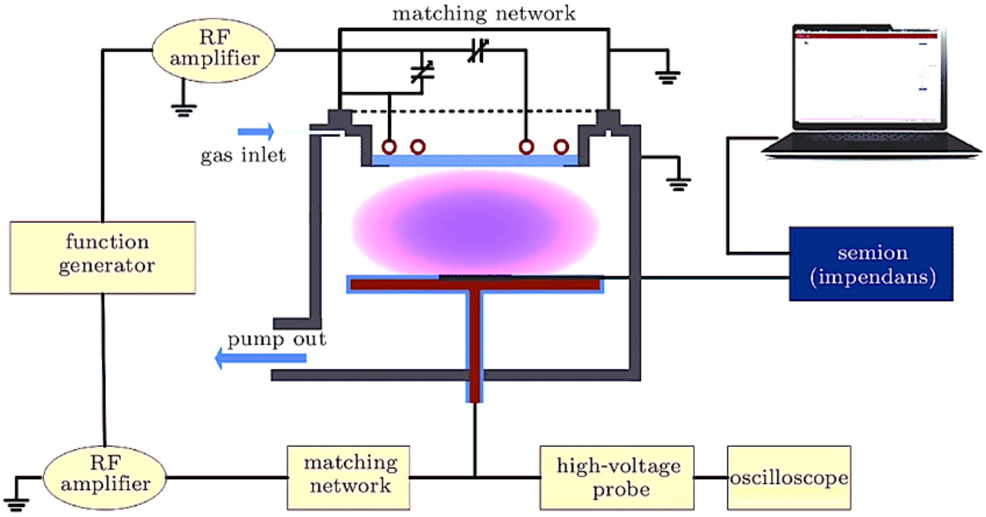

Fig. 5: The Plasma Reactor Equipped by REFA Diagnostic Model

Microwave-assisted pyrolysis

Plastic waste microwave-assisted pyrolysis (MAP) process is enhanced when an extremely microwave-absorbed dielectric substance is combined with the plastic [14]. The microwaves heat the polymers by conduction. Microwave radiation sources enable the electrical energy efficient conversion into heat and its transport to a load, resulting in high heating rates and temperatures.

The outcomes of MAP of various biomass feedstocks are influenced by conditions like the specific type of biomass waste (e.g., sludge wastes, agro-industry residues, forest biomass) and the characteristics of the feedstock (e.g., HHV (higher heating value), ultimate and proximate analysis components and moisture content, particle size, lignocellulosic analysis fractions, LHV (lower heating value), etc.). The MAP of several biomass materials, such as bamboo leaves, maize stover, rice husk, straw, sugarcane peel, bagasse, and spent coffee grounds, was achieved using similar treatment conditions. The investigation indicated that all seven feedstocks exhibited similar and satisfactory yields. However, the amount of both liquid and gas produced is highly dependent on the kind of feedstock used. The maize stover had the greatest gas production (40 wt%) but the minimum liquid production (25 wt%). In contrast, rice husk had the minimum gas emission (30 wt%) and the greatest liquid yield (48 wt%).

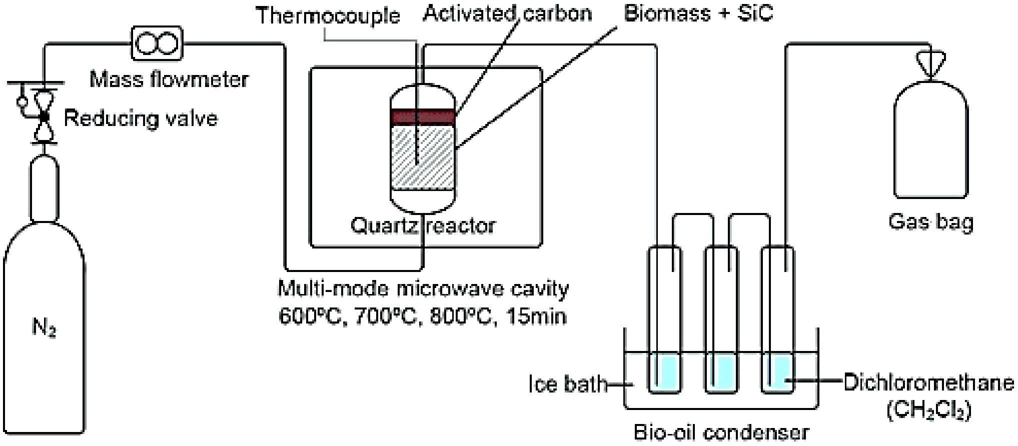

Fig. 6: The Process of Using Microwaves for Pyrolysis of Biomass, which is Then Combined with Reforming

The study performed Microwave-assisted pyrolysis (MAP) in [15] utilizing a multi-mode microwave cavity (2.45 GHz) with a highest power output of 3 KW. A K-type thermocouple, encased in a Niobium alloy tube, was employed to measure operational temperatures. A custom-made quartz tube, with a diameter similar to that of the vertical tube furnace typically used in conventional pyrolysis, served as the microwave reactor. **Fig. 6** displays the experimental setup. A 5.0 g biomass sample, which had been air-dried, was used. Furthermore, activated carbon`s additional 5.0 g was introduced into the SiC-biomass mixture, maintaining a mass ratio of 5/50. Subsequently, the blend was introduced into a quartz tube and carefully situated within the pyrolysis reactor. The reactor was heated to either 800, 700, 600°C when operated in a temperature-controlled mode. The temperature regulation has been mechanized to avoid surpassing the specified value by permitting the microwave generator to be turned on and off. A nitrogen gas flow rate of 200 ml/min was utilized to generate an inert ecosystem. Following the completion of the pyrolysis process, the char and SiC components were segregated via a sieve-based technique. The ice bath was used to expedite bio-oil condensation, then liquified in dichloromethane. Concurrently, the gas generation was gathered with a gasbag.

Catalytic Cracking

Catalytic cracking is a flexible conversion method that may be used to many feedstocks, such as gas oil, heavy crude oil, and residuum. Catalytic cracking is a refining technique that is similar to thermal cracking, but it differentiates itself by using a catalyst that is theoretically not exhausted throughout the process. One of the many practical uses in a refinery is the use of a catalyst to improve both the efficiency of the process and the variety of products generated.

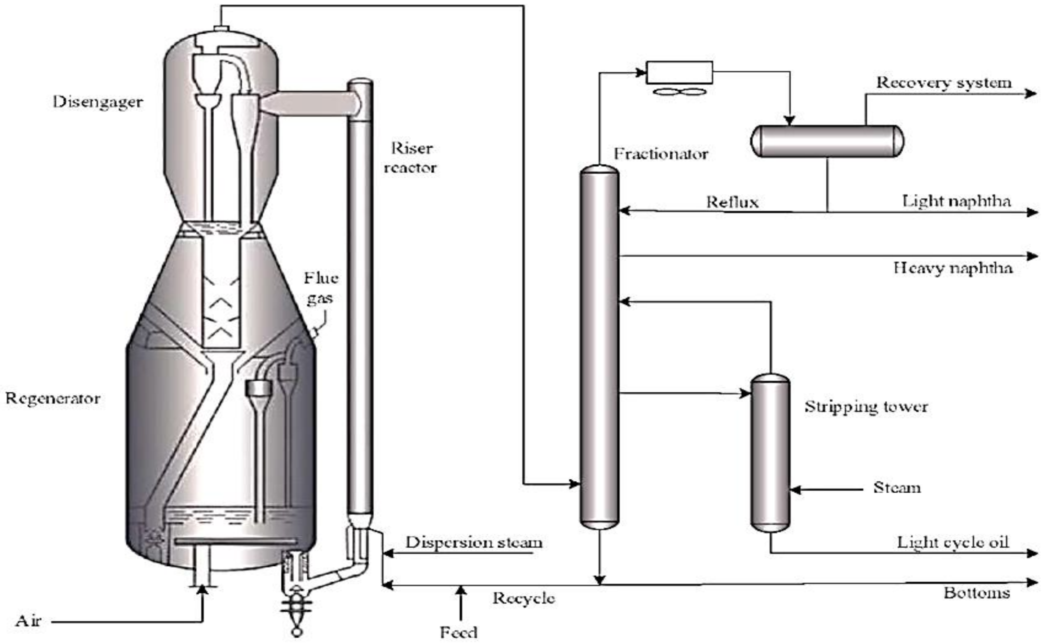

Fig. 7: Standard Procedural Illustration of a Fluid Catalytic Cracking Unit

In the conventional industrial procedure of catalytic cracking, a catalyst is used to chemically react with a feedstock, typically consisting of a gas oil fraction. The occurrence of this reaction is contingent upon certain conditions of temperature, pressure, and residence time. Using this technique, a substantial proportion (over 50%) of the raw material is converted into gasoline and other highly volatile compounds, often in only one operation. Nevertheless, while undergoing the cracking process, a carbonaceous material is generated and accumulated on the catalyst, resulting in a notable decline in its efficiency. Hence, it is essential to eliminate this deposit. The carbonaceous deposit is created by the heat decomposition of bulky, polar molecules (such as asphaltene and resin constituents) in the unprocessed substance. The removal of the deposit from the catalyst is often accomplished by burning in the presence of air until the catalyst's activity is reinstated.

The fluid catalytic cracking (FCC) [16] process is the primary element of a modern refinery that aims to maximize the production of gasoline. Out of all the steps in the refinery process, this specific step shows the most potential for increasing profitability. Even a little improvement that results in increased gasoline output might provide substantial economic advantages. The FCC method employs carbon rejection in a continual fashion to increase the H/C ratio. It is used to convert hydrocarbon fractions that have high boiling temperatures and large molecular weights. These fractions usually consist of a combination of heavy straight-run gas oil, light vacuum gas oil, and heavy vacuum gas oil. The goal is to generate higher-value products, such as olefinic gases, gasoline, and other derivatives. A typical modern FCC unit consists of the following phases (see **Fig. 7**): The process consists of two sequential stages: (a) The feed is heated and mixed with the recycled slurry oil collected from the lower part of the column of distillation, and (b) The resulting mixture is introduced into the catalyst riser, where it undergoes vaporization.

(c) The vaporized feed undergoes molecular fragmentation upon contact with the high-temperature powdered catalyst from the regenerator. (d) The fragmented product vapors are isolated from the discarded catalyst by being sent through a sequence of two-stage cyclones. (e) Employing steam to remove hydrocarbon odors from the used catalyst before reintroducing it to the regenerator. The process of regenerating the used catalyst by using compressed air to combust the coke deposits that have collected on its surface. The process is thermally exothermic and emits a substantial amount of heat. (f) The regenerated catalyst absorbs a portion of this heat, subsequently providing the required heat for vaporizing the feed and facilitating the endothermic cracking processes in the catalyst riser. (g) Diverting the hot flue gas, which is at a high temperature, from the regenerator through several sets of cyclones in order to remove catalyst particles that are being carried by the gas. (h) The fragmented product vapors are efficiently isolated from the reactor and any catalyst particles that may be transported by using a cyclone. Subsequently, these vapor fractions are sent to the recovery section of the FCC unit to meet the requirements of the product stream.

The addition of a catalyst to the pyrolysis process has the potential to reduce energy consumption and production costs by lowering the required process temperature. Utilizing a catalyst allows for a decrease in the operating temperature of the process to a range of 300-350 °C, as opposed to the higher temperature of about 450 °C needed for pyrolysis. Utilizing a catalyst may also augment the generation of things with larger economic value. Catalysts have been used to augment the caliber of goods and generate oil with analogous attributes to conventional fossil fuels. Most studies on plastic catalytic cracking have mostly focused on using unpolluted polymers, since the presence of contaminants in mixed waste plastic streams might potentially affect the process.

Catalytic cracking, when using the suitable catalyst, provides a higher oil yield and a reduced operating temperature in comparison to the conventional cracking technique for the majority of polymers. Catalysts expedite the reaction while preserving their initial condition throughout the process. The process of plastic waste cracking has the potential to reach a conversion rate of up to 100%, leading to an oil yield that may range from 86% to 92% [17]. The main obstacle is the deactivation of the catalyst due to the presence of nitrogen and chloride components in the untreated waste stream, which clog the catalyst's pores with inorganic compounds. Therefore, it is often essential to do waste pre-treatment as a preventive action.

The effectiveness of product selectivity and distribution is optimized by catalysts, particularly in the domain of petrochemical chemicals and automotive fuels. The catalytic degradation of polymers and hydrocarbon catalytic cracking in petroleum refineries both involve comparable reaction pathways and catalysts. Various commercial catalytic cracking methods are used in the industry. Multiple multinational businesses worldwide interact with technology. The maximum waste plastic liquefaction facility in Japan is jointly operated by Sapporo Plastics Recycling and Toshiba, making it one of the globe`s highest catalytic cracking facilities. The factory changed 15,000 tons of diverse plastic waste into light oil, which was used as a primary ingredient for manufacturing new plastic products, medium fuel oil with characteristics like heavy oil, and diesel employed for power generation [18]. Nevertheless, Sapporo Plastic Recycling ceased its participation in the business in 2010 due to financial challenges.

IV. CONCLUSION

This article provides a comprehensive analysis of several methods used in the recycling of plastics, with a particular emphasis on the cracking and gasification processes. The selection of technologies was based on a thorough examination of existing literature, and the evaluation was conducted using crucial performance metrics, peer-reviewed scientific papers, material supplied by technology creators, and expert interviews. The research highlights the importance of temperature in the processing of mixed waste plastics, since various polymers possess distinct melting temperatures and need certain stages of decomposition. This research examines at the relationship between the amount of logistical work needed and a technology's vulnerability to waste contamination. As shown, more precise waste segregation is required for devices with greater sensitivity. The study highlights the effectiveness of chemical recycling, namely the cracking and gasification processes, in removing plastics from municipal waste. One method involves breaking down plastic polymers chemically into monomers, which may then be put back together to create new polymers for making new plastic items. A thorough analysis is conducted on several cracking techniques, such as catalytic cracking, microwave-assisted pyrolysis, plasma pyrolysis, and conventional pyrolysis. Every approach has pros and downsides, taking into account variables like temperature, energy requirements, pollutants, and product yield. In the end, this study offers insightful information on various plastic recycling techniques and their potential to create a self-sufficient plastic system. It highlights the need for more research and development in this field in order to improve the efficiency and robustness of plastic recycling methods.

CRediT Author Statement

The author reviewed the results and approved the final version of the manuscript.

Data Availability

The datasets generated during the current study are available from the corresponding author upon reasonable request.

Conflicts of Interests

The authors declare that they have no conflicts of interest regarding the publication of this paper.

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Competing Interests
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