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Photocatalytic assisted microwave-based plasma pyrolyser: A solution for COVID-19 related wastes?[†]

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The aim of this mini-review is to explore the potential of indigenous photocatalytic assisted microwave plasma based pyrolyser for COVID-19 related wastes (gloves, masks, bottles, personal protective equipment (PPE), etc.) conversion into liquid oil. Plasma pyrolysis can provide solution for complete pyrolysis of typical hospital wastes not limited to PPE wastes. Literature survey reveals that pyrolysis of polystyrene (PS) plastic waste feedstock can yield up to 80% of pyrolytic oil including styrene, ethylbenzene and toluene with their physico-chemical properties such as density, viscosity, flash point, freezing point, pour point, and high heating values very similar to the conventional diesel and therefore, possess potential as an alternative source for power generation. It is suggested that char produced from pyrolysis has requisite properties for electro-oxidation of CO_2. Moreover, if such a system could be integrated with a microwave heating process having absorbents like silicon carbide and different catalyst combinations, up to 32% energy saving could be done in the process that would be rapid, uniform and energy-efficient.

Keywords: COVID-19 wastes, PPEs, microwave, pyrolysis, catalysts, fuel, oils, char.

1. Introduction

The increasing rates of plastic production and waste plastic accumulation pose a serious challenge to the society, environment, and economy. A wide range of plastics are available in the modern world to solve our day to day problems due to their excellent durability characteristics, especially in the medical field. This has its own merits such as inexpensive cleaning and sterilization, hygienic packaging ensuring food safety, reduction of pathogen transmission, disposal of medical wastes, etc.[1]. In 2018, global production of plastic was 358 million tons[2]. Eriksen *et al.*[3] and Lebreton *et al.*[4] have reported accumulation of 5 trillion plastic pieces in the water bodies, with average annual accumulation of 1.2–2.4 million tons. Mismanagement and improper use/disposal of plastics, often lead to environmental contamination, and various natural calamities by creating imbalance of ecological communities, etc.[5,6]. The Central Pollution Control Board[7], GoI has issued latest guidelines for disposal of COVID-19 waste, and has directed that discarded masks, gloves and PPE used during the current pandemic, need to be shredded in separate bags or bins for 72 h minimum resistive time as per the disposal standard protocols.

Other than PPE waste generated by COVID patients like empty juice and water bottles, food waste, and other solid wastes also collect in closed bins or bags to prevent the transmission of SARS-CoV-2 virus. World Health Organization (WHO) has advised the frequent usage of PPEs (especially gloves, masks, etc.) to combat the effects of the current global pandemic situation created by the deadly virus SARS-CoV-2 since March, 2020[8]. The estimated requirement per month to deal with such an emergency situation is expected to be 89 million medical masks, 76 million gloves and 1.6

[†]**Invited Lecture.**

million eye protective glasses, while half a million sets of PPE have already been shipped to 47 countries by WHO[9,10]. To meet the rising demand, industries are expected to increase their production by 40% for uninterrupted supply chain. Often, the mismanagement of COVID-19 waste leads to disturbance of biological eco-system. Huge quantities of waterlogged COVID-19 related plastic waste have been observed in water bodies, creating extra pressure on routine waste management practices[11].

Therefore, this calls for an urgency to develop more responsible and sustainable strategies for waste recycling and disposal, particularly in the healthcare sector. Current mechanical recycling process of plastic waste is limited to segregation/pre-treatment followed by degradation. In developed countries, land filling post incineration is the most widely used method for such waste[12,13]. However, incineration has its own demerits like gas emission, high construction cost of the incinerator, etc[1]. Moreover, prioritization of human wellbeing over environmental health have created a barrier for implementation of policies reinforcing the ban of single use plastics in healthcare. As a result, the cost of manufacture of virgin plastics from fossil fuels has plummeted as compared to its recycling cost[14]. Several methods are available to convert plastic into fuel such as gasification, pyrolysis, plasma catalysis, etc.[15]. Among these, pyrolysis is an effective method for converting waste plastic into fuels with three end products (pyrolytic oil, gases and char[16]). It depends on different process influening parameters not limited to moisture or toxic elements presence, reaction time, temperature, feed composition, and heating rates[17].

Research investigators have successfully converted waste plastic into fuel in small scale pyrolytic reactors[18–24]. Although, aromatic compounds recovery mainly toluene, ethyl-benzene, and styrene from produced pyrolytic oils from municipal plastic waste with its applications' studies have been limited[25–28].

Non-thermal plasma technology for conversion of waste plastic into fuel is one of the best and effective methods available till date, as compared to other methods due to low energy consumption. Microwave-induced plasma has advantages like simplicity, compactness, lightweight reactor, uniform heating and the ability to operate under atmospheric pressure. Ethaib *et al.*[29] have studied microwave-assisted pyrolysis of biomass waste. Ho *et al.*[30] have reviewed dominating factors on the performance of microwave induced plasma for processing of solid waste into fuels. Liu *et al.*[31] have reviewed the affecting parameters and the end product formation in pyrolysis of biomass along with microwave assisted pyrolysis. Khongkrapan *et al.*[32] have studied the conversion of solid wastes (paper, biomass, and plastic) through plasma-based pyrolysis. Catalytic microwave-assisted pyrolysis could also be a feasible approach for chemical recycling of waste plastics and producing fuel and petrochemical feedstocks such as naphtha[33].

To the best of our knowledge, no specific study on photocatalytic assisted microwave-based plasma pyrolyser is available till date for the conversion of COVID-19 related wastes thereby, contributing in carbon sequestration. The aim of this review is to analyse the synergistic effects of catalyst and microwave based plasma pyrolyser along with the affecting parameters for decomposition of COVID-19 related wastes i.e. PPEs, because catalyst resolves the problem of hydrogen deficiency in the pyrolytic process.

2. Current status of the photocatalytic assisted or microwave-based plasma pyrolyser for conversion of COVID-19 related wastes (PPEs)

To meet the energy requirements as per the consumer demands due to the uncertainty in the availability of fossil resources and environmental concern, it would be a judicious step to explore the waste to energy routes in the long run. The accumulation of COVID-19 wastes if not dealt as per the standard protocols, could lead to a havoc situation worldwide disrupting biological ecosystems, as the man-made virus is novel and actively mutates under normal environmental conditions, therefore it can affect different organisms differently. In the absence of extensive research studies in biological model systems other than humans, and till the advent of effective vaccines, the best practice would be to ensure safe disposal of COVID-19 wastes. The plastic footprint in the environment has considerably increased due to decline in the profit margins of recycling and hence, supply of recycled plastic material has dropped. Circular economical strategies including recycling practices and firm policies to mitigate plastic pollution for attaining the United Nations' Sustainable Development Goals is the need of the hour[34].

In 1940s, Percy Spencer discovered microwave heating for cooking food and numerous other applications which improved quality production resulting in development of new end products/processes. Since then, microwave heating has become more acceptable for food preparation and a microwave oven is one of the common appliances in most modern house kitchen and restaurants. Fig. 1 shows the schematic of microwave setup for pyrolysis. Microwaves (wavelengths between 0.001–1 m corresponding to 300 and 0.3 GHz frequency range) lie between infrared radiation and radio waves in the electromagnetic spectrum region. Federal Communications Commission (FCC) holds two different microwave heating frequencies (0.915 and 2.45 GHz) used for industrial, scientific, and medical purposes mainly cellular phones, radar, and television satellite communications.

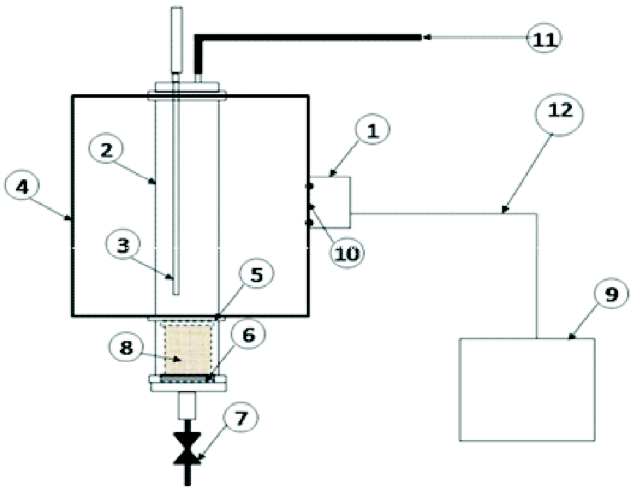

Fig. 1. Schematic drawing of the microwave reactor: (1) magnetron; (2) quartz reactor; (3) thermocouple; (4) oven casing; (5) quartz distributor plate; (6) spring for quartz holder fitting; (7) N_2 gas inlet; (8) quartz holder fitting; (9) cooler system for spring for quartz holder fitting; (10) hole for wave transmission into cavity; (11) gas outlet; (12) electrical connection (Source: Rosi *et al.*[35]).

The yield characteristics of the final products are determined by the material properties and operating conditions in a pyrolysis reactor. However, additional complications in the chemical reactions may arise affecting the mass and energy balance in the microwave assisted pyrolytic reactor system. Heterogeneous reactions or de-volatilization is usually favoured by microwave heating of the solid material[36], whereas, the gas-phase homogeneous reactions are favoured by conventional heating process. Moreover, the occurrence of the undesirable reactions can be prevented by the reduced temperatures in the microwave cavity that also helps in the condensation of the final pyrolysis vapours in the target area.

In UK, Tech-En Ltd. initially developed microwave heating techniques for plastic waste conversion[37]. Plastic wastes are highly transparent to microwaves with high carbon content as the microwave-absorbent material. In a microwave field, conversion of plastic waste into carbon at 1000°C temperature is more energy efficient process, with uniform heat distribution than conventional one and also prevents formation of undesirable oxygenated hydrocarbon products. Hussain *et al.*[38] studied the microwave heating process of PS into hydrocarbons using iron metal as antenna (microwave receptor or microwave absorbing dopant). Ludlow-Palafox and Chase[39] studied degradation of high-density PE (polyethylene) and aluminium/polymer laminates (toothpaste tube) in a novel microwave assisted pyrolytic reactor and concluded that problematic wastes such as laminates can be dealt with such process effectively[40]. High quality aluminium was recovered along with liquid and gaseous hydrocarbon production during this process with feed mixture of aluminium and toothpaste tube. This study showed that the process has good potential for the plastic wastes treatment on a commercial scale.

Similarly, sterilization of hospital wastes can also be done using microwave technology[41]. Large volumes of hospital waste (HW) generally includes pathological, microbiological, sharps, etc. The advantage of *in situ* treatment with microwave source is that it requires less time to convert such hazardous waste into inert ash. Although, few reports state that microwave heating produces more gas and less oil than conventional pyrolysis[42]. The major constituents of the gaseous products formed during pyrolysis are H_2, CO, CH_4, and other lighter hydrocarbons. Compared to the conventional heating, microwave heating produce more H_2, and CO content along with other hydrocarbon fractions[42].

Nema and Ganeshprasad[43] stated that plasma pyrolysis is a state-of-the-art technology for safe medical waste disposal. Department of Atomic Energy, GoI has endorsed the use of a pyrolysis system developed by the Institute of Plasma Research, Gandhinagar, Gujarat which is successfully demonstrated at the Gujarat Cancer Research Institute for medical waste treatment and now this technology is commercialized by M/s Bhagwati Pyrotech Pvt. Ltd., Ahmedabad, Gujarat, India. Plasma pyrolytic technology is an effective solution for complete pyrolysis of typical hospital waste not limited to PPE wastes. The pathogens are completely destroyed upon the exposure to high temperatures integrated with high UV radiation flux in the thermal plasma pyrolysis system. Although, concerted efforts are required to explore the potential of fluidized bed microwave assisted plasma based pyrolyser in this regard.

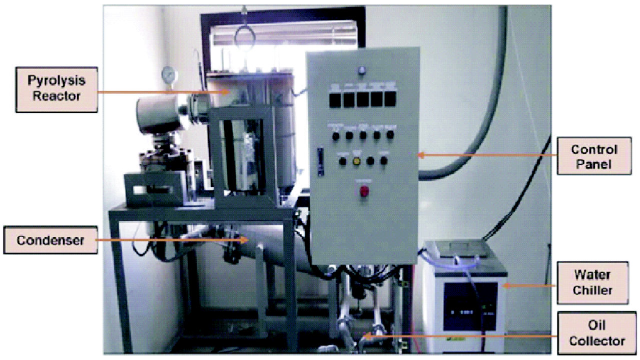

Fig. 2. Batch type pyro-reactor (Source: Miandad *et al.*[44]).

Miandad *et al.*[44] studied the effect of various plastic waste types (polystyrene (PS), polypropylene (PP), polyethylene terephthalate (PET), and poly-ethylene (PE)) on quality of production of pyrolysis oil. Fig. 2 shows the pictorial representation of batch pyro-reactor experimental setup used for pyrolysis process operated at room temperature to 450°C at constant retention time of 75 min.

Fig. 3 shows the produced pyrolytic oil from the pyrolyzer with different types of plastics (PS, PP, and PE) and their combination (PS/PP, PS/PE, PS/PP/PE and PS/PP/PE/PET) used as feed at constant 450°C temperature with 75 min retention time except for PE (having longer carbon chain structure) that gets converted into wax instead of oil[21,45]. From the figure, it is evident that maximum conversion of 80.8% was achieved when PS was used as the feedstock. This is because of the simple chemical structure of PS in contrast to other plastic types that follow four basic degradation steps i.e. initiation, transfer, decomposition, and termination[28]. Similar results were reported by Ciliz *et al.*[22]. However, the gas product and char yield were significantly low as compared to all other feedstock. Lower amount of pyrolytic oil and char were produced from PP feedstock due to fast pyrolysis that operated at higher heating rate of 10°C/min. However, maximum gas yield of 54.6% was obtained from PP

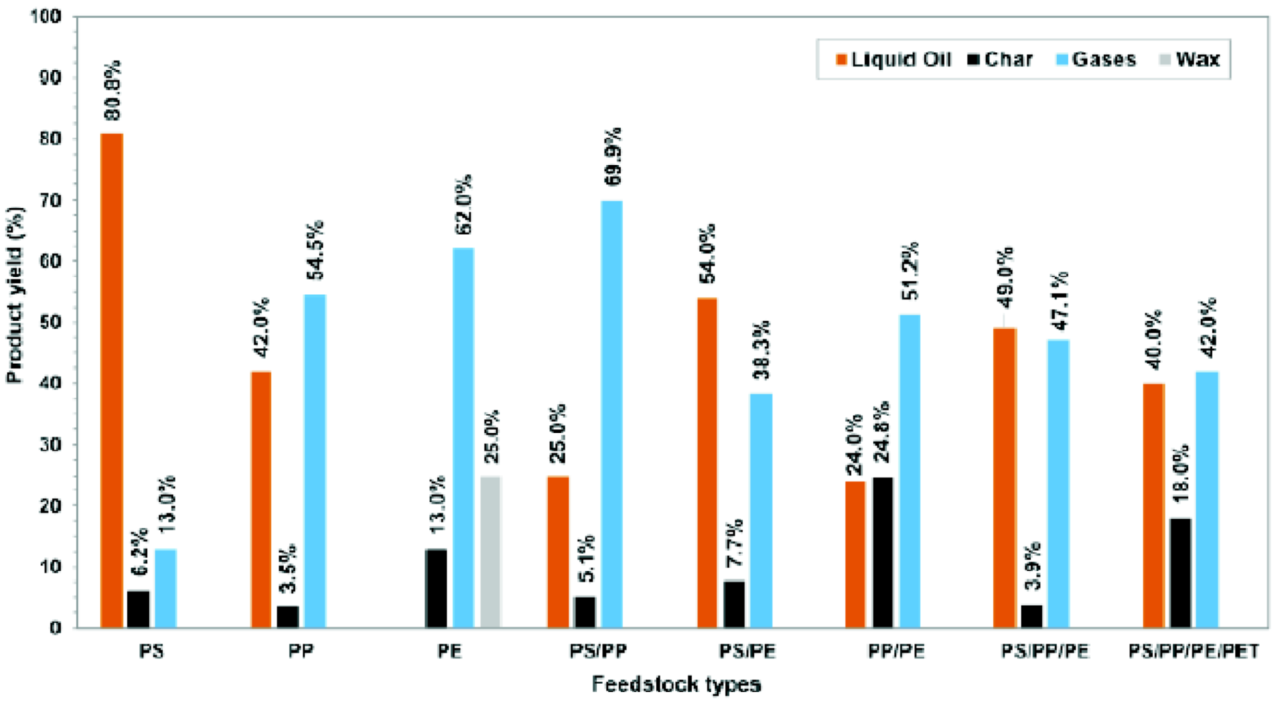

Fig. 3. Type of plastic waste with pyrolysis yield (Source: Miandad *et al.*[44]).

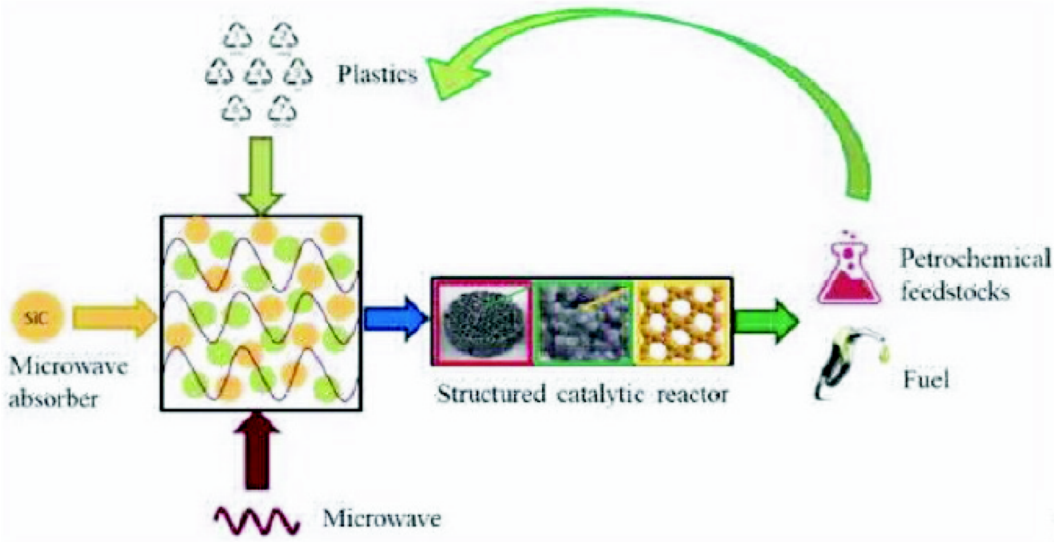

Fig. 4. Schematic view of conversion of plastics into fuels from waste plastic through microwave assisted process (Source: Ruan *et al.*[33]).

feedstock. Koo and Kim[46] have reported higher pyrolytic oil yields in slow pyrolysis process. Moreover, mixed feedstock (PS/PE 50%:50%) has been shown to produce 54% pyrolytic oil. Miandad *et al.*[21] have performed gas chromatography analysis of the pyrolytic oil fraction and found that ethylbenzene (21.2%), toluene (25.6%), and styrene (48.3%) are the main components in case of PS pyrolysis.

Fig. 4 shows the abstract overview of the catalytic microwave-assisted pyrolysis for plastic waste[33]. The developed continuous lab-scale pyrolytic chemical reactor consisted of downdraft operation and a mixing ball bed reactor with operating capacity of 200 kg plastics per day. The ZSM-5 catalyst system was used by Ruan *et al.*[33] in combination with Al_2O_3 and obtained 57 wt% yield of C_5-C_{22} liquid hydrocarbons through microwave plasma pyrolytic reactor with 32% energy saving during the process.

In case of sequential catalytic treatment by Al_2O_3 followed by ZSM-5, conversion into monoaromatics and C_5-C_{12} alkanes/olefins up to 100% can be achieved at a catalyst to plastic ratio of 4:1. This higher conversion was achieved due to promotion of alkanes/alkenes production in the C_5-C_{23} range by Al_2O_3. The group also reported formation of C_{13}-C_{23} alkanes and alkenes with a selectivity of 86.6% in the case of MCM-41, whereas, ZSM-5 facilitated the production of aromatics with 70% selectivity. Moreover, the reactor design plays a crucial role in providing sufficient heat and mass transfer within the catalyst bed during reactor operation and catalyst regeneration. Ruan *et al.*[33] also synthesized ZSM-5 coatings on a SiC foam support for upgrading pyrolytic vapours with higher catalytic activity, stability and regenerability.

Biomedical waste is categorized as highly toxic/hazardous pollutants due to rapid mobility/transmission of biological organisms from micro to macro level. Fig. 5 shows biomedical waste categorization as hazardous (risk waste) comprising of sharps, pathological, pharmaceutical, chemical, and radioactive waste and non-hazardous (non-risk) waste include paper, packaging, and food waste[46].

Although different methods (adsorption method, microbial degradation, ion exchange method) have been reported for waste treatment, their application is limited by poor efficiency, low affordability, secondary pollution and more complicated mechanism. Photocatalysis chemical method is quite promising due to its simplicity, affordability, nontoxic, high degradation efficiency and higher stability[47]. Photocatalysts/nano-photocatalysts are widely used in the field of environmental and ecological safety for their advantages including efficient degradation of biological contents, destruction of specific pathogens, low energy consumption, capability to treat lower concentration of pollutants using sustainable approach[48]. Nano-photocatalyst possess significantly high photoactivity due to larger surface area which makes them unique for being able to absorb solar energy for degrading higher levels of organic compounds and biological micro-organisms under controlled conditions[49,50]. The photocatalytic performance for specific applications could be enhanced by range of intensity of light via catalysts by exploring methods such as coupled/metal-doped semi-conductor, noble metal deposition to maximize their ionized bandgap. For example, graphite carbon nitride nanocomposites, CuO-doped titania, nitrogen-doped TiO_2 photocatalysts used for degradation of oxytetracycline antibiotic, inactivation of cancer cells, etc. Hooshmand *et al.*[51] have reviewed the activity of nano-photocatalysts for biomedical waste management, citing their importance in health care sector. Now-a-days, nano-photocatalysts combined with thermal degradation methods is a new explorable route for decomposing biomedical waste.

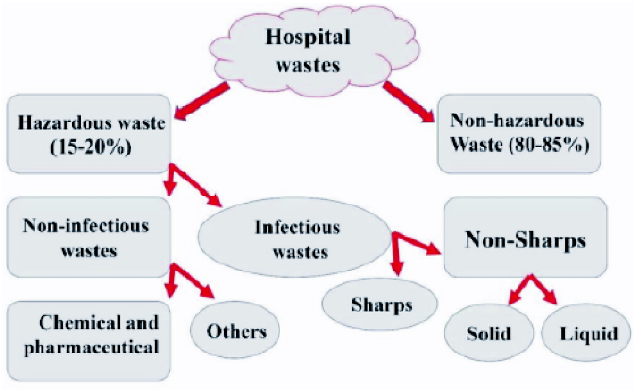

Fig. 5. Classification of biomedical/hospital wastes (Source: Hooshmand *et al.*[51]).

Khongkrapan *et al.*[32] investigated pyrolysis process using constant 800 W microwave plasma with varying argon gas flow rate from 0.50 to 1.25 lpm of refuse derived fuel (RDF) conversion along with plastic, paper, and biomass

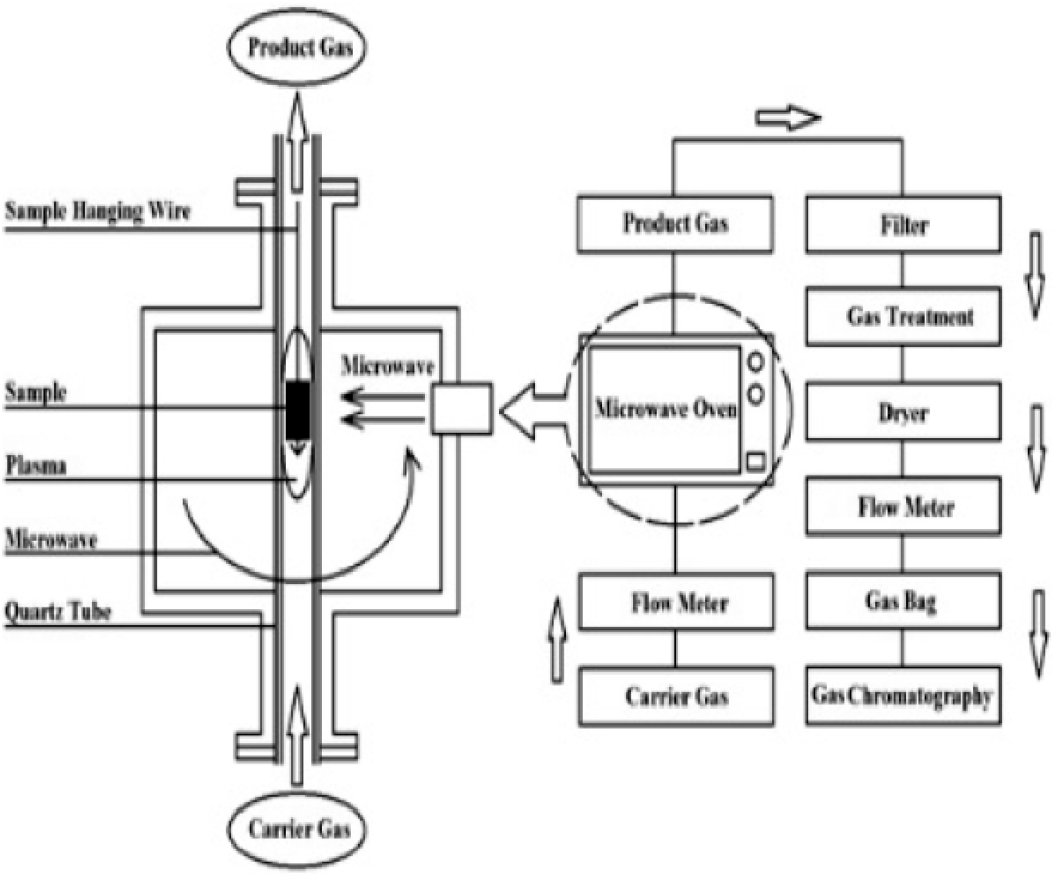

Fig. 6. Schematic view of microwave plasma reactor (Source: Khongkrapan *et al.*[32]).

combustible components. Fig. 6 shows their schematic experiment setup which consists of a plasma reactor assisted with plasma microwave generator (800 W) whose inner diameter, outer diameter, and length were 27 mm, 30 mm, and 250 mm, respectively.

Converted char has 39 MJ/kg GCV, and yield about 12–21% of the original mass, and combustible gas (1.0–1.7 m^3/kg containing approximately 14% H_2, 66% CO and 4% CH_4 and heating value of 11 MJ/m^3), along with plasma light emission and maximum power density generation of 35 W/cm^3. The products formed were potentially marketable forms of clean energy[32].

Fig. 7a-d shows variation in the formation of different products H_2, CO, CH_4, and CO_2 based on argon flow rate and types of wastes. From the Fig. 7a-c, an increasing trend upto 0.75 lpm argon gas flow rate is observed for H_2, CO, CH_4, followed by a decreasing trend in all the products, except CO_2. However, in case of CO_2 (Fig. 7d), with increase of argon gas flow rate, the trend decreased with flow rate upto 0.75 lpm and followed by a steep increase. The trends were similar to those observed for the post plasma temperature. At 0.75 lpm gas flow rate, the calorific value and carbon conversion efficiency were reported to be highest. The reported volume percentage of produced H_2 were 24, 22, 10 and14%,

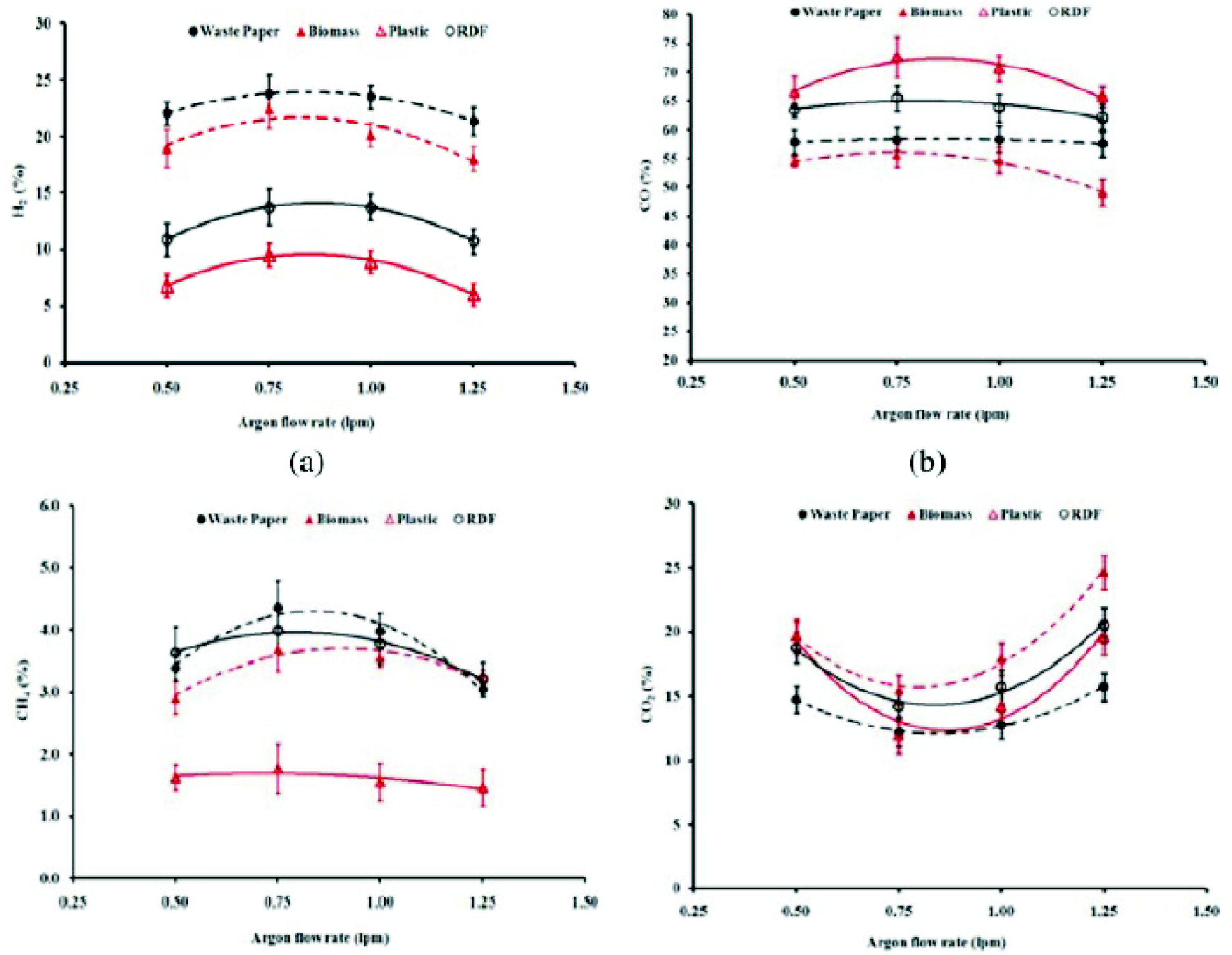

Fig. 7. Effect of different wastes on argon flow rate (lpm): (a) percent of H_2, (b) percent of CO, (c) percent of CH_4, and (d) percent of CO_2 (Source: Khongkrapan *et al.*[32]).

respectively for paper, biomass, plastic and RDF. Moreover, high CO content (56–73%) was reported in the product gas that used mainly plastic as the feedstock.

Khongkrapan *et al.*[32] showed significant increase in the carbon and energy content of the carbonized feedstock in their proximate and ultimate compositional analysis. However, volatile content was decreased due to the conversion of organic constituents into gaseous phase. Plastic showed higher solid residue formation as compared to all other feedstock duringa 3 min pyrolysis process. This also implies that longer residence time is required for carbonization of plastic waste with generation of higher calorific value char yields[52,53].

Suriapparao *et al.*[54] developed mechanism for co-pyrolysis of synthetic plastics, polystyrene (PS) and polypropylene (PP) mixed with different co-substrate such as *Prosopis juliflora* (PJF), bagasse (B), groundnut shell (G), mixed wood sawdust (MWSD) and rice husk (RH) operated at 450 W microwave source. Figs. 8–9 shows mass and energy yield of products (%) on different co-pyrolysis feed.

They found lower viscosity value in the PS-biomass mixture than biomass based oil. However, in case of pyrolytic oil from PS-biomass, high monoaromatic and polyaromatic hydrocarbon content is present. In case of PP-biomass mixture, high viscosity (640–702 cP) was found which contains mainly long chain aliphatic hydrocarbons and non-additive nature. The density of produced pyrolytic oil from PS-mixtures was 2.0–2.75 cSt which was similar to the reported value[55,56] and this value was higher than conventional fuels like gasoline.

Fig. 8 shows yields of products (%) obtained by PS co-pyrolysis with different biomass as studied by Suriapparao *et al.*[54]. When polymers and biomass are mixed, higher HHV were found as compared to that of the biomass alone. Within 10–11.5 min, the pyrolysis temperature went up to 600°C for all the feed materials, corresponding to 51–59°C/min heating rate. The evolution of oil has been reported when the temperature varies (250–450°C) during the pyrolysis reaction. Beyond 450°C, only gas evolution was observed. This reveals that localized micro-plasma generation causes the complete conversion in a pyroreactor with moderate temperatures. At higher temperatures, char decomposes post devolatilization, resulting in the gas evolution. Therefore, no significant change in the oil yield or its composition has been reported for consequent change in the pyrolysis duration (9–12 min).

From Fig. 8, it is imperative that the overall bio-oil yields of feed mixtures are high in the range of 74–81%, and trends are (PS:RH) > (PS:MWSD) > (PS:B) > 77.6% (PS:PJF) > 73.8% (PS:G)[54]. It is noteworthy to mention that these values are higher than in case of individual material feed. Therefore, it is concluded that RH is a desirable biomass for mixing with PS for production of high-quality bio-oil. For a majority of the feedstock mixtures, char's energy densification ra-

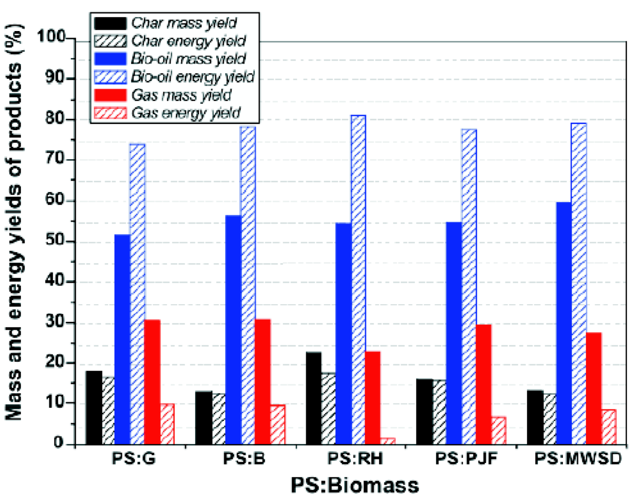

Fig. 8. Mass and energy yields of products (%) obtained by PS co-pyrolysis with different biomass (Source: Suriapparao *et al.*[54]).

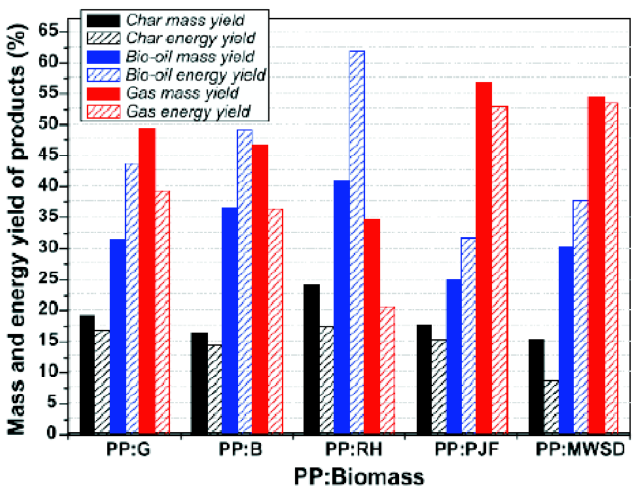

Fig. 9. Mass and energy yields of products (%) obtained by co-pyrolysis of PP with different biomass (Source: Suriapparao *et al.*[54]).

tio is close to unity, implying that co-pyrolysis chars have the HHVs (high heating values) very similar to that of the feedstock.

Fig. 9 shows that bio-oil yields in the PP-biomass mixture through microwave assisted pyro-reactor was in the range of 31.7% to 62% and trend are PP:RH > PP:B > PP:G > PP:MWSD > PP:PJF. However, PP:RH mixture showed maximum product heating value with 1.24–1.5 energy densification ratio as compared to other feed mixtures. This is due to active inter-bonding of oxygen and hydrogen during this co-pyrolysis process. It is also evident to note that although the mass and energy yield patterns for all the feedstock PS-biomass mixtures in co-pyrolysis are alike as shown in Fig. 8, patterns are significantly different for PP-biomass mixtures (Fig. 9). Studies revealed that mixture of feed materials such as PP:PJP and PP:MWSD produced higher energy content hydrocarbon gases, as compared to individual waste alone. Finally, Suriapparao *et al*.[54] concluded that energy efficiency was lower in case of biomass alone than polymer materials which contains high level of hydrocarbons. It is suggested that improving the microwave reactor design for processing large feedstock volumes would enable lower energy losses.

Conclusion

This review provides scientific perspective on photocatalytic assisted microwave-based plasma pyrolyser for conversion of COVID-19 related wastes with demonstrated high carbon conversion efficiency. High efficiency, lower operating costs with minimum energy consumption, more energy dense products, and also curtailing pollution are the main advantages of pyrolysis which as seen in the literature discussion. Although, pyrolysis being a self-sufficient treatment process, still has a long way to go in waste-to-energy conversion industrial process. The processing cost will go down significantly if, the batch processes can be converted into the continuous processes. Often, the pyrolysis char is used as solid fuel/additives for boilers, activated carbon, production of carbon nanofilaments, generating high surface area catalysts for use in electrochemical capacitors and production of carbon nanoparticles in gas phase secondary reactions.

Pyrolytic oil contains organic and inorganic compounds, having multiple industrial uses such as combustion fuel, blended transportation fuel, power generation, chemicals and resins, wood preservatives, making adhesives, production of anhydro-sugars like levoglucosan, pelletizing binder and briquetting of combustible organic waste substance, etc.

Due to the presence of high aromatic compounds in pyrolytic liquid oil, its blending with conventional diesel, post distillation and refining treatment is required to use it as a suitable transportation fuel. If the aromatic compounds especially styrene could be recovered from pyrolysis oil, it can be a potential precursor chemical in polymer industries for manufacture of styrene products. All the studies discussed in the paper imply that pyrolytic oil product generated from microwave plasma reactor are similar in characteristics to the conventional diesel.

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