CONTEMPORARY ADVANCES ON CONVERTING MARINE PLASTIC WASTE INTO READY TO USE FUELS

Introduction

Since 1990, the global demand for plastic has increased by 5% every year. In 2019, the global plastic market size was valued at 568.9 billion USD, with an expected compounded annual growth rate of 3.2% over the next decade [1]. As the use of plastic in both industrial and municipal settings increases, plastic waste pollution becomes an increasingly major threat to both ecological and human health.

Marine ecosystems in particular have been negatively impacted by global reliance on plastics. The ecological consequences of marine plastic waste cannot be understated. According to Barnes et al., up to 80 percent of waste that accumulates on shorelines, the seabed, and the ocean surface is plastic. As plastics do not biodegrade, the waste eventually deteriorates into microplastics, which are then consumed by marine life [3]. According to Wabnitz et al., the bodies of most marine species, ranging from plankton to whales, now contain plastic. Plastic waste has become dangerously intertwined with the ecology of marine life, with plastic being seen in bird nests, worn by hermit crabs, and appearing in the intestines of turtles and albatross [5]. Nearly 300 marine species have been shown to ingest or become entangled in plastic waste, resulting in biological harm and death [6]. Because of its light weight, durability, and buoyancy, plastic waste is a serious biological hazard once seaborne. The inherent buoyancy of most plastic waste means the waste is easily swept by ocean currents, where it contaminates even remote regions of the oceans.

Roughly 50% of waste plastic is deemed "unrecoverable" and subsequently disposed of [7]. Among conventional techniques, landfills and incineration are the most common methods of disposal [8]. Incineration as a means of energy recovery is directly linked to the release of harmful compounds, including dioxins and furans, into the environment, which negatively impacts human and ecological health [9]. Recycling is regarded as the best possible solution for the plastic waste crisis. While there is unmistakable value in primary and secondary recycling of plastic solid waste (PSW), or the repurposing of plastics into new plastic products, through some blending of virgin materials, tertiary recycling is the ideal approach for converting PSW into high-value chemicals: namely, hydrocarbon fuels. Tertiary recycling, or cracking, is a process of degrading plastics, typically through pyrolysis or catalytic cracking. Pyrolysis and catalytic thermal cracking are the most promising and effective methods of depolymerizing post-consumer plastics.

Methods of Plastic to Fuel Conversion

Pyrolysis is a method of thermal depolymerization of plastics in the absence of oxygen. Typically, plastic is fed into a chamber, where it is heated to a narrow temperature range, without excessive variations. Oxygen is then purged from the pyrolysis chamber. The formation of carbonaceous char by-products is managed before it lowers the heat transfer to the plastic. Finally, the pyrolysis vapors are condensed to produce consistent, high-quality distillate [10].

The pyrolysis process can be further broken down into process subsets. Slow pyrolysis involves the use of a slow heating rate, while fast and flash pyrolysis requires rapidly heating the plastic to temperatures as high as 1300K [11]. Catalytic thermal degradation is a subset of traditional pyrolysis, in which catalysts are added to pyrolysis reactions. The addition of catalysts has been shown to offer numerous advantages over purely thermal pyrolysis. In polyolefin pyrolysis, the addition of a catalyst lowered pyrolysis temperatures and residence time. The reduction of the degradation temperature and reaction time results in an increase of conversion rates from many polymers compared to thermal pyrolysis alone [12-15]. Specifically, catalytic pyrolysis has been shown to provide better control over the hydrocarbon product distribution in the pyrolysis of low and high-density polyethylene, as well as with polypropylene and polystyrene [16-20]. Several catalysts, such as zeolite [21], silica-alumina, and clay [22-23], have been shown to improve fuel quality, increase selectivity, and lower both the pyrolysis temperature and residence time [22].

Also promising is the method of hydrocracking. Hydrocracking, the cracking of larger hydrocarbons into fuel-range hydrocarbons. Hydrocracking is simply pyrolysis performed in the presence of hydrogen at elevated temperatures [24].

By far, the most discarded plastic types are polyolefins, such as polyethylene, polypropylene, and polystyrene. Overall, polyolefins account for more than half of all plastic waste [25-26]. As a result, a notable amount of recent research has been focused on methods of converting this waste into useful materials. Polyethylene (PE) and polypropylene (PP) are polymers containing only carbon and hydrogen [27]. As a result, fuels derived from this plastic waste are comparable to fossil fuels, and typically don't require further upgrading.

Process Optimization

While effective, thermal pyrolysis is often economically infeasible, as the reaction requires significant energy. As catalysts lower the activation energy required for pyrolysis, their use is essential for widespread adoption. Reducing energy input through catalysts reduces the reaction temperature and time, narrowing the product distribution.

Optimization of the plastic to fuel conversion process has been the subject of several studies. Several factors, including catalyst type, feed composition, particle size, and catalyst-to-polymer ratio have been shown to impact both fuel and conversion quality. Notably, zeolite (aluminosilicate mineral) based catalysts are also more effective, and substantially reduce the temperature of cracking. Reforming catalysts, including Pt/SiO2Al2O3 are shown to increase the octane number (i.e., the measure of a fuel's ability to withstand compression in an internal combustion engine without detonating) while maintaining the carbon number. Feed composition can affect the speed of plastic decomposition. The decomposition of polyethylene and polypropylene is faster when mixed with polystyrene because it catalyzes the radical formation reaction. Acidic sites also play a role in cracking. With an increase in pore size, hydrocarbons degrade into smaller hydrocarbons, such as gas. Particle size should be considered. Catalysts with smaller particle sizes have larger surface areas for catalytic activities. However, this can translate to a smaller pore size for cracking. Catalyst loading methods are also of interest. Two methods of catalyst loading can be employed. Liquid phase contact is an effective method in which the catalyst is mixed with plastic. While conversions using this method are effective, catalyst recovery is extremely poor [22]. The alternative method utilizes vapor phase contact and requires the loading of the catalyst into a basket [23, 28].

In plastic pyrolysis, mass and energy transfer, due to the high viscosity and low thermal conductivity of plastics, are notable constraints. The reactor type, operating conditions, and product distributions all impact mass and energy transfer [29]. Uniform heat distribution is needed for effective mass and energy transfer. The use of fluidized bed reactors (Figure 1) can facilitate a uniform distribution of energy, which is a crucial component in obtaining desirable hydrocarbons. Because of the rapid circulation and the turbulent gas flow in fluidized bed reactors, a much higher efficiency in heat exchange is attained, as well as better temperature control [30]. Sharratt et al. tested the catalytic cracking of HDPE (high-density polyethylene) with ZSM-5 (Zeolite Socony Mobil−5) in a fluidized bed reactor, obtaining more than 65% of hydrocarbons, with a range of C3-C5. Screw kiln reactors have been shown to be more effective than batch reactors, producing lower percentages of heavy products. Another area of interest in optimizing heat transfer is the viscosity reduction of molten plastic. This can be achieved through the incorporation of solvents or oils, such as lubricating oil or vacuum gas oil. The

use of these oils lowers the pyrolysis temperature, resulting in an increased yield and a decreased energy cost [32]. Serrano et al. tested lubricating oil with LDPE (low-density polyethylene) in a range of concentrations in a screw kiln reactor (Figure 2) and was able to achieve complete conversion at acceptable temperatures.
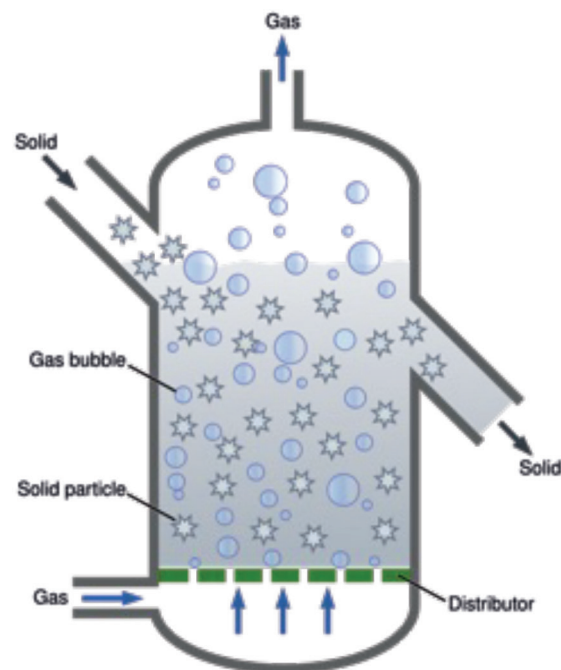

Figure 1: Schematic of Fluidized Bed Reactor [33]

Also of interest is the kinetics of pyrolysis. Kinetic evaluation involves performing plastic cracking via thermogravimetric analysis techniques. The amount of plastic mass degraded and the reaction temperature are measured at regular time intervals [35]. The results of kinetic studies show that cracking is a first-order reaction, consisting of a single-step degradation process. This generally holds true for polyamide (PA), polycarbonate (PC), and both LDPE and HDPE. Polyvinyl chloride (PVC) is reported to decompose over multi-reaction mechanisms, likely due to the release of HCl during degradation [36]. Activation energies can also be determined via kinetic evaluation. Sorun et al. examined PS, LDPE, HDPE, and PP, using a single reaction model. It was found that the lower activation energy bound for PS degradation is 311.5 kJ/mole, and the upper bound for HDPE is 445.1 kJ/mole. The activation energy is determined by the dissociation energy for the different bonds, such as C=C, C-H, and C-C.

With increasing pyrolytic temperature, the yield of gasoline-range hydrocarbons increases. The yield of gaseous products also increases. In a pyrolysis experiment on mixed plastics, the highest yield of liquid fuel (76%) and gaseous products (28.5%) was obtained at 370 degrees celsius [38-39]. Also of note are comparative studies of yields from varying pyrolysis temperatures. Kamal et al. tested the pyrolysis of mixed plastics (2:1 ratio of PE to PP) at both 900 and 425 degrees celsius. It was determined that there were significant differences in the calorific values of the two products, with the 900°C pyrolysis product showing 46.8 MJ/kg, and the 425°C product showing 41.8 MJ/kg. Interestingly, the higher temperature reduced the levels of carcinogenic compounds, including boric acid and cyclopentanone. Olefin content is similarly influenced by temperature, showing a decrease with increasing pyrolysis temperature [40] This is likely a result of longer reaction times with lower temperatures, making flash pyrolysis a potentially more attractive method.

Successful Plastic to Fuel Processes

In real-world cases, all of these methods show promise. For non-catalytic pyrolysis, a temperature range as broad as 350-900°C can be used, with varying results. In the 650-700°C range, the weight percent of gases produced increases. When temperatures are near 500°C, oil containing aromatic hydrocarbons are produced [41].

Sarker et al.[19] tested pyrolysis at temperatures between 370 and 420°C waste plastic degradation. In this study, the plastic was degraded to a slurry and then condensed, resulting in a range of hydrocarbons between C4H8 and C28H58 [42]. The differences between hydrocarbon products from waste plastic mixtures and individual plastics have been studied as well. Williams et al. pyrolyzed both waste mixtures and pure plastics in the presence of nitrogen and hydrogen. It was found that while the compositions of gas products from mixtures and individual plastics were similar, the mixed plastics produced higher concentrations of alkanes and aromatics. Sharma et al. pyrolyzed high-density polyethylene in the form of grocery bags. The
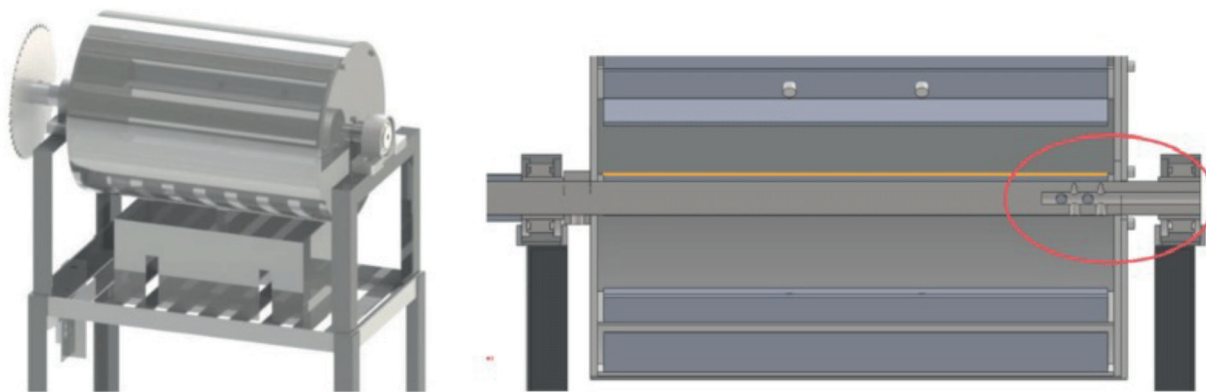

Figure 2: Schematic of rotary kiln reactor with output hole for pyrolysis gas [34]

resulting oil had paraffinic hydrocarbons as high as 96.8% and an absence of any oxygenated products. The resulting crude oil was synthesized to obtain: 20% motor oil, 41% diesel #1, 23% diesel #2, and 16% vacuum gas oil-range fractions. This conversion occurred at 440 celsius without the use of a catalyst.

Also noteworthy are the pyrolysis tests performed using catalysts. Sarker et al. pyrolyzed waste polyethylene terephthalate to produce hydrocarbons at a temperature of 405°C. Hydrocarbons ranging from C3 to C27 were obtained, with very few oxygenated compounds. The study of the conversion of contaminated plastics to fuel is particularly interesting, as virtually all ocean MSW is contaminated. Miskolczi et al. examined the catalytic conversion of contaminated HDPE and PP. While the contaminants (Ca, P, C, and N) did distort the fuel properties, when the trial was run with the addition of ZSM-5, these distortions were negated. This lower concentration of impurities can be attributed to the contaminants attaching to the catalyst, and therefore becoming separated from the hydrocarbons. The presence of the catalyst also increased the percentage of lighter hydrocarbons and gasoline products. Isobutane formation was also higher when the catalyst was used.

Also tested was the impact of catalyst pore size on pyrolysis conversions. The use of HZSM-5, HY, and ⬚ zeolite catalysts was compared. Elordi et al. determined that smaller pore-sized catalysts, such as ZSM-5, resulted in an elevated yield of lighter olefins and gases. With the use of a catalyst with larger pores, non-aromatic compounds and higher hydrocarbons are predominantly produced. Also worth considering is the use of multiple catalysts in combination. Uemechi et al. utilized both HZSM-5 and SiO2-Al2O3, producing a high yield of gasoline with a high octane rating.

The potential of plastic pyrolysis cannot be understated. The conversion of waste plastic to fuel simultaneously addressed the issues of plastic pollution and reliance on fossil fuels, making it an attractive solution for the pollution and energy crises.

Conclusion

In the last 20 years, the use of plastics has skyrocketed. Yet, the same properties that make plastic so desirable, including its durability, light weight, and low cost, also make plastic a unique environmental danger. As plastics continue to accumulate in the ocean, the damage to marine ecosystems and wildlife becomes an increasingly urgent concern. Simultaneously, there is a continuous rise in the consumption and cost of fossil fuels. As the world continues to further its reliance on both plastic and fossil fuels, the conversion of waste plastics to fuel presents an attractive opportunity. As the majority of plastic waste is either polyethylene or polypropylene, most of the existing literature on plastic pyrolysis concerns PE and PP.

Municipal plastic wastes are easily converted into fuel-range hydrocarbons via pyrolysis and catalytic cracking processes. The thermal degradation of plastics has been proven an effective alternative fuel source and has potential for widespread adoption. For pyrolysis processes to become commonplace in both the plastics and energy industries, further study is recommended in several subsets. The use of catalysts is an opportunity for both enhanced efficiency and greater selectivity in the fuel conversion process. The choice of catalyst is also impactful, with the pore size impacting the yield. Optimization is also possible through procedural changes, including catalyst loading, temperature, and reactor type. As sufficient literature is published on the optimization of the degradation process and the fuel yields become uniform, the use of cracking techniques to repurpose plastic waste will become sustainable and widely adopted.

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Authors

Dr. Raj Shah is a Director at Koehler Instrument Company in New York, where he has worked for the last 27 years. He is an elected Fellow by his peers at IChemE, CMI, STLE, AIC, NLGI, INSTMC, Institute of Physics, The Energy Institute and The Royal Society of Chemistry. An ASTM Eagle award recipient, Dr. Shah recently coedited the bestseller, "Fuels and Lubricants handbook", details of which are available at ASTM's Long-Awaited Fuels and Lubricants Handbook 2nd Edition Now Available - Jul 15 2020 - David Phillips - Petro Industry News Articles - Petro Online (petro-online.com)

A Ph.D in Chemical Engineering from The Penn State University and a Fellow from The Chartered Management Institute, London, Dr. Shah is also a Chartered Scientist with the Science Council, a Chartered Petroleum Engineer with the Energy Institute and a Chartered Engineer with the Engineering council, UK. Dr. Shah was recently granted the honourific of "Eminent engineer" with Tau beta Pi, the largest engineering society in the USA. He is on the Advisory board of directors at Farmingdale university (Mechanical Technology) , Auburn Univ (Tribology) and Stony Brook University (Chemical engineering/ Material Science and engineering).

An adjunct professor at the Dept. of Material Science and Chemical Engineering at State University of New York, Stony Brook, Raj also has over 470 publications and has been active in the alternative energy arena for over 3 decades. More information on Raj can be found at Koehler Instrument Company's Director elected as a Fellow at the International Institute of Physics Petro Online (petro-online.com)

Nikhil Pai is an engineering student whose interests and research are focused on additive manufacturing and material science. He is currently also part of a thriving internship program at Koehler instrument Company, which encourages engineering students to explore the field of alternative energy technologies

Author Contact Details

Dr. Raj Shah, Koehler Instrument Company • Holtsville, NY11742 USA • Email: rshah@koehlerinstrument.com
• Web: www.koehlerinstrument.com

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