

A Thermo-Catalytic Pyrolysis of Polystyrene Waste Review

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Global polystyrene (PS) production has been influenced by the lightness and heat resistance this material offers in different applications, such as construction and packaging. However, population growth and the lack of PS recycling lead to a large waste generation, affecting the environment. Pyrolysis has been recognized as an effective recycling method, converting PS waste into valuable products in the chemical industry. The conversion of PS into a liquid with high aromatic content (84.75% of styrene) can be achieved by pyrolysis. In addition, PS favors the production of liquid fuel when subjected to co-pyrolysis with biomass, improving its properties such as viscosity and energy content.

Keywords: polystyrene ; pyrolysis ; waste ; aromatics

1. Introduction

Plastic is a non-biodegradable material referred to as the “material of every application” ^[1]. The demand for plastics continues to increase owing to their inherent properties such as light weight, low cost, flexibility, durability, resistance to erosion, versatility, moldability, and ease of production and maintenance ^{[2][3][4][5]}. Currently, plastic products are an indispensable part of people’s daily lives. They are applied in various industries, such as construction, healthcare, electronic and electrical components, agriculture, automotive, aviation, textiles, household, and packaging ^{[5][6][7][8][9]}.

Since 1950, global plastic production has grown at a compound annual growth rate of 8.40%; the annual plastic production was 390 million tons in 2021, and plastic production is estimated to reach 500 million tons in 2025 ^{[10][11]}. Worldwide, the plastic demand is increasing annually by 4% ^[12]. Polystyrene (PS) is heat resistant and light in weight; it has good strength and durability, making this polymer suitable for various applications such as food packaging, beverages, household appliances, the automotive field, and insulating systems for the construction industry ^{[13][14][15][16]} ^[17]. PS accounts for around ten wt.% of the total plastic waste produced annually in the last ten years ^[18].

The plastic industry is ubiquitous worldwide, and the generation of “plastic waste” is a consequence of industrial development steadily increasing to the point of being considered a high-impact pollutant and causing severe environmental problems ^{[19][20][21]}.

Recycling and reusing plastic waste is vital for various reasons, and the most important of all is the conservation of natural resources and the reduction in environmental pollution ^[19]. Plastic recycling provides a “waste-to-value” strategy to utilize used plastics as a resource to produce valuable commodities, which simultaneously contributes to plastic waste management and sustainable development ^[22]. In recent years, increased attention has been paid to recycling synthetic polymer waste, solving pollution problems, and reusing cheap and abundant waste products ^[13].

Plastic recycling technologies began to develop in the 1970s, and there have been many advances since then. Primary and secondary recycling involves the mechanical reprocessing of used materials into products with little or no effects on their physical properties. It can also be considered physical recycling; sorting, extrusion, segregation, grinding, and melt-processing are the most commonly used methods in the industry ^{[23][24]}. Mechanical recycling is often referred to as the most promising recycling technology in energy consumption; nevertheless, it prefers pure single-polymer streams, which are difficult to obtain due to the mixture of several plastics ^[25]. Recycling plastics has been proven to be a problematic, costly technique due to various constraints such as water contamination, and good sorting is needed before recycling ^[26].

On the other hand, tertiary recycling involves valuable chemical intermediate recovery (depolymerization, solvolysis, pyrolysis, gasification, hydrocracking, and others) ^{[27][28][29][30]}. Nowadays, chemical or feedstock recycling is attracting much attention, as it is environmentally friendly ^[31]. Chemical recycling overcomes the quality issue of thermoplastic recycling by enabling the production of virgin materials such as monomers, oligomers, and higher hydrocarbons from

chemically-recycled feedstock [27][32]. Finally, quaternary recycling involves energy recovery (incineration) [20]. Solutions such as landfilling and incineration have many drawbacks (high energy cost, loss of carbon, and others) and thus, low economic efficiency [33]. In addition, these strategies are unaffordable for developing countries because of their global warming mandate [13][34]. Incineration for energy recovery can cause environmental issues by emitting dioxins, NO_x, SO_x, heavy metal oxides, or polycyclic aromatic hydrocarbons [35][36].

2. Pyrolysis as an Alternative Plastic Waste Recycling Route

Plastic polymers are, in principle, the derivatives of fossil fuels and petrochemicals.

The long chains of polymers have several thousand repeating units of monomers, which makes plastic durable [15]. Remarkably, the high degradation stability and low density of PS cause significant problems when disposed of in a landfill; thus, processing PS waste is an important issue. Since non-renewable hydrocarbon feeds are used for the production of polymers, the chemical processing of polymer waste to produce fuels or petrochemical feedstocks is more favorable compared to other recycling methods [33][37]. Additionally, chemical recycling of plastic waste to base chemicals represents a promising recycling route since it is expected to be more robust towards impure and contaminated plastic waste streams. In pyrolysis, plastic waste is decomposed thermal or catalytic chemically at elevated temperatures (300 to 900 °C) without oxygen to convert polymers into small molecules to obtain liquid, gas, and solid fraction [71][20][24][38].

Managing plastic wastes via pyrolysis is an up-and-coming valorization technique, as it permits the use of this waste, without previous processing, to obtain products with high added value [39][40]. The main applications of pyrolytic oil derived from plastic waste are fuels [41][42][43][44][45][46][47] or chemical feedstock [23][48][49][50][51] that can be used to manufacture virgin-quality polymers [5]. Pyrolysis is a flexible recycling method because the pyrolytic products can be modified by varying the reaction conditions [14][52]. The significant factors influencing the plastic pyrolysis product molecular distribution include plastic chemical composition, reaction temperature, residence time, heating rate, operation pressure, reactor type, and catalyst application [8][37][53][54][55][56][57].

3. Polystyrene Characterization

Plastics are synthesized from organic polymers or long chains of carbon atoms in addition to hydrogen, oxygen, nitrogen, sulfur, and chlorine [15]. Moreover, thermoplastics are a family of plastics that can be melted when heated and hardened when cooled. These characteristics, which lend the material its name, are reversible. It can be reheated, reshaped, and frozen repeatedly [58].

Polystyrene is one of the polymers found in 1930, made through a polymerization process with additives. PS is made up of a long hydrocarbon chain with phenyl groups linked to alternating carbon atoms, and it is obtained from the liquid petrochemical and can be found in solid or foam states [14][15]. PS is identified by code 6 and has a chemical formula of (C₈H₈)_n and the International Union of Pure and Applied Chemistry (IUPAC) name of poly(1-phenylethene) [59]. There are different presentations of PS; among them, the following stand out: expanded polystyrene (EPS) and high-impact polystyrene (HIPS). PS is transparent but can be colored by colorants [14].

In contrast, a typical EPS is made up of 98% air and 2% PS [60]; EPS is produced by the expansion of styrene pellets [61]; the process is known as pre-expansion [13][61]. These pre-expanded beads are kept in mesh storage silos for around 24 h to diffuse air into the beads [62]. Finally, HIPS is the co-polymer of polystyrene and butadiene rubber with improved toughness compared to PS [63].

PS represents a high carbon content plastic; virgin PS can contain at least 91.23 wt.%, and it is observed that when it is discarded, the carbon content decreases. However, HIPS waste is the type with the least amount of carbon, possibly because additives accompany this material. The additives in HIPS also increase the oxygen and ash content, HIPS being the material with up to 17.84 and 7.60 wt.%, respectively. In addition, PS material is composed of volatile matter with a maximum of 99.90 wt.% when it is virgin, decreasing to 88.90 wt.% when discarded. The high amount of volatile matter makes PS an excellent candidate for chemical recycling, achieving high conversion yields.

All PS types are difficult to be degraded by nature, producing a problem for the environment [64][65].

4. Statistical Analysis of PS Pyrolysis

4.1 Thermal Degradation through TGA Analysis

Thermogravimetric analysis (TGA) determines the quantity and the frequency of the weight variation of the samples against temperature and time in a controlled atmosphere [66]. TGA is used to study the degradation behavior of polymeric materials, including homopolymers, copolymers, and others [67]. In addition, TGA helps determine degradation trends of operation parameters of the pyrolysis process, such as temperature, heating, oxygen absorption rates, and others [68]. In the literature, different studies on thermal or catalytic pyrolysis of PS have been developed; in these publications, the authors reported as a first step a thermogravimetric analysis of their samples to determine the optimum reaction conditions in the pyrolysis process and to develop their design of experiments.

Fuentes et al. [69] reported the initial, final, and maximum temperatures of two types of PS, purchased and waste. Their results showed slight differences in the degradation temperatures. It was observed that the purchased and the waste had a weight loss of 98.50 and 95.60% at a final temperature of 452 and 463 °C, respectively. Despite these differences, the maximum degradation temperature for both samples was about 420 °C.

Figure 1 visualizes box plots of data collected from the literature to visualize the distribution of virgin PS [69][70][71][72][73][74][75][76] and PS waste [77][78][79][69][71][80][81][82] degradation temperatures (initial, final, and maximum). The results contemplate data from TGA analysis of samples without catalyst in a nitrogen environment with a flow rate range of 10 to 100 mL min⁻¹ and a heating rate of 3 to 100 °C min⁻¹.

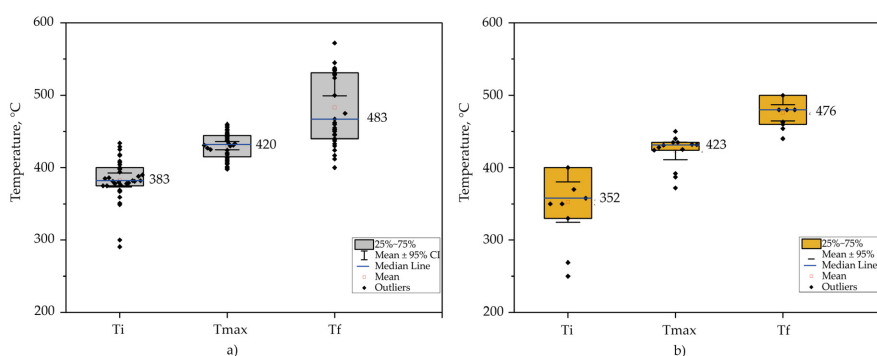


Figure 1. Degradation temperatures by TGA [initial (Ti), maximum (Tmax), and final (Tf)]: (a) virgin PS; (b) PS waste.

It can be observed in TGA results that the mean initial temperature of the PS waste is about 31 °C lower compared to virgin PS; an influential factor may be the impurities contained in the sample when the waste has not been pre-treated, such as in food. However, both maximum and final temperatures have similar behavior.

4.2. Thermal Pyrolysis of PS

Thermal pyrolysis is the simplest form of chemical recycling in which carbon-carbon bonds are broken by applying heat [76]. A simple process can recover valuable chemicals; an example is the study by Lu et al. [70]. In their experiments, pyrolysis was performed under an inert nitrogen atmosphere and heating of 5 °C min⁻¹ until reaching a temperature of 420 °C for 2 hours. The investigation yielded 76.24, 13.01, and 10.75% of liquid, solid, and gas, respectively. The results stand out because they achieved a single styrene component in their liquid sample, reaching a 73% yield.

4.2.1. Temperature

Temperature controls the cracking reaction in the polymer chain, which is why it is considered one of the most influential parameters in the pyrolysis process [83][84]. Studies report that, in the pyrolysis process, the production of a liquid with long hydrocarbon chains is improved at low temperatures. In contrast, the liquid yield decreases at high temperatures, and gas production improves. Furthermore, high temperatures lead to secondary reactions within the reactor, which reduce the obtaining of solid products[85].

A study of the influence of temperature on the thermal pyrolysis of PS waste is that of Verma et al. [86]. They evaluated the process at temperatures from 400 to 700 °C with 50 °C intervals and a heating rate of 15 °C min⁻¹. Their results showed that the pyrolytic liquid yield increases with increasing temperatures up to 650 °C. However, the liquid yield decreases at temperatures higher than 650 °C. Moreover, the same behavior was obtained even when a catalyst was used.

4.2.2. Reactor types

Reactor design is an essential parameter in the PS pyrolysis process since it affects how the reaction develops by influencing the method of heating, reactant mixing, residence time, and heat transfer.

Batch reactors are those where the system does not allow the flow of reactants or the exit of the products while the reaction is taking place; this allows the reactants to remain inside the reactor for a longer time, achieving a high conversion efficiency [87]. Moreover, these reactors are recognized for their simple design and ease of control of the operating parameters involved. Nevertheless, the main disadvantages of batch reactors are the requirements to refill feedstock and, in addition, that it is impossible to use them at a high production scale.

Unlike a batch reactor, in semi-batch reactors, it is possible to feed reactants and collect the resulting products simultaneously. However, coinciding with the disadvantage of the batch type, they are suitable for smaller-scale production. Finally, fixed-bed reactors are categorized as simple in design and are generally used as secondary reactors [88]. Yet, a two-stage process is not considered economically viable since the products obtained are very similar to single-stage processes [85].

Different types of reactors have been studied in the pyrolysis of PS to obtain products of interest in the chemical or petrochemical industry. The most used reactors are batch [89][78][76][90][91], semi-batch [92][68][76][80][82], fixed bed [93][94], and laboratory scale, which include basic systems such as a simple steel tube or using Pyrex material [79][70][81][95][96][97]. Lopez et al. [98] reported that reactor design is vital because an inadequate design leads to undesired reaction conditions, decreasing the quality of the products and promoting the formation of undesired by-products, such as solid residues or tar. Therefore, an optimal design must ensure a high heat transfer rate for rapid heating of the polymer and reliable temperature control to avoid operating problems due to the nature of the plastic melting process.

4.3. Catalytic Pyrolysis of PS

Thermo-catalytic pyrolysis has proven to be an alternative technology to reduce the impact of polymeric waste on the environment [99]. A catalyst is a substance that changes the performance of the chemical reaction without being altered in the process [100]. Using catalysts during a hydrocarbon degradation process can decrease the temperature and reaction time, increase the conversion rate, and promote a desired selectivity in the products [101]. These advantages make catalysts play an important role in waste pyrolysis processes [102].

The catalytic pyrolysis of PS has been less studied than other polyolefins; however, basic and acid catalysts have shown potential in the depolymerization of PS. Nevertheless, literature has shown differences using these catalyst types, such as the conversion efficiency of the products or their selectivity. An example is the study conducted by Inayat et al. [74], where they evaluated the catalytic pyrolysis of different PS wastes, experimenting with zeolite (HZSM-5) and MgO as acid and basic catalysts, respectively. Their results showed that implementing basic catalysts influenced the composition of the products slightly compared to those obtained by thermal pyrolysis. In conclusion, they indicated that the PS feedstock type affects the composition more than basic catalysts. Unlike acid catalysts, they significantly influenced the composition of the products.

Furthermore, in their experiments with zeolite, they added the operation mode as a factor (ex-situ and in situ). The results indicated that mixing the PS waste with the zeolite increases the production of waxes. In contrast, zeolite promotes mono aromatics' formation in the ex-situ mode, such as benzene, toluene, and xylenes (BTX). The authors concluded that acid catalysts have the potential to transform PS-based materials into higher-value compounds.

Figure 2 compares two types of catalysts used in the catalytic pyrolysis of PS. The analysis involves data collected from experiments with different types of bentonite [68][86][71][103][104] and zeolites [104][105] and evaluates the influence on liquid hydrocarbon yield and styrene formation. In this case, the effect of another operating parameter was not contemplated; the analysis includes temperatures from 300 to 600 °C. Despite the wide scatter of the data collected, the results show that zeolite achieves the highest performance for both liquid yield and styrene formation. The yields mean between 37.70 and 25.90% liquid and styrene, respectively, with the use of bentonite; on the other hand, with zeolite, it is possible to reach, on mean, up to 72.80% liquid and 63.40% styrene.

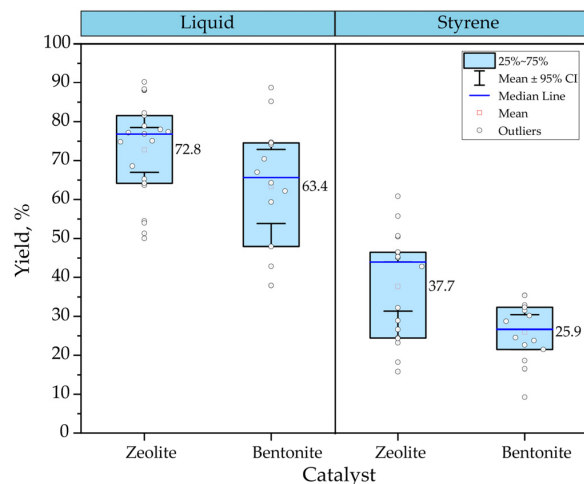


Figure 2. Comparison between zeolite and bentonite in the catalytic pyrolysis of PS.

Part of the data collected includes those reported by Dewangga et al. [104], in which they evaluated the behavior of bentonite and natural zeolite at different catalyst percentages, from 0 to 25%. Their results showed that, for both catalysts, the higher the catalyst percentage, the higher the liquid yield obtained. Specifically for zeolite, Rehan et al. [103] evaluated the performance of liquid yields with natural and synthetic zeolite in the catalytic pyrolysis of PS. Their results showed that using natural zeolites slightly favors the production of liquid hydrocarbon with 4% more than that obtained with synthetic zeolites. In contrast, gas production increases with synthetic zeolites, reaching approximately 10% more. Regarding styrene formation, the difference between the two types of zeolites is more notorious, obtaining about 60.80% and 15.80% for natural and synthetic zeolites.

5. Production of Fuels from PS Pyrolysis

Obtaining alternative fuels from plastic waste has received significant attention because it can solve the problems of the final disposal of this type of waste. In addition, the growing demand for energy worldwide, the depletion of oil resources, and the high cost of petroleum-based fuels have led to the development of alternative fuels from different types of plastic waste [106][107].

Despite that, PS pyrolytic oil has significant aromatic content, and its application as a fuel is questionable; however, its main advantage is its high calorific content. Therefore, studies on producing a combustible derived from PS thermo-catalytic pyrolysis have been developed [108][77][79]. Nisar et al. [97] evaluated the obtaining of fuel from the pyrolysis of PS waste; the experiments were carried out in a salt bath reactor using copper oxide (CuO) as a catalyst. They reported that using the catalyst reduced the temperature and time of the process and increased the liquid produced during pyrolysis. Liquid properties were compared with some standard values from fossil fuels, and finally, they concluded that the liquid obtained had a great potential to be a substitute for commercial fuels.

Budsareechai et al. [109] produced oil from plastic packaging waste in a bench-scale fixed pyrolysis batch reactor. Among their objectives, they studied the influence of heating rate on oil production; the results showed that the liquid yield decreases as the heating rate increases. Moreover, they concluded that oil derived from catalytic pyrolysis with commercial clay bentonite resulted in greater engine power, comparable engine temperature, and lower carbon monoxide (CO) and carbon dioxide (CO₂) emissions than uncatalyzed oils and commercial fuel in the gasoline range. **Figure 3** shows the distribution of carbons from C₅ to higher C₁₃ for the pyrolytic oil derived from this research, compared to diesel and gasohol 91. It is remarkable to identify the high content of aromatic components (C₅–C₉) in the PS oil derived from thermal pyrolysis with a value of 60%. In comparison, diesel only contains 2.18%, evidencing the significant difference between them. PS oil contains short-chain (C₅–C₉) and long-chain (>C₁₃) hydrocarbon chains, while medium-chain carbons characterize diesel.

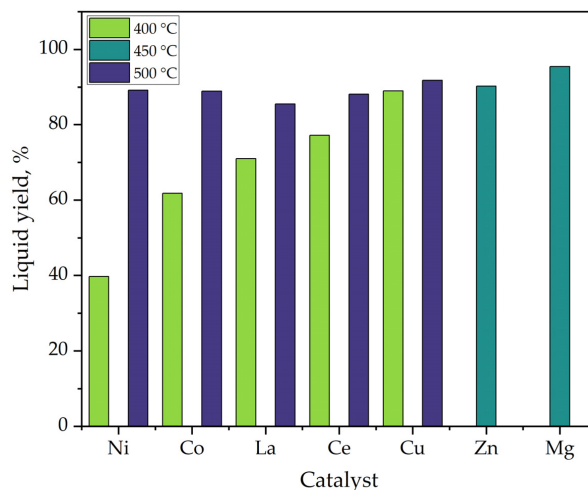


Figure 3. Carbon distribution of PS pyrolysis oil and commercial fuels.

Table 1 summarizes the comparison of the described properties of commercial fuels with pyrolytic PS oils found in the literature.

Table 1. Summary of fuel properties from PS pyrolysis and commercial fuels.

Fuel properties	PS Pyrolysis Oil						Commercial Fuels		
	Temperatures [°C]						Gasoline [97] [110][111][112]	Diesel [110] [111][112][109] [97]	Kerosene [97]
	150– 379 [113]	400 1 [97]	500 [109]	550 [79]	~695 [108]	~782 [108]			
Density, g cm ⁻³	0.95	0.79	0.85	0.92	0.95	0.98	0.72–0.78	0.80–0.87	0.78–0.82
Kinematic viscosity, mm ² s ⁻¹	0.92	1.34	1.71	0.88	0.97	1.26	1.08–1.17	1.90–5.30	1.54–2.20
Flashpoint, °C			48	30.50	79	79	>42	>48	
Pour point, °C	<–35		19		–39	–39		–6 to 19	
Calorific value, MJ kg ⁻¹			43.55		40.89	40.02		46.95	

¹ Mixed with CuO catalyst, 98:2.

The summary of fuel properties shows that the density of the liquid derived from PS at any temperature is relatively high, placing it out of the range of fuels such as gasoline, diesel, and kerosene. However, its value resembles marine application fuels, such as marine residual fuel (RMG-380) [79]. There is no trend in kinematic viscosity with pyrolysis temperatures; however, the values of 400, 500, and ~768 °C are within the established range for all commercial fuels compared. Gasoline and diesel must comply with a flash point above 42 °C, so pyrolytic oil, except for 550 °C, is compliant. Finally, one of the beneficial properties of PS pyrolytic oils is their high energy content. Due to the high aromatic content of PS oil, it is not recommended to be used directly as fuel. However, it is possible to blend it with other combustibles or biocombustibles to improve this property.

6. Co-Pyrolysis of PS and Biomass

Biomass is the biodegradable fraction of products, residues, and wastes of biological origin from agricultural activities, including substances of plant and animal origin, forestry and related industries, fishing, and aquaculture. Biomass is a biodegradable fraction of industrial and municipal waste of biological origin ^[114]. Biomass can produce alternative fuels, mitigating fossil fuel environmental and climate change impacts ^[115]. Moreover, co-pyrolysis refers to the process of thermo-catalytic pyrolysis in which different feedstocks are mixed, and the process is carried out. Several co-pyrolysis studies of biomass with waste plastics have been evaluated to enhance fuel production yields and improve fuel properties ^{[116][117][118][119][120]}.

Regarding fuel energy content, another reason for blending PS pyrolytic oil with biomass or conducting co-pyrolysis of both feedstocks is the increase in the calorific value of the biofuels. Reshad et al. ^[121] evaluated the energy content behavior of biofuels derived from the pyrolysis of rubber seeds (RSC) with PS at different operating conditions. Their results showed that PS pyrolytic oil contained 42.10 MJ kg⁻¹, and its high calorific value influenced the co-pyrolysis with the seeds. In the experiments with varied RSC: PS ratios, the results showed that the higher the amount of PS, the greater the increase in the calorific value of the biofuels, achieving up to 41.10 MJ kg⁻¹. This same behavior can be observed in **Table 2**, except for Niger and Karanja seeds, where a seed:PS ratio of 2:1 maximizes the calorific value of the biofuels.

Table 2. Energy content of co-pyrolysis of PS and biomass seeds.

Type	Seed: PS Ratio	HHV [MJ kg ⁻¹]	References
Grape seeds	100:0	35.49	
	90:10	36.90	^[122]
	80:20	40.40	
	80:20	40.90 *	^[123]
Coffee grounds	100:0	25.91	
	75:25	26.68	
	50:50	33.98	^[124]
	25:75	39.66	
Rubber seeds	1:0	32.25	
	2:1	37.48	
	1:1	37.61	^[121]
	1:2	41.10	

Karanja seeds	1:0	37.65	[125]
	1:1	38.88	
	2:1	42.18	
			[126]
	4:1	29.02	
Niger seed	8:1	26.53	
	1:0	35.87	[127]
	1:1	32.15	
	2:1	41.42	
			[126]
	4:1	31.82	
	8:1	29.77	

* LHV, MJ kg⁻¹.

7. Conclusions

Statistical analysis showed that low temperatures are recommended if a pyrolytic liquid is required; in contrast, increasing temperatures enhance the production of the gaseous fraction. The PS pyrolytic liquid can contain in its composition high yields of styrene, benzene, toluene, xylene, and ethylbenzene, among others. Styrene has the highest yield, reaching up to 84.74% [86]. The application of PS pyrolytic oil as a fuel is questionable due to its high aromatic content, differentiating it from diesel; however, it has a similarity of about 60% with gasohol [109].

Furthermore, the mixture of PS with biomass in a co-pyrolysis process favors the liquid production that biomass by itself would not achieve and, in addition, improves its properties, such as increasing the heating value to values close to 41 MJ kg⁻¹ and reducing up to 8.67% of its viscosity value [126][121]. It is concluded that the thermo-catalytic pyrolysis process is an effective recycling method to reduce a large amount of PS waste in the environment and convert it into high-value products for the chemical and petrochemical industry.

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