

# Catalytic Pyrolysis of Plastic Waste

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Plastic is a non-degradable material that can persist in the environment for long periods of time. On the other hand, it is a very special material with a great number of advantages: it is affordable, versatile, light, and resistant. Plastic originates from petrochemicals and contains mostly hydrocarbons and some additives, such as antioxidants, flame retardants, and stabilizers, which make the material very bio-undegradable. Pyrolysis can be thermal or catalytic. Thermal pyrolysis requires higher temperatures (from 350 to 900 °C), which can lead to a low molecular weight and low quality products. Gases evolved during pyrolysis have a high calorific value, and could be used in different gas machines and engines for the generation of electricity without any other treatments or modifications. The addition of catalysts overcomes the limitations of thermal pyrolysis, by reducing either the reaction temperature or the time. The addition of catalysts improves conversion, reduces activation energy of pyrolysis, and enhances the fuel quality. The most commonly used catalysts for the pyrolysis of plastics are zeolites, polyciliate components, and clays. Compared to thermal pyrolysis, the catalytic process has the advantage of a lower process temperature, a reduction of solid residues such as carbonized char and volatile fraction, shorter time process, high product selectivity, and high-octane-number products. Zeolite-based catalysts are the best option for the pyrolysis of plastic waste, and for co-pyrolysis of combined plastic waste/biomass.

catalyst

pyrolysis

plastic waste

## 1. Introduction

Catalytic pyrolysis involves the addition of a catalyst, with a higher conversion rate of plastic into oils and higher quality of the products, which is related to the lower temperatures and lower reaction time of the process [1][2]. Catalysts are also used in pyrolysis for the optimization of product distribution and increase of product selectivity, and provide yields similar to the conventional fuels, such as diesel and gasoline [3][4][5][6]. Catalysts are usually classified into two groups: homogeneous and heterogeneous. Generally, homogeneous catalysts are in a single phase (reactants, products, and catalysts are in the same phase), i.e., liquid solution, while the heterogeneous catalysts are in solid form [7][8]. The most used homogeneous catalysts for plastic pyrolysis are aluminum chloride ( $\text{AlCl}_3$ ), potassium hydroxide (KOH), sodium hydroxide (NaOH), and sodium methoxide ( $\text{CH}_3\text{ONa}$ ). Commonly used acidic catalysts are hydrochloric, phosphoric, sulphuric, and organic sulphonic acids. Homogeneous acid catalysts are preferred to base ones, because of the lower activity of the latter [7].

Heterogeneous catalysts are mostly in the solid form, and they are added to solid or liquid or gaseous compounds in the mixture [8]. Heterogeneous catalysts are of more common use, because of their easy separation from the liquid pyrolysis product, which allows the catalyst to be reused and regenerated [9]. Another important advantage of

heterogeneous catalysts over homogeneous ones is that they are non-corrosive. The surface area, porosity, arrangement of functional groups, and acidity are important features for the pyrolysis process [7][10]. Most used catalysts are the following: zinc oxide—ZnO; magnesium oxide—MgO; calcium-carbonate—CaCO<sub>3</sub>; calcium carbide—CaC<sub>2</sub>; silicon dioxide—SiO<sub>2</sub>; aluminum oxide—Al<sub>2</sub>O<sub>3</sub>; silicon dioxide alumina doped—SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>; zeolite Socony mobil-5—ZSM-5; zeolite; red mud; and fluid catalytic cracking—FCC [3][4]. The FCC catalysts are widely used in petroleum refineries for the cracking of heavy oils into gasoline and liquid oil petroleum [1][9]. The FCC contains three parts: zeolite crystals and a non-zeolite acid matrix that is made of silica-alumina with a binder in the structure [10]. Silica-alumina catalysts have Lewis acid sites, acting as electron acceptors, and Brönsted acid sites with ionizable hydrogen atoms. Acidity of these catalysts influences the production of liquid oils from the pyrolysis of waste plastics. When the acidity is very high, the production of liquid oils is reduced [1]. The presence of open pores in zeolites is the most important feature of this type of catalyst. Zeolites with high acidity are more active in the cracking process, thus increasing the production of light olefins and decreasing the heavy fractions [1][11].

## 2. Catalyst Structure and the Pyrolysis Mechanism

Zeolite catalysts have a crystalline structure that contains primary structural TO<sub>4</sub> tetrahedron units. T represents the central atom, mostly Si or Al, surrounded by O atoms connecting the units to each other. Interconnection between the building units leads to three-dimensional microporous structures with different geometries and structural characteristics [11]. Zeolites also promote hydrogen transfer reactions, due to the presence of many acid centers [12]. Silica-alumina catalysts and zeolites result in an increase of the gaseous yield. Lighter molecules, supporting gas formation, are obtained as a consequence of the increased cracking reactions and acidity [11]. Compared with zeolites, silica-alumina catalysts can be assumed not to contain stable crystalline structures with pore volumes larger than those in zeolites, which are microporous [13]. Many studies have been published on the pyrolysis of plastic waste polymers due to the advantageous microporous structure of ZSM-5 zeolite [14][15][16][17]. These papers also provide exhaustive surveys of the studies performed on the catalytic pyrolysis of plastic waste in the presence of zeolites (ZSM-5).

The activation energy is known to decrease in the presence of catalysts, thus suggesting an increase of the reaction rate, although no information is available on the value of the pre-exponential factor. As a consequence, the use of catalysts reduces the optimum temperature needed for the pyrolysis process allowing for energy saving, therefore reducing the most relevant cost for industries [18].

Catalysts can be placed either in the pyrolysis reactor (in situ) or in a separate catalytic bed (in-line). Catalysts, such as the protonated (H<sup>+</sup>) form of ZSM-5 or HZSM-5, protonated form of Y zeolite (HY), protonated form of beta zeolite (HB), and hierarchical H-style ultra-stable Y HUSY zeolite, promote the carbocationic cracking of pyrolysis volatiles and isomerization, oligomerization-cracking, and hydrogen transfer. The main properties of catalysts that influence the cracking performance are strength and porosity. During the catalytic processing of plastics, the polymer matrix melts upon heating and is dispersed over the catalyst surface. Mechanisms of catalytic pyrolysis also include chain scission, isomerization, oligomerization, H-transfer, and aromatization. The first step of polymer cracking is ascribed to adsorption of the reactant molecule on the acid site, where it undergoes protonation with the

formation of carbonium ions. This intermediate promotes the cracking of molecules. As a consequence, the reaction rate is mostly affected by acid site strength, density, and distribution. A catalyst's acid active site promotes cracking of olefinic compounds and hydrogen transfer reactions. When catalyst sites are strongly acidic, the formation of olefins (via end-chain scission) is favored, whereas, when the catalyst sites are weakly acidic, the formation of waxes (random scission) is enhanced. The obtained products (olefins or waxes) undergo further reactions to produce low-molecular-weight compounds [11]. In the study carried out by Cai et al. [19], the catalytic pyrolysis of various plastic types with  $\text{Fe}/\text{Al}_2\text{O}_3$  was studied in order to obtain high value hydrogen, aromatic chemicals, and carbon nanotubes as reaction products. This research showed that plastic polymers tend to decompose through a random scission mechanism, and ethylene obtained from polyethylene (PE) decomposition was more involved in the catalytic coke deposition with respect to propylene (from PP decomposition), which further increased the amount of solid carbon. Furthermore, high impact polystyrene (HIPS) and general-purpose polystyrene (GPPS) deposited more carbon than that produced during the catalytic pyrolysis of polyolefin plastics. This is explained by the difference in the molecular structure of the different plastic types, because of the presence of benzene rings in the HIPS and GPPS structures. When GPPS and HIPS are heated to their degradation temperatures, the large polymer moiety breaks up into smaller aromatic compounds such as styrene or styrene oligomers. When the catalysts are used, the ethenyl group in the styrene molecule is removed and benzene rings are accessible to produce high value hydrogen, aromatic chemicals, and carbon nanotubes as reaction products. At the same time, C-C and C-H bonds in the ethenyl groups are broken. On the catalyst, carbon atoms are dissolved and suddenly recombined by melting iron, thus forming graphitic carbon. The residual hydrogen atoms are responsible for the formation of low-molecular-weight gases. The heavier aromatic hydrocarbon compounds are decisive for the carbon-generating process in comparison to micro alkane and olefin molecules. Due to the higher degree of conversion into solid carbon, the amount of gaseous products obtained from polystyrene (PS) is lower than that produced from PE and PP [19].

Paula et al. [20] have performed thermal and catalytic pyrolysis of plastic waste (PE, PP, PS) with ZSM-5 zeolites and modified mordenite (MOR). Mordenite was treated with NaOH in order to improve the pore volume. By treatment with alkali, a mesoporous morphology can be produced in the catalyst, thus preserving its acidity. Mordenite has an irregular pore size, which could contribute to the occurrence of secondary reactions taking place on the catalyst surface. Reactions on the catalyst surface may contribute to bimolecular secondary processes and the production of aromatic compounds, which further lead to lower conversion rates and increase light fraction hydrocarbons. Low conversion rates can also be related to mono-dimensional channel systems and low mesoporosity, which limited the catalyst's access to polymer chains on the active sites. However, the treated ZSM-5 catalyst showed better performance in the catalytic processing of plastics compared to mordenite. Another study where the effect of small pores of the ZSM-5 catalyst was discussed is reported in [14].

### **3. Application of Zeolites for the Catalytic Pyrolysis of Plastic**

PE, polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET) were pyrolyzed in the presence of HZSM-5 zeolite in a tandem micro-pyrolyzer. Pyrolysis was conducted in situ and ex situ. The in situ catalytic

pyrolysis of plastics gave a higher amount of solid residue and led to the production of aromatic hydrocarbons, with the exception of PS. During the ex situ catalytic pyrolysis of PS, a high amount of aromatics was produced compared to the in situ catalytic pyrolysis, with a large production of styrene. In the case of PET, in situ catalytic pyrolysis generated a large amount of CO<sub>2</sub> and a small amount of CO, compared to the ex situ catalytic pyrolysis. Pyrolysis of PE and PP yielded comparable amounts of alkenes and alkanes in ex situ and in-situ catalytic pyrolysis, with the amount of produced alkene higher than the alkane yield during ex situ catalytic pyrolysis. These results show that in situ and ex situ catalytic pyrolysis of plastics have different reaction mechanisms [21]. Another study that also confirmed catalytic pyrolysis of the polymers to depend on the chemical and physical characteristics of the catalysts, and on the chemical and structural nature of polymers, was reported by Marcilla et al. [22].

Colantonio et al. [23] investigated the pyrolysis of plastic residue from a material recovery facility. The goal of this study was to investigate the effect of the HUSY catalyst and hydrogen zeolite socony mobil-5 (HZSM5) catalyst on the quality of the pyrolysis products. The plastic residue used as a feed was composed of 45 wt.% of PE, 30 wt.% of PP, 20 wt.% of PS, and 5 wt.% of PET. Thermogravimetric analysis showed that HUSY reduces the degradation temperature of all plastic polymers, with the highest effect on polyolefins. HZSM5 showed a significant effect on the degradation of PE. The pyrolysis reaction was carried out in a laboratory semi-batch reactor at 370, 450, and 650 °C, on the basis of TG analysis. The use of zeolites resulted in a reduction of the heavy oil fraction and inhibition of wax formation. HUSY showed the best results in regards to the monoaromatic yield, whereas, HZSM5 influenced the production of gases.

A noticeable improvement in the production of light oil to the detriment of tar was obtained in a recent study [23], which carried out catalytic pyrolysis of thermoplastics extracted from waste electrical and electronic equipment (WEEE) using various catalysts prepared from fly ash. The pyrolytic degradation of common polymers found in packaging materials was recently studied in [24]. The cracking temperature affects the type and number of the segments produced, with higher temperatures favoring the conversion of the polymer chains to monomers and the release of smaller molecules, and lower temperatures are more likely to produce oligomers. Faujasite (FAU) zeolites typically induced the formation of char due to their large cavity volume where secondary reactions can take place [25].

The pyrolysis of multilayered plastic and mixed resin plastic waste in the presence of the iron-modified zeolite ZSM-5 catalyst was examined in the study carried out by Kremer et al. [26]. The FTIR analysis showed that PET was present in the exterior part of the packaging, mixed with PE and PP. Some packaging also contained small amounts of PA. The study showed how the activation energy of pyrolysis decreases in the presence of a catalyst and that the catalyst lowered the content of polyaromatic compounds. Kinetic analysis of the pyrolysis of non-recyclable plastics and its influence on the product yield was studied by Kremer et al. [27]. In this study, plastic waste that was not mechanically recyclable was used. The plastic waste was separated from PVC, so that the main components of the used feedstock were PE, PET, PA, PP, and a very small amount of PS. Two different catalysts were employed in the pyrolysis process. The first catalyst was FCC, and the second one was iron (III) oxide (Fe<sub>2</sub>O<sub>3</sub>) loaded on the zeolite support ZSM-5 (Fe-ZSM). Pyrolysis was conducted in a fixed-bed reactor, and TG and DTG analyses were carried out at 5, 10, and 15 °C. The study concluded that the order of polymer

degradation was  $PS < PET < PP < LDPE < HDPE$ . The decomposition temperature of pure PS was in the range between 350 and 450 °C. PET degradation occurs in the temperature range between 390 and 470 °C, whereas PP, LDPE, and HDPE require the highest temperatures (450–510 °C) for degradation. In the presence of catalysts, the pyrolysis of plastic waste is faster and the formation of wax is evidently reduced [27]. Another kinetic study on the pyrolysis of waste plastic polymers (PP, PE, PET, and PS) and three synthetic mixtures, representing commingled post-consumer plastic waste output from recovery facilities, was reported by Tuffi et al. [28]. Volatilization that occurs in one step of mass loss is complete for all polymers tested, with the exception of PET, because of the formation of complex aromatic compounds [28].

Nanoporous MCM-41 (mobil composition matter no. 41), which has suitable properties for catalytic applications of polymers, such as the specific area and pore size, can be used for the degradation of PET [29]. The effect of pore shape on the catalytic performance of zeolites during HDPE degradation was investigated in the presence of several zeolites with different pore sizes. Good results (high conversion rates of HDPE) were obtained using BEA and MFI zeolites, due to their bent pores, which slowed down catalytic deactivation because of carbon deposit, defeating the formation of longer molecules. Sudden blocking of linear pores by a small amount of carbon deposit decreased the MOR zeolite's activity. FAU zeolite had large pores connected through supercages, leading to better mass transfer and a high yield of liquids. On the other side, MWW zeolite had numerous large pores that influenced the slow diffusion of cracked products, enhancing yields of small hydrocarbons. By taking all these findings into account, it can be concluded that the pore shape of the studied zeolites was a crucial parameter for the determination of catalytic activity and product selectivity in the degradation of polymers [30].

Catalytic pyrolysis of HDPE on HZSM-5 and iron-alumina pillared montmorillonite (FAMO) was performed in a conical spouted bed reactor. The acid sites of catalysts affect the activity and the cracking products. When the acid site is stronger, the produced hydrocarbon mixture is lighter. In view of the bulky nature of polymers, another important factor for the activity of catalysts is the availability of the acid sites. In the pyrolysis performed with the HZSM-5 catalyst, the high activity and high selectivity for gases could be explained by the strong acidity of the zeolite, related to the easy and full access to acid sites located on its outer surface. On the other hand, FAMO showed low selectivity towards gases, because of the lower acid strength of catalyst site. However, when pyrolysis is performed at a temperature of 550 °C, the catalyst showed a better performance in the production of diesel-range products [31].

Garforth et al. [32] have investigated the catalytic pyrolysis of HDPE at temperatures between 290 and 430 °C in a fluidized-bed reactor using HZSM-5, H-mordenite zeolite HMOR, HUSY, and SAHA catalysts. This study showed that initial cracking of HDPE has to be restricted to the outer surface and pore entrances of the zeolite, and the subsequent initial cracked products are decomposed within the catalyst. Zeolite catalysts, namely HZSM-5, HMOR, and HUSY, were more effective in the conversion of polymers into volatile hydrocarbons than SAHA. Another study focused on the influence of the zeolite catalyst (ZSM-5) and red mud was carried out by Lopez et al. [33]. ZSM-5 and red mud were tested on a plastic mixture simulating the municipal plastic waste. Pyrolysis was performed in semi-batch reactor at 440 and 500 °C, respectively. ZSM-5 produced a higher proportion of gases and liquids with

aromatics compared to the thermal process. Red mud also showed positive results in plastic waste degradation, although higher temperatures were required than in the process with ZSM-5 [33].

## 4. Application of Natural Catalysts for Plastic Pyrolysis

In order to make the catalytic pyrolysis more affordable, catalyst costs need to be reduced, because scaling up the process on the continuous scale requires a large amount of catalysts that further influences the overall cost of the process [7]. Naturally occurring silica-rich zeolites (clinoptilolite and mordenite) can be used for plastic pyrolysis [15]. Several studies closely examined the application of natural zeolites [34][35][36][37]. Natural zeolites frequently have impurities such as Na, Mg, Ca, K, Ti, and Fe that can promote further reactions, therefore, could not be used for the process on a larger scale [15].

Clays may be used as catalysts for plastic degradation. However, they require higher temperatures for pyrolysis than zeolites. Clays act like solid acid catalysts and they have been used in the petroleum industry before zeolites were discovered. Because of their affordable price and availability, clays were extensively studied and applied for the plastic pyrolysis process. The acidity and activity of clays can be enhanced by pillaring them (PILC) and, subsequently, by applying an acid treatment. Layered clay forms a two-dimensional porous network, with a microporous size larger than zeolites. PILC have a grid of voids and they are not as flexible as the parent clays [38]. Mixed plastics (HDPE, PS, PP, and PET) have been pyrolyzed in the presence of modified pillared clays (PILC) and Al-PILC [39]. Budsareechai et al. [3] have investigated the application of pelletized bentonite clay as a catalyst for the pyrolysis of PS, PP, LDPE, and HDPE. As previously stated, PS contains mainly aromatic hydrocarbons (gasoline-range), whereas longer aliphatic hydrocarbons (for diesel engines) are present in PP, LDPE, and HDPE. The pelletized form of catalysts eliminates the pressure drop and reduces the pyrolysis processing time. Furthermore, due to the high acidity of the bentonite catalyst, no wax is formed during the process.

Experiments were carried out by Auxilio et al. [40] on five zeolitic and clay-based catalysts (pelletized and powdered zeolite-based catalysts and clay) for the pyrolysis of virgin HDPE, HDPE waste, and mixed plastic waste. A two stage thermo-catalytic process was detected and monitored in a bench scale reactor, with a continuous feeding system. This research outlined important information for the possible upscaling process: diesel fuel can be produced by two-stage thermo-catalytic cracking, by optimizing the waste plastic composition. The acid strength of the catalysts is crucial for reaching a good selectivity of the hydrocarbon fraction, since a higher acidity leads to gasoline formation, whereas a lower-to-mild acidity leads to the formation of diesel. In addition, mesopore volume is an important factor for avoiding catalyst coking, especially when the mesopore volume is small and the coke formation is more prominent; a pelletized form of the catalyst is favorable over a powder form, because it avoids the large pressure drop in the reactive distillation column [40].

Five different plastic types (HDPE, LDPE, PP, PS, and PET) were mixed and pyrolyzed in a fixed-bed reactor (temperature up to 400 °C, 90 min). Natural catalysts (clays and dolomite) and a synthetic catalyst (pelletized zinc oxide) were used for the plastic pyrolysis. Based on the calculations of liquid and gas yields, the study has concluded that the use of natural catalysts does not give a significantly different amount of fuel yields compared to



synthetic catalysts [41]. Thermal and catalytic pyrolysis of PP, LDPE, and HDPE, as well as mixtures of these three materials, were carried out in a batch reactor, in the presence of calcium bentonite. The presence of a catalyst had accelerated the reaction and improved the quality of the condensable fraction. Furthermore, the reaction time decreased with the increase of the catalyst concentration. This means that the reaction rate increased with increasing the catalyst concentration, due to the high surface area and the catalyst's acidity. The ideal catalyst-to-plastic ratio is found to be 1:3 at 500 °C [42]. By using this optimal ratio, the highest yields of liquids were obtained: 88.5 wt.% for PP, 82 wt.% for LDPE, 82.5 wt.% for HDPE, and 81 wt.% for mixed plastics [42].

An interesting study was carried out by Nguyen et al. [43] in which the pyrolysis of PP waste plastics in the presence of natural-clay-mineral NCM, with LaFeO<sub>3</sub> nanoparticles, was studied. The degradation of PP was investigated using thermogravimetry (TG) and differential scanning calorimetry (DSC). The results showed that pyrolysis of PP takes place between 460 and 480 °C, and the formation of coke was observed on the surface of the catalyst. The study also reported that in the presence of the catalyst the cracking component consisted of 49.3% and 34.4% of alkenes and alkanes, respectively. Recently, a comprehensive overview of polystyrene pyrolysis was reported in the literature [44]. The effects of temperature, reactor type, and catalysts on the oil yield were also discussed. When bentonite catalysts are used for the pyrolysis of PS, mostly styrene compounds were formed. On the other hand, zinc catalysts promoted better selectivity of the end products, while the spent FCC catalyst was highly efficient in the production of liquid yields. Calcium oxide represents an economical and very effective catalyst in the depolymerization of PS into styrene compounds.

A modified natural zeolite was used by Miandad et al. [45] as a catalyst for the pyrolysis of plastics containing PP, PS, PET, and PE. In order to improve the catalytic properties, natural zeolites from Saudi Arabia were treated in a novel thermal activation process followed by an acid treatment with HNO<sub>3</sub>. The degradation of PET and PE require higher temperatures compared to other types of plastic. PE is a long chain branch structure and its degradation occurs via random chain scission, which requires a high temperature. The decomposition of PET follows the ester link random scission, where oligomers are formed as the resulting compounds. The highest amount of liquid oil is obtained via the catalytic pyrolysis of PS in comparison with those obtained through the catalytic degradation of PE and PP. Liquid oil produced from the catalytic pyrolysis with two selected catalysts showed high aromatic content with aliphatic and other hydrocarbon compounds. The obtained liquid oils had higher heating values (HHV) in the range of conventional diesel.

## 5. Application of Other Types of Catalysts for the Catalytic Pyrolysis of Plastic

An interesting study was conducted by Cocchi et al. [46], where the plastic film residue (PFR) from plastic waste recycling (polyolefin mixture residual) was pyrolyzed in the presence of coal fly ash (CFA) and CFA-derived zeolites, aiming at investigating the catalyst's effect on yields and the quality of the obtained oils. CFA-derived zeolites were produced from CFA by melting in the presence of NaOH, followed by a hydrothermal method NaX/CFA, and by a further acidification HX/CFA. The best results were achieved with HX/CFA, since it reduced the

endothermic contribution, and, at the same time, improved the gas and oil yield, as well as the selectivity towards the gasoline-range products.

Dai et al. [47] have studied the conversion of polyolefin waste into low aromatic naphtha in a microwave pyrolysis reactor. Various catalysts were screened in order to choose the best one for the conversion of HDPE into shorter-chain olefins with the smallest aromatic selectivity. Mesoporous silica SBA-15( $\text{SiO}_2$ )-supported metal catalysts (Al, Co, Ni, Zn, etc.) were tested for the pyrolysis process. Among the tested catalysts, Zn/SBA showed to be the most promising candidate for the pyrolysis of polyolefins. Integration of ZnO altered the mesoporous structure of SBA-15 (Santa Barbara Amorphous) catalyst and increased its acidity. This led to higher catalytic activity for the conversion of polyolefins into shorter hydrocarbon chains with a low aromatic selectivity. In the same study, zeolites were also tested and their performances compared with that of SBA-15 for the pyrolysis of polyolefins. In particular, USY and ZSM-5 were chosen for this reaction. In comparison to SBA-15 catalysts, zeolites seem to have a better cracking performance, but the main constituents in the liquid oil were aromatic hydrocarbons, which are not suitable for converting naphtha into plastic monomers [47].

Hydrocracking of LDPE was monitored over commercial zirconia nano powders with a fixed amount of sulfate and various amounts of Pt [16]. The addition of Pt on sulfated zirconia resulted in an increase in the catalyst's acidity and reduction in the crystallinity of  $\text{ZrO}_2$ . As a consequence, when higher concentrations of Pt were used, gasoline production was increased.

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