

A review on the pyrolytic conversion of plastic waste into fuels and chemicals

Original

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(Article begins on next page)

1 **A Review on the Pyrolytic Conversion of Plastic Waste into Fuels and** 2 **Chemicals**

3 Maddalena Laghezza ^a, Silvia Fiore ^b and Franco Berruti ^{a,*}

4 ^a Institute for Chemicals and Fuels from Alternative Resources (ICFAR), Department of
5 Chemical and Biochemical Engineering, Faculty of Engineering, Western University, London,
6 Ontario, N6A 5B9, Canada

7 ^b Department of Engineering for Environment, Land and Infrastructure (DIATI), Politecnico
8 di Torino, Corso Duca degli Abruzzi 24, 10129, Torino, Italy

9

10 *corresponding author: Prof. Franco Berruti (fberruti@uwo.ca)

11

12 **Abstract**

13 We reviewed 195 references discussing the pyrolysis of plastic waste, with the aim of analyzing
14 the technical aspects (feedstocks, process parameters, reactors, products, catalysts, kinetics,
15 and pollutants), the economic feasibility for the industrial implementation, and the
16 environmental assessment through Life Cycle Analysis (LCA). In the last decade, the scientific
17 community primarily studied individual virgin polymers (79% of the references) through batch
18 lab-scale tests, mostly at temperatures around 500°C (46% of the selected references).
19 Depending on the polymer (PE, PP, PET, PS), reactor type and operating conditions the relative
20 amounts of gas/condensable/solid products varied, whereas PVC was considered undesirable
21 due to the formation of corrosive gases. Liquid products are the most common targets, due to
22 their application as fuels. Several technical issues (such as the impact of residence time and
23 pressure, and low-cost catalysts) need further attention. Very few LCA studies explored plastic
24 waste pyrolysis, with insufficient results for a thorough environmental assessment of the
25 processes. Only seven references investigated the techno-economic feasibility of plastic waste

26 pyrolysis at full-scale, achieving results that are interesting but too scarce for any conclusive
27 evaluation. This represents the most significant knowledge gap identified in the review, as the
28 techno-economic sustainability is a fundamental factor for the technological transfer. Pyrolysis
29 has been proven to be a viable technology to convert plastic waste into high value-added
30 products that can be utilized as fuels, chemicals and/or building blocks. This review provides
31 guidance for further research and aims at stimulating the interest of the industrial world
32 regarding the possible implementation of this technology.

33

34 **Keywords:** chemicals; circular economy; fuel; plastic; pyrolysis; waste.

35

36 **Highlights**

37 - Analysis of feedstock, process parameters, reactors, products, catalysts, and kinetics

38 - “Real” plastic waste samples should be considered instead of virgin polymers

39 - Further efforts are needed to scale-up the research oriented to added-value products

40 - The environmental impacts have been scarcely investigated

41 - Technical-economic feasibility at full-scale is the key knowledge gap

42

43 **List of abbreviations**

44 AP: acidification potential; ADP: abiotic acidification potential; ASTM: American Society for

45 Testing and Materials; BTE: break thermal efficiency; BTX: benzene toluene xylene; BTEX:

46 benzene toluene ethylbenzene xylene; CNT: carbon nanotubes; CSTR: continuous stirred

47 reactor; EC: European Commission; EP: eutrophication potential; FBR: fluidized bed reactor;

48 FAETP: freshwater aquatic eco-toxicity potential; FCC: Fluidized catalytic cracking; GC-MS:

49 gas chromatography-mass spectroscopy; GHG: greenhouse gas; GWP: Global Warming

50 Potential; LCA: life cycle analysis; HDPE: high-density polyethylene; HHV: higher heating
51 value; LDPE: low-density polyethylene; MFR: Mechanically fluidized reactor; MPW: mixed
52 plastic waste; NPW: net present value; PET: polyethylene terephthalate; POCP: photochemical
53 ozone creation potential; PP: polypropylene; PS: polystyrene; PUR: polyurethane; PVC:
54 polyvinylchloride; PW: plastic waste; RMP: Recyclable materials rich in plastics; TEA:
55 techno-economic assessment; WEEE: waste from electrical and electronic equipment; WPO:
56 waste pyrolysis oil.

57

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88 **1. Introduction**

89 Plastic waste production, dominated by the packaging sector, accounts worldwide for 380 Mt/y
90 [1], with 29.5 Mt/y only in Europe [2]. According to the Ellen MacArthur Foundation [3],
91 global flows of plastic packaging in 2013 (corresponding to 78 Mt) were based on 98% virgin
92 feedstock, with an end-of-life management consisting in 14%-wt recycling, 14%-wt
93 incineration and/or energy recovery, 40%-wt landfilling and 32%-wt loss in the environment.
94 It is evident that actual waste management operations make plastic pollution one of the most
95 significant global environmental challenges. Across the world, international regulations and
96 policies have focused on plastic pollution prevention since the end of last century, with an
97 approach that evolved from the ban of specific plastic products towards the whole plastic value
98 chain [4]. In 2020, post-consumer plastic waste in Europe was destined to energy recovery (42
99 %), recycling (34.6 %) and landfill disposal (23.4 %) [2]. The Packaging and Packaging Waste
100 Directive 94/62/EC of the European Commission (EC) set targets on recovery and recycling
101 equal to 55%-wt for plastic packaging by 2030. The Single-use Plastics Directive 2019/904
102 imposed in Europe that from 2030 beverage bottles must contain at least 30%-wt recycled
103 plastic and introduced appropriate waste management operations for single-use plastic items.
104 In this framework, aiming at a Circular Economy of plastics and at reducing CO_{2eq} emissions,
105 there is an urgent need for novel and efficient recycling technologies to improve the material
106 recovery quota from plastic waste at the expenses of other management options. Recycling 1 t

107 of plastic could avoid 2 t CO₂ eq [5], therefore recycling 15 Mt of plastics per year by 2030 (i.e.,
108 achieving the 55%-wt recycling target set by EC) would save CO₂ emissions equivalent to
109 taking 15 M cars off the roads.

110 Plastics' recycling processes are classified into mechanical (re-extrusion of single polymer
111 materials) and thermo-chemical [6,7]. Mechanical recycling still faces several challenges due
112 to plastics heterogeneity, contamination, and degradation; its economic viability relies on the
113 market value of the recycled products, it is limited by the associated energy demand, and it can
114 be applied only to homogeneous plastic waste flows. On the other hand, thermo-chemical
115 processes (e.g., pyrolysis, liquefaction, gasification) can convert plastic waste into high value-
116 added products, such as fuels, char, and chemicals, depending on feedstock, process
117 parameters, reactor type, and catalyst.

118 The pyrolysis of plastic waste has been extensively explored in the literature, considering
119 different technical aspects of the process (e.g., type of reactor and operating conditions,
120 catalyst, and products, individually or in different combinations) [8–11], as well as its
121 environmental impacts [12–14] and economic assessment [15,16]. Chang [17] reviewed the
122 influence of plastic waste feedstock for pyrolysis oil production. Li [18] carried out a
123 comprehensive review on both technical and modeling aspects of plastic pyrolysis, and how
124 these data can be used in machine learning environments to optimize the process. Cheng [19]
125 applied statistical approaches, such as response surface methodology (RSM) and the Taguchi
126 method, to optimize pyrolysis parameters for oil production. Fisal [20] conducted an extensive
127 review on thermal and catalytic pyrolysis of plastic waste focusing on yields and oil quality.
128 Kusenberg [21] assessed the available pre-treatments, pyrolysis additives and oil upgrading
129 technologies to achieve high-quality petrochemical feedstocks. Al-Fatesh [22] evaluated
130 catalytic pyrolysis and its integrated processes (e.g., pyrolysis + gasification, pyrolysis +
131 plasma) as sources of hydrogen production. Catalytic pyrolysis has been reviewed tackling

132 different problems. For example, Christopher [23] studied the influence of catalyst on products
133 distribution and quality, mainly focusing on liquids. Yuan [24] extensively reviewed zeolite
134 catalyst and their influence on polyolefins cracking, products, yields, catalyst deactivation and
135 regeneration. Nabgan [25] carried out a bibliometric review on non-precious catalysts,
136 highlighting process strengths and knowledge gaps. To the best of our knowledge, a
137 comprehensive review of plastic waste pyrolysis, addressing at the same time the technical
138 aspects of the process, its scale-up and economic feasibility, and the environmental impacts
139 has not been published. The novelty of this review consists in a systematic and updated analysis
140 of the available scientific literature, based on three criteria: 1. the technical aspects of plastic
141 waste pyrolysis (e.g., feedstock, process, products, pollutants, catalysts); 2. its environmental
142 impacts (through life cycle assessment (LCA); and 3. the techno-economic feasibility of the
143 scale-up at the industrial-scale. This review has two main goals: (a) emphasizing the
144 technological advancements on plastic waste pyrolysis discussed in up-to-date literature, with
145 a specific focus on high value-added products; and (b) highlighting the existing research gaps,
146 to provide guidance for further research and facilitate the technology transfer to the industrial
147 world.

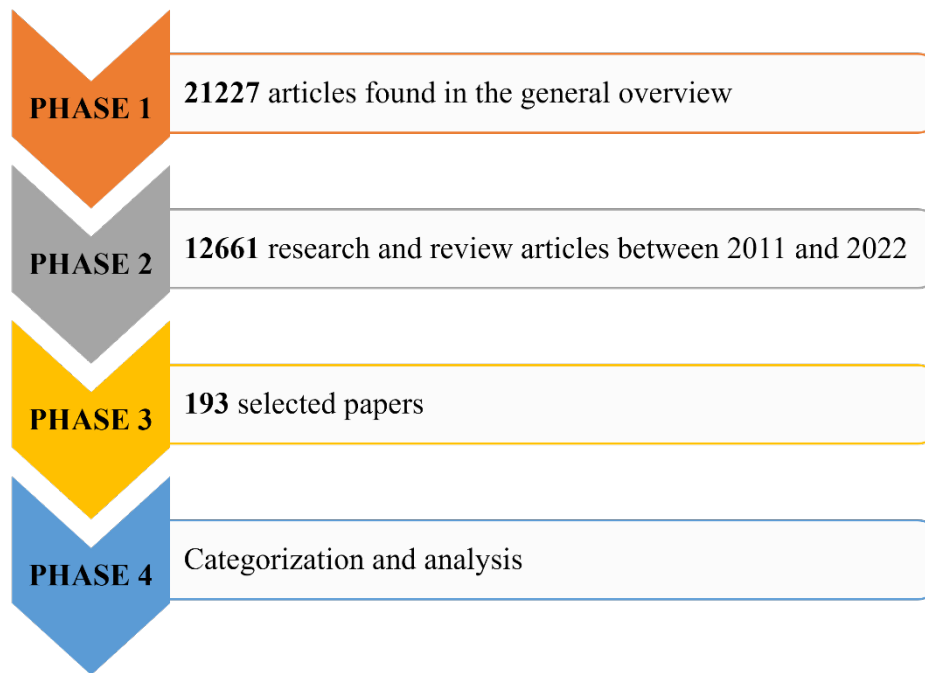
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149 **2. Methodology**

150 A systematic review of the most recent available literature, from 2011 to 2023, on the pyrolysis
151 of plastic waste has been carried out applying a four-phase procedure (Figure 1), as follows.

152 1. Scopus, Science Direct and Web of Science databases have been scrutinized adopting the
153 keywords “plastic waste, PE, PP, PS, PVC, PET” AND “pyrolysis, waste-to-energy, recycling”
154 AND “Life Cycle Assessment, LCA, techno-economic assessment, circular economy” in
155 various combinations.

156 2. The 21,227 references identified have been screened based on the publication language, type
157 of reference, and year of publication, thus selecting only research and review papers in English
158 published between 2011 and 2023 (12,661 references).
159 3. The titles and abstracts of the selected references have been analyzed to verify consistency
160 with the specific scope of this review study (i.e. references not including pyrolysis, or based
161 on co-pyrolysis have been excluded), thus finally obtaining 195 references.



162 **Figure 1.** Outline of the applied methodology for literature selection

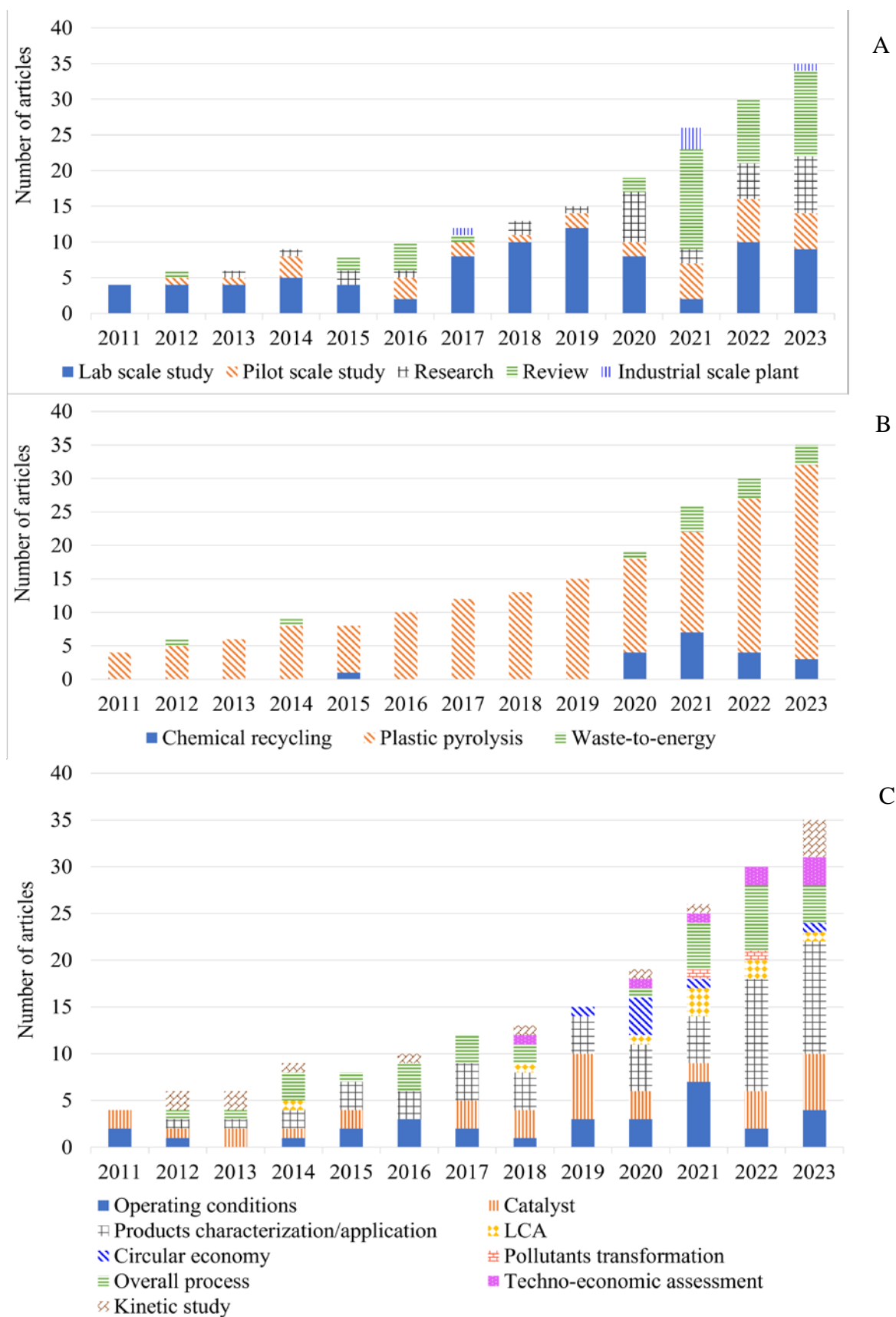
163 4. The selected 195 references have been inventoried according to the following categories:
164 type of study (research or review), scale of approach (lab-, pilot-, industrial-scale), type of
165 process (including processes for value-added products, such as gasification, pyrolysis,
166 hydrocracking, liquefaction, waste-to-energy,...), process features (operating conditions,
167 feedstock, products characterization and applications, catalysts, kinetics/mechanisms,
168 pollutants), life cycle assessment (LCA) study, and techno-economic assessment study.

169 3. Results and discussion

170 3.1. Analysis of the selected references

171 The overview of the 195 selected references (149 research articles and 46 review papers)
172 highlighted an intensification of the interest of the scientific community in the thermochemical
173 treatment of different plastic-based feedstocks from 2011 to 2023 (Figure 2A). In 2021, a
174 decrease in the number of research papers and an increase in review studies, compared to 2011-
175 2020, was noticed, probably because of the global COVID-19 pandemic and the lockdown
176 happening in several countries. Considering the scale of the research, lab-scale studies were
177 predominant (Figure 2A), while several pilot- and industrial-scale studies were observed just
178 in the last three years (20 out of a total of 36 in the whole period considered). This draws
179 attention to the urgent need for research applied at larger scale to identify optimized pilot/full-
180 scale process parameters and define a database required for the industrial implementation of
181 this technology. Considering the process type (Figure 2B), chemical recycling, plastic waste
182 pyrolysis, and waste-to-energy were the most common topics. The selected references were all
183 related to, or had sections, dedicated to pyrolysis; however, in 2020-2023 a slight increase in
184 papers on chemical recycling (+ 10%) and waste-to-energy processes (+ 7%) was noticed.

185



186 **Figure 2.** Categorization of the selected references about: A) approach, B) topic, and C) focus
 187 of the research.

188 The findings suggest that the scientific community deems it crucial to critically compare
189 pyrolysis with alternative technologies aimed at producing energy/energy sources or
190 chemicals. The conventional meaning of the term “pyrolysis” (i.e., thermochemical process
191 aimed at producing solid, liquid, and gaseous fuels) can be outdated nowadays [26]. It should
192 be mentioned that “modern pyrolysis” is an integrated (bio)refinery process, able to convert
193 many different waste flows – not only biomasses, into high-added value products that can be
194 valorized as fuels, chemicals and/or building blocks. Future research should acknowledge the
195 modern pyrolysis concept and use it as a driving force. In fact, pyrolysis is mature from a
196 technological point of view thanks to its extensive application in the domains of fossil fuels
197 and waste-to-energy processes. It is expected that, in the upcoming years, pyrolysis application
198 in the field of integrated (bio)refineries will likely become pivotal to achieve circularity in the
199 plastic waste management [26].

200 Considering the specific focus (Figure 2C) of the selected references under the “pyrolysis”
201 umbrella, the overall process, operating conditions, and products characterization/application
202 were the most researched subjects. The interest in catalysts was also evident in the last decade.
203 Kinetics and degradation studies decreased over the years, but there was a growing interest in
204 exploring the interactions during pyrolysis between different polymers and new green catalysts.
205 LCA, techno-economic studies, and circular economy evaluations were explored particularly
206 in the last five years. There is an urgent need for these evaluations to assess the feasibility of
207 industrial-scale plants depending on feedstock availability, transportation, products market,
208 and socio-economical aspects. Lastly, only in 2021 some interest in pollutant management has
209 been identified. This research area is very important, especially when industrial or mixed
210 plastics are fed to the pyrolysis process, and hazardous pollutants may form.

211 The following sections of this review analyze and discuss the contents of the selected
212 references. Specifically, the main findings and existing knowledge gaps about the technical

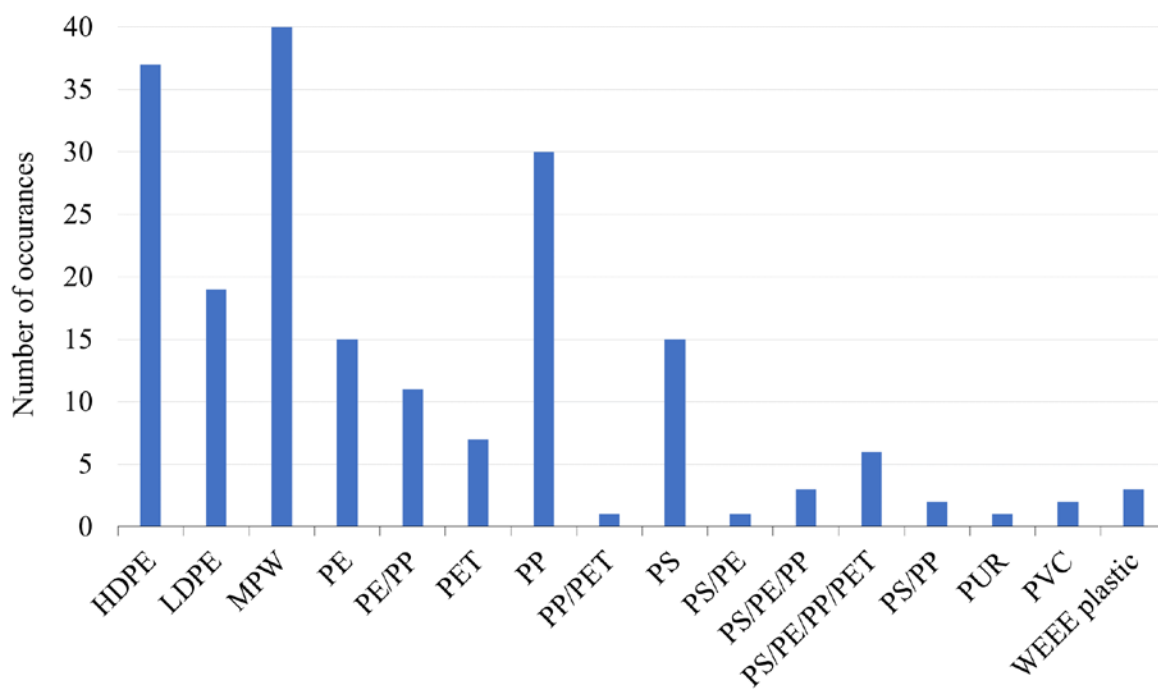
213 aspects of pyrolysis (feedstock, type of reactor, operating conditions, products, catalysts,
214 pollutants) will be described. Finally, the contents of the selected references describing LCA
215 studies and the techno-economic assessment of the scale-up of plastic waste pyrolysis will be
216 critically analyzed and discussed, as well as the applications at industrial scale.

217

218 **3.2. Feedstock**

219 Plastic waste is a suitable feedstock for pyrolysis since it can be thermally decomposed through
220 cracking reactions based on free radicals or involving different polymeric chains [27]. Within
221 the selected references, polyethylene (low-density (LDPE), and high-density (HDPE)),
222 polypropylene (PP), polyethylene terephthalate (PET), polystyrene (PS), polyvinylchloride
223 (PVC), and polyurethane (PUR) were the polymer types occurring individually or in various
224 combinations, also classified as MPW (mixed plastic waste) (Figure 3). The selected references
225 explored the pyrolysis of PE (37% of the total including both HDPE, and LDPE) i.e., the most
226 abundant polymer in post-consumer plastic waste [2], followed by MPW (21%), PP (16 %)
227 and PS (8%). These findings show that the scientific literature primarily focused on the
228 pyrolysis of individual polymers, usually tested as virgin materials in the experimental studies.
229 The research should move forward to explore the pyrolysis of real plastic waste, as the typical
230 mix of PE/PP/PVC/PS and associated additives found in post-consumer plastic waste, or the
231 pyrolysis of plastic waste deriving from agricultural and industrial activities. Plastic waste
232 thermal decomposition mechanisms depend on the type of polymeric material, as described in
233 the following sections.

234



235 **Figure 3.** Categorization of the selected references about the types of polymeric materials

236

237 3.2.1. Polyethylene

238 Polyethylene has different uses based on its density. Linear low-density PE (LLDPE) and low-
 239 density PE (LDPE) account for the 17.5% of the total resin demand in Europe and the main
 240 products are food packaging films, agricultural films, bags, and others [2]. Medium and high-
 241 density PE is crafted into pipes, shampoo bottles, cleaning products bottles, toys, and it takes
 242 the 12.2% of the European resin demand [2]. High-density polyethylene has a packed structure
 243 where linear chains of polymer with low branching degree (7 branching points per 100 carbon
 244 atoms) are linked together, hence high density is achieved. Low-density polyethylene presents
 245 a higher degree of branching (60 branching points per 1000 carbon atoms); thus, lower
 246 crystallinity makes it suitable for filming [27]. Both types of PE are decomposed through
 247 random chain scission, producing alkanes, alkenes, and dienes [27,28]. HDPE pyrolysis has
 248 been extensively investigated, demonstrating the possibility of producing high yields of
 249 condensable products (pyrolysis oil) [29]. The temperature should exceed 250 °C [30] to

250 achieve HDPE degradation, and condensable yields (up to 81%) were observed at around
251 350°C. The same authors reported lower (54%) condensate at higher temperatures which
252 favored gaseous products. Other studies reported an 84% condensable yield at 450°C in a
253 mechanically fluidized bed reactor operated in batch with a mixing velocity of 8 rpms and
254 heating rate of 5°C per minute [31], 88% in a continuously stirred reactor (CSTR) at 425°C in
255 argon atmosphere with an agitation speed of 125 rpms and feeding rate of 600 grams per hour
256 [32], 88% in a fixed bed reactor at 500°C in nitrogen atmosphere and heating rate of 5°C min⁻¹
257 [33], and 83% in a semi-batch reactor at 500°C in N₂ environment and heating rate of 1°C
258 min⁻¹ [34]. Considering LDPE, a condensable yield of 83% was observed in a semi-batch
259 reactor at 500°C in N₂ environment and heating rate of 1°C min⁻¹ [34]. A 79% condensable
260 yield was obtained in a mechanically fluidized bed reactor at 450°C operated in batch mode
261 with a mixing velocity of 8 rpms and heating rate of 5°C per minute [31] and a yield of 78% in
262 a fixed bed reactor at 300°C in nitrogen atmosphere and heating rate of 5°C min⁻¹ [33]. In
263 conclusion, the literature on PE pyrolysis reported mostly liquid products, with yields of 80-
264 88% within a relatively wide temperature range (350-500°C), through different types of
265 reactors (fixed/mechanically fluidized bed, semi-batch, CSTR).

266 3.2.2. Polypropylene

267 Polypropylene resin has the highest market share with a 19.3% demand to produce pipes,
268 automotive parts, microwave containers, candy wrap, food packaging and more [2]. PP is
269 synthesized in three different forms but only the isotactic one is employed commercially.
270 Isotactic polypropylene presents a highly crystalline structure (65%), melting point of 170°,
271 and decomposition temperature of 386.85°C [27]. PP degradation happens, as for PE, through
272 random chain scission. Condensable yields have been reported equal to approximately 70% at
273 400-500°C [30] in a batch reactor, 89% at 450°C in a mechanically fluidized bed reactor

274 operated in batch with a mixing velocity of 8 rpms and heating rate of 5°C per minute [31],
275 83% in a semi-batch reactor at 500°C in N₂ environment and heating rate of 1°C min⁻¹, and
276 83% at 450°C in a batch reactor with a heating rate of 15°C per minute [35]. Better yields are
277 achieved in mixed reactor due to improved heat transfer. In conclusion, PP pyrolysis reported
278 mostly condensable products with variable yields (69-89%) within a relatively narrow
279 temperature range (400-500°C), using limited types of reactors (mechanically fluidized bed
280 and batch).

281 3.2.3. *Polyethylene terephthalate*

282 Polyethylene terephthalate is mainly applied to produce bottles, such as water, soft drinks, and
283 juice bottles accounting for a 10% of the resin market in Europe [2]. PET decomposes into
284 polyene structures [27], the reaction starts by cleavage of the C-C bond in the backbone
285 followed by the cleavage of adjacent C α -O bonds [36]. Having PET the lowest volatile matter
286 content compared to the other polymeric materials considered as potential pyrolysis feedstock,
287 oil production is not the preferred route, while gaseous products [27] are favored. The reported
288 gas yields were 19 % in a tube reactor at 500 °C for 60 min in helium atmosphere [37], 76% at
289 540°C in a batch reactor in argon atmosphere [38], and 43% at 550°C in a different tube reactor
290 for 40 min in N₂ environment [39]. It was observed [40,41] that the oil fraction deriving from
291 PET pyrolysis is rich of benzoic acid, which is corrosive and thus hinders market applications
292 of the liquid products. In conclusion, the literature on PET pyrolysis reported mostly gaseous
293 products with highly variable yields (19-76%) within a narrow temperature range (500-550°C)
294 and corrosive pyrolysis oil, using limited types of reactors (tube and batch).

295 3.2.4. *Polystyrene*

296 Polystyrene accounts for 6.4% of the European resin market and it is mainly used in food
297 packaging, insulation materials, electrical and electronic equipment, eyeglasses frame and

298 more [2]. PS is a polymer consisting of a backbone hydrocarbon chain with phenyl groups
299 attached to every carbon. Depending on its method of synthesis polystyrene structure changes
300 showing a different degree of irregularities, hence the decomposition rate varies: 275.85,
301 332.85, 363.85°C are some of the values reported [27]. PS decomposition leads to styrene,
302 benzene, and toluene, resulting in a high production of oil [42]. Condensable yields have been
303 reported equal to 96% at 500°C for 20 minutes [43] in a fixed bed reactor in nitrogen
304 atmosphere and with a heating rate of 20°C min⁻¹, 94% at 450°C in a mechanically fluidized
305 bed reactor operated in batch with a mixing velocity of 8 rpms and heating rate of 5°C per
306 minute [31], 79% at 540°C in a batch reactor in argon atmosphere [38]. In conclusion, the
307 literature on PS pyrolysis reported mostly high yields of aromatic liquid products with variable
308 yields (79-96%) achieved within a relatively narrow temperature range (450-550°C), using
309 limited types of reactors (mechanically fluidized bed and batch).

310 3.2.5. *Polyvinylchloride*

311 Polyvinyl chloride is the main component of window frames, floor and wall covering, profiles,
312 pipes, cable insulation, inflatable polls and many more; it takes up 10% of the resin market in
313 Europe [2]. PVC is a mixture of chlorine (47.5%) and carbon (39.6%) deriving from
314 hydrocarbons [44]. PVC decomposition takes place by dichlorination [45]; chlorine is released
315 by β -elimination from the polymer chain to react with hydrogen and form hydrogen
316 chloride[46]. This reaction has low activation energy and, therefore, starts at lower temperature
317 (200-360 °C), meaning that it is possible to eliminate chlorine from the products by performing
318 a double stage pyrolysis process with a chlorine elimination step in between [47]. The literature
319 on PVC pyrolysis is not common, due to the formation of corrosive and toxic compounds. To
320 avoid them, PVC should be dechlorinated before pyrolysis [48], thus limiting the economic
321 sustainability of the overall process chain. Honus [49] reported yields of 53.6% for gas, 8.8%

322 for liquid, and 42.6% for solid products from PVC in a tube reactor at 500°C in helium
323 atmosphere. In conclusion, the literature on PVC pyrolysis is scarce, and it reported mostly
324 gaseous (53.6%) products, which are corrosive and toxic.

325

326 *3.2.6. Mixed plastic waste (MPW)*

327 Mixtures of polymers affect the overall decomposition [50,51], product distribution and quality
328 achieved during pyrolysis [52]. Increasing PS concentrations tend to yield more pyrolysis oil,
329 an increase of 5%-wt in oil yield has been reported when the polystyrene concentration
330 increases from 5 to 15%-wt [53]. However, the remaining plastic mixture affects the
331 composition and yields of the final product. Fixed bed reactors at 500°C are the most employed
332 at the lab scale to treat MPW. Under thermal pyrolysis conditions, yields are reported to be
333 31.7% gas, 36.7 oil, 1.7% char for a simulated mixture of PS (10%), PET (13%), HDPE (20%),
334 LDPE (27%), and PP (30%) [54]. The same set up and operating conditions yielded 13.7% gas,
335 65.9% oil, and 13.2% char for unwashed residual plastic waste DKR-350 [55], and 36.7% gas,
336 56.7% liquid and 6.6% solid for a rejected plastic fraction with an average composition of
337 18.6% PS, 56.1% PP, 12.6% PP film and 12.6% PE films [56].

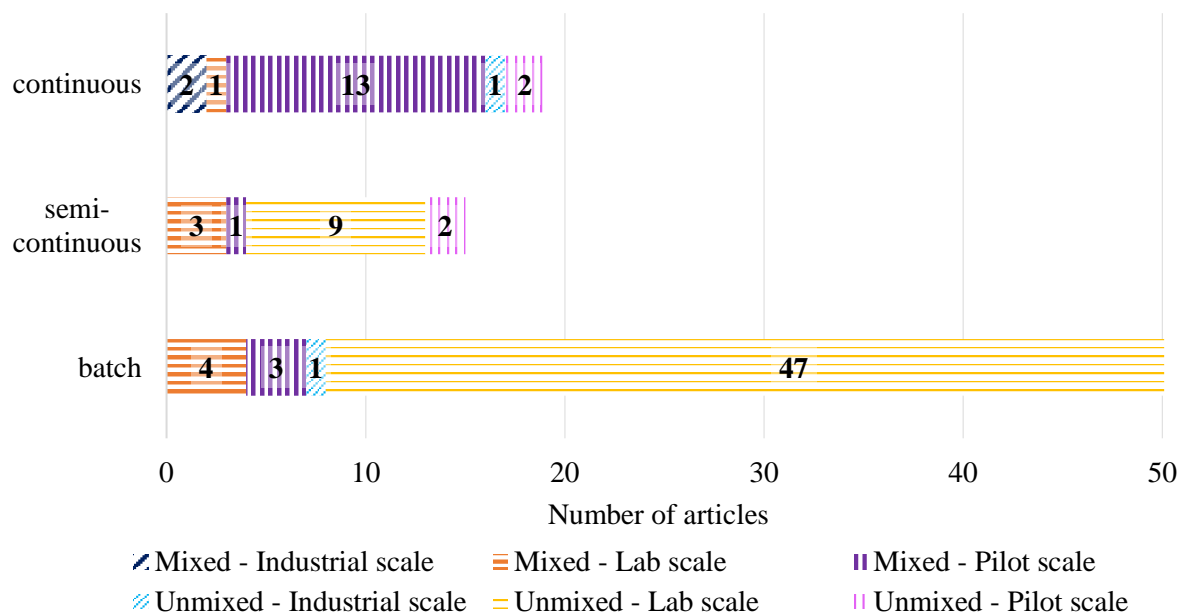
338 Up-to-date literature focused on virgin polymers (and their mixtures) unreliably mimicking
339 true mixed plastic waste. It is critically important to understand the effects of contaminants
340 present in real plastic waste and identify their possible catalytic properties as well as
341 detrimental impact, corrosiveness and toxicity. This knowledge is crucial for pyrolysis'
342 optimization and process industrialization.

343

344 **3.3. Reactor type**

345 Many reactors for plastic pyrolysis have been studied and described in the literature [29,57];
346 they differ about the feeding mode (batch, semi-continuous, continuous), presence/absence of
347 mixing, and overall design (fixed bed, mechanically mixed or fluidized bed reactors) (Figure
348 4).

349 Depending on the feeding mode, pyrolysis reactors are categorized as follows [58]. Batch
350 reactors (i.e., closed systems fed at the beginning of the process) are easy to design and operate,
351 thus very common in laboratory-scale studies [59]; they may be not suitable for large-scale
352 applications due to their labor intensiveness and non uniformity across batches [60], and
353 mixing is required if catalysts are used [60]. Semi-continuous reactors (i.e., systems
354 simultaneously fed and discharged for a discrete time intervals) improve the selectivity of the
355 process [61] and provide results that may be scaled-up. Continuous processes (i.e., materials
356 continuously flow in and out during the process), even if more complex in design and operation
357 compared to batch and semi-continuous, provide higher productivity [62], generate a more
358 constant product composition [63] and findings are easier to scale-up.



359 **Figure 4.** Categorization of the selected references about the type of process (feeding mode
 360 and presence/absence of mixing) and scale of the study.

361
 362 Moreover, 70% of selected studies did not apply mixing in their reactors. Continuously mixed
 363 reactors showed better results compared to unmixed (batch) reactors, as the mixing improves
 364 the heat transfer leading to higher conversion and better-quality products. Singh [64] reported
 365 yields of 16% gas, 24% liquid, and 60% solid for the pyrolysis of waste HDPE in a batch
 366 unstirred reactor at 500°C, while yields of 27.2% gas, 71.7% liquid, and 1.1% solid were
 367 observed from the same input material and operating conditions in a continuous stirred tank
 368 reactor [65]. Compared to unmixed reactors, continuous reactors are easier to operate at the
 369 industrial size, they present lower operating costs and yield a constant composition of the
 370 products [66].

371 Considering the design, fixed bed reactors (i.e., vessels containing a packed bed of material)
 372 are mostly applied in batch mode (18% of the references) due to their easy design and
 373 operation. However, they offer limited surface area for the contact between catalyst and
 374 feedstock [67,68]. To overcome this drawback and to enhance the life of the catalyst, two-step
 375 pyrolysis is implemented adopting fixed bed reactors as second reactor, whose feed is liquid or

376 gaseous [69]. This enhances capital and operational costs of the process, compared to single-
377 stage pyrolysis, but higher yields are not typically achieved. Considering single-stage fixed bed
378 pyrolysis of HDPE, Al-Salem [70] obtained 25% gas, 70% liquid, and 5% solid at 550°C, and
379 51% gas, 40% liquid, and 9% wax was obtained by [71]. Two-stage fixed bed catalytic
380 pyrolysis of HDPE showed yields of 18.2% gas and 66.3% liquid at 500°C with ZSM-5 as
381 catalyst [72], and yields of 36% gas, 45% liquid, and 10% solid at 600°C with Y-zeolite [73].
382 Mechanically fluidized reactor (MFR) and fluidized bed reactor (FBR) are the designs most
383 commonly used for plastic waste pyrolysis at the pilot scale (44% of the references) [74]. In
384 MFRs, the bed is fluidized by rotating paddles; in FBRs the injection of fluidizing gas (typically
385 nitrogen) allows the solid to be suspended and well mixed[58,75], improving the contact
386 between catalyst and feedstock and promoting heat transfer. Furthermore, MFRs and FBRs are
387 continuous systems, with lower operational costs compared to batch reactors [60]. The main
388 drawback of FBRs is bed defluidization, happening when melted plastic adheres on the bed
389 particles surface creating agglomerates and potentially leading to bogging the entire reactor.
390 To avoid this, conical spouted bed reactors -i.e., a subcategory of FBRs, have also been adopted
391 for plastic waste pyrolysis [58]. MFRs are very common in the literature, as they are less
392 affected by defluidization than FBRs, they offer good heat transfer characteristics[76] while
393 maintaining a simple design.

394 Microwaved-assisted pyrolysis has been applied to plastic waste pyrolysis [77–79]. Plastic
395 waste needs to be mixed with microwave absorbent material (such as carbon), and the
396 application of microwaves allows efficient heat transfer directly to the feedstock. This
397 technology has been scarcely explored at pilot scale, with few interesting applications [80,81].
398 In conclusion, most literature studies (see also Section 3.1) refer to lab scale and unmixed
399 (batch) reactors. In the last three years an increase in interest in pilot scale experimentation is
400 noted and it accounts for 52% of the research references. In these studies, 55% employ mixing

401 in the pyrolysis reactor. Further efforts are needed to scale-up the research to support the
402 transfer of the results to the industrial world, and single-stage mixed MFR design seems the
403 most appropriate.

404

405 **3.4. Operating conditions**

406 *3.4.1. Temperature*

407 Temperature is the most critical parameter controlling plastics' pyrolysis and reported the
408 highest Pearson Correlation Coefficient (0.857) for yields regulation [82]. Temperature
409 controls the cracking of polymers, thus it must be carefully monitored and adjusted to produce
410 the desired products, generally liquid or gaseous [83]. At lower temperatures, waxes are the
411 main product formed. Reaching the lower decomposition temperature is crucial to decrease the
412 formation of wax-like solid products. Increasing the temperature, the relative abundance of
413 liquid products increases, reaches a maximum and then decreases until the highest gas yield is
414 reached (Table 1).

415 The analysis of temperature values cited in the selected references (Figure 5) showed that
416 temperatures equal to 250-350°C (melting range for plastic) were associated to yields of 9%
417 gas, 89% liquid, 2% solid for HDPE, and 12% gas, 78% liquid, 10% solid for LDPE in a fixed
418 bed reactor [84]. The range 400-550°C has been deeply researched, with 500°C as most applied
419 value for plastics' pyrolysis (46% of the selected references) achieving yields of 0-66% gas, 8-
420 99% liquid and 0.1-82% solid products (Table 1). Temperatures above 600°C were utilized
421 when maximization of gas yields (>30%) was required.

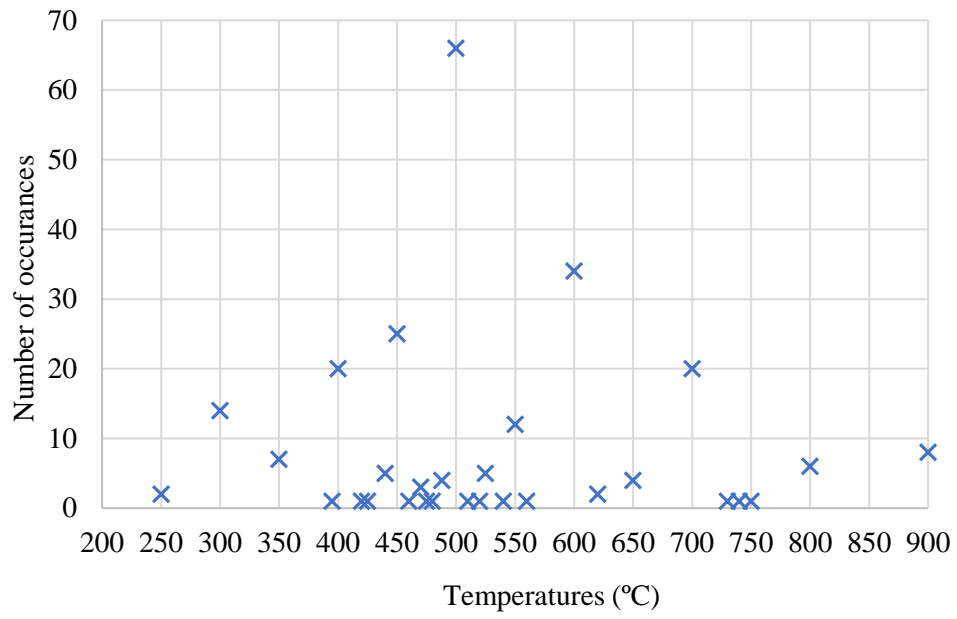


Figure 5. Temperature values applied in the selected references.

Table 1. Temperature influence on product yields in the selected references

Reactor	Feedstock	Temperature (°C)	Heating rate (°C/min)	Reaction time (min)	Carrier gas	Yields (%)				Reference
						Gas	Liquid	Wax	Solid	
Batch	PP	250	-	-	N ₂	29.05	57.27	-	13.68	[30]
		300				28.84	69.82	-	1.34	
		350				30.00	67.74	-	1.56	
	PE	400				31.07	63.23	-	5.70	
		300				36.25	30.70	-	33.05	
		350				17.24	80.88	-	1.88	
		400				45.29	57.17	-	0.54	
Batch	HDPE	300	5	-	N ₂	9.12	88.54	-	2.34	[84]
		400				9.91	87.87	-	2.22	
		500				10.22	87.62	-	2.16	
		600				10.34	87.55	-	2.11	
	LDPE	700				14.00	83.86	-	2.14	
		300				11.49	78.39	-	10.12	
		400				14.30	76.58	-	9.12	
		500				21.37	69.19	-	9.44	
		600				18.68	73.20	-	8.12	
		700				20.71	72.85	-	6.44	
Batch tube reactor	PET	500	-	60	N ₂	18.80	17.80	-	63.40	[49]
		700				31.60	14.40	-	54.00	
		900				56.40	14.60	-	29.10	
	PP	500				8.80	20.70	-	70.50	
		700				37.00	21.40	-	41.60	
		900				66.90	5.50	-	27.60	
	PE	500				5.50	13.00	-	81.60	
		700				35.10	13.00	-	51.80	
		900				65.10	8.90	-	26.20	
	PVC	500				53.60	8.80	-	42.60	
		700				57.20	5.80	-	37.10	
		900				60.50	6.70	-	32.70	
	PS	500				0.00	99.90	-	0.10	

		700				0.50	92.60	-	6.90	
		900				5.90	62.70	-	26.90	
	Plastic mixture (EU)	500				13.40	12.90	-	73.70	
		700				30.90	17.50	-	51.60	
		900				54.70	21.30	-	24.00	
	Plastic mixture (US)	500				10.00	11.70	-	78.30	
		700				31.20	21.60	-	47.20	
		900				56.20	19.70	-	23.70	
Batch tube reactor	Waste PP	450	15	60	N ₂	5.60	82.40	-	5.70	[85]
		488				5.80	85.60	-	3.10	
		525				8.40	80.90	-	2.60	
		600				27.00	63.00	-	2.50	
Batch tube reactor	Virgin PE	550	-	40	N ₂	49.00	26.00	25.00	-	[39]
		600				67.00	28.00	2.00	-	
	Virgin PP	550				61.00	31.00	8.00	-	
		600				61.00	37.00	2.00	-	
	Virgin PS	550				69.00	28.00	2.00	1.00	
		600				65.00	34.00	1.00	-	
	Virgin PET	550				43.00	-	44.00	13.00	
		600				48.00	-	40.00	12.00	
PP/PE	550	59.00	29.00	12.00	-					
	600	67.00	32.00	1.00	-					
Batch mechanically fluidized reactor	PP	360	10	20	N ₂	-	-	93.30	-	[86]
		380				-	-	86.70	-	
		400				-	-	70.40	-	
Semi-batch	Plastic waste mixture	460	20	30	N ₂	26.90	72.00	-	1.10	[87]
		500		30		34.00	65.20	-	0.80	
		600		30		56.20	42.90	-	0.90	
Semi-batch	PP	420	25	-	N ₂	8.30	88.60	-	3.10	[88]
		450				4.10	92.30	-	3.60	
		480				12.50	82.40	-	5.10	
		510				17.10	76.10	-	6.80	
Semi-batch		400	-	50	N ₂	3.00	86.00	-	10.90	[89]

	WEEE plastic	500		50		2.00	94.00	-	3.30	
		600		50		3.20	93.20	-	3.50	
Semi-batch	PE	500	-	35	N ₂	18.02	60.48	-	21.50	[90]
		600				20.00	63.80	-	16.20	
		700				16.82	68.02	-	15.16	
		800				18.33	72.32	-	9.35	
Conical Spouted Bed Reactor	LDPE	450	-	-	N ₂	20.00	-	80.00	-	[91]
		500				31.00	-	69.00	-	
		600				49.00	-	51.00	-	
	LDPE	450				20.00	-	80.00	-	
		500				32.00	-	68.00	-	
		600				51.00	-	49.00	-	
	PP	450				8.00	-	92.00	-	
		500				25.00	-	75.00	-	
600		50.00	-	50.00	-					
Bubbling Fluidized Bed Reactor	Polyolefin mixture	650	-	-	N ₂	37.00	48.00	-	15.00	[92]
		730	-	-		42.00	44.00	-	14.00	
Fixed bed reactor	Plastic mixture	450	55	45	N ₂	47.87	26.68	-	25.46	[93]
		475				59.99	28.26	-	11.75	
		500				65.75	32.80	-	1.46	
		525				69.98	28.80	-	1.23	
Fixed Bed Reactor	MPW	450	20	60	N ₂	10.80	77.60	-	11.60	[94]
		500				10.00	80.50	-	9.50	
		550				11.70	82.00	-	6.30	
		600				12.10	84.00	-	3.90	
Microwave reactor	Waste HDPE	500	-	60	N ₂	17.60	33.00	40.50	8.90	[95]
		560				23.80	47.40	24.50	4.30	
		620				40.20	37.60	15.70	6.50	
		740				74.70	18.20	1.30	5.80	

425 3.4.2. *Residence time*

426 The residence time of vapors during pyrolysis has extensively studied until 2010 as reported in
427 the literature review by Al-Salem [57]. During the time period (2011-2023) analyzed in this
428 work only one article on the influence of residence time on plastic waste pyrolysis was found.
429 Residence time can influence the relative abundances and composition of the products (Table
430 2); the production of low molecular weight fractions (i.e., non-condensable gases) is favored
431 by longer residence times. Papuga [93] studied the influence of residence time at a fixed
432 pyrolysis temperature of 500°C on a theoretical mixture of plastic waste (e.g., PP, HDPE, and
433 LDPE). They recorded an increase in gas and condensable yields (Table 2) at the expenses of
434 the solid product, finding that 60 minutes was the value which did not any longer compensate
435 the additional energetic and operational costs associated to higher residence times. The
436 influence of residence time on pyrolysis' products yields was scarcely explored. This issue,
437 crucial for an optimized operation of plastic waste pyrolysis, needs further efforts towards a
438 wide range of virgin polymers and real plastic waste samples. This parameter has been
439 neglected in the recent years but in light of the advancement of plastic waste pyrolysis it is vital
440 to resume its study and explore the effect on monomer yields for new plastic production.

441

442 **Table 2.** Product distribution in mixed plastic waste pyrolysis at 500°C depending on residence
443 time [93]

Residence time (min)	30	45	60	90
Gas (wt%)	66.13	65.75	67.91	68.17
Pyrolysis oil (wt%)	28.80	32.80	30.66	30.37
Solid residue (wt%)	5.08	1.46	1.43	1.47

444

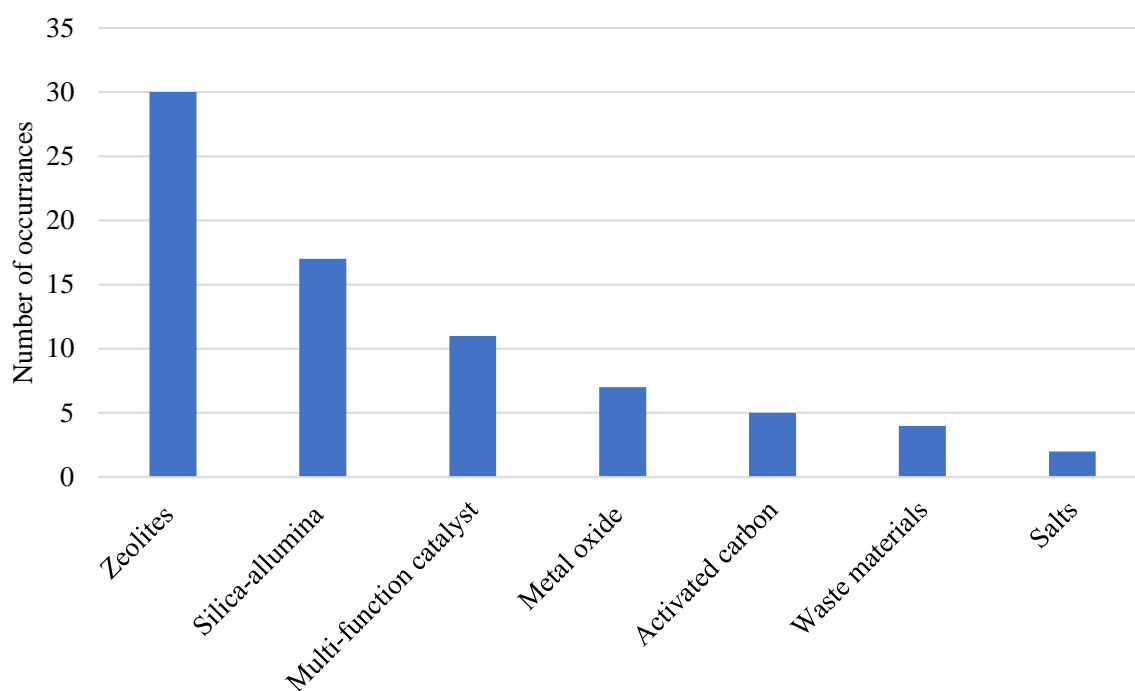
445 3.4.3. Pressure

446 Plastic waste pyrolysis generally happens at atmospheric pressure, to limit the operational
447 costs, thus pressure effects on product yields have not been fully researched. Parku [85]
448 investigated the effect of vacuum on PP pyrolysis, finding that in a batch reactor at 450-600°C,
449 vacuum pyrolysis produced 2-7% gas and 79-93% condensable yields, while the heating rate
450 (e.g., applying slow or fast pyrolysis) was the key parameter. Moreover, their investigation of
451 different heating rates (15 and 180°C/min) showed no influence on the overall yield of products
452 until 525°C. When the temperature was increased to 600°C, a drop in liquid production was
453 reported favoring the non-condensable fraction. In the last two years, literature studies have
454 been showing some interest in the effect of pressure on pyrolysis, with applications at the pilot
455 scale. Different pressure conditions were applied to virgin HDPE to understand if there was
456 any effect on degradation [96]. It was reported that vacuum conditions (0.1 bar) increased the
457 oil yield at all temperatures when compared to atmospheric and over pressure achieved with
458 nitrogen gas (2 bar) [96]. They also reported higher linear hydrocarbons yields in vacuum
459 pyrolysis, while N₂ pressurized environments lead to cyclization [96]. The same conditions
460 have been applied to polypropylene, low density polyethylene and real mixed plastic waste
461 [93]. LDPE followed the same trends as high-density polyethylene, while PP a lower pressure
462 increases both iso- and di-olefins in the oil but an over pressure significantly decreases the
463 diolefins content [97]. Pressure affects reactions taking place at the vapor-liquid interface, end-
464 of-chain β scission happens at this interface and it influences gas yields and oil quality [17]. In
465 conclusion, the pressure affects the degradation mechanism of plastics, which directly
466 influences oil production and composition. This is a major aspect when pyrolysis is used for
467 oil production, as better pyrolysis oil quality translates into lower upgrading costs and emission
468 limits conformity for direct application.

469

470 3.4.4. Catalysts

471 Catalytic pyrolysis is widely applied at both lab and industrial scale [26,98,99]. It was
472 developed to decrease the operating temperature (to limit the energy demand), avoid product
473 contamination from the feedstock, and for products upgrading by increasing the gas yield while
474 enhancing the quality of the oil products [100]. Catalysts can be homogeneous or
475 heterogeneous. The latter are the most applied because they can be easily separated from the
476 gaseous output and be regenerated and reused [101]. The key features of heterogeneous
477 catalysts to maximize the yield of gas products are [101]: high specific surface area (151-412
478 m²/g), micro-porous structure, crystalline structure, and acidity (0.176-115 mmol NH₃/g).
479 Catalysts, involved in 39% of the selected references, were particularly based on zeolites
480 (crystalline), silica-alumina (amorphous), and multi-function materials (e.g., mixture of
481 catalysts bind together to enhance functionality) (Figure 6).



482

483

Figure 6. Types of catalysts considered in the selected references.

484 **Table 3.** Effect of zeolite-based catalysts on pyrolysis products' yields in the selected references

Reactor	Feedstock	Catalyst	Catalyst dose ^a (%)	Temperature (°C)	Heating rate (°C/min)	Reaction time (min)	Carrier gas	Yields (%)			Catalyst effect	Reference
								Gas	Liquid	Solid		
Batch	MPW	Y-zeolite	10	500	-	-	N ₂	31.10	46.00	22.90	<ul style="list-style-type: none"> • Volatile product yields increased in the presence of a catalyst, while the full pyrolysis time necessity decreased. • Aromatic and cyclic compounds predominantly formed in the presence of catalysts. • Catalyst pore size and Si/Al ratio had the main role in their efficiency. 	[102]
		b-zeolite	10	500	-	-		32.50	44.60	22.90		
		HZSM-5	10	500	-	-		37.90	41.60	20.50		
Batch	HDPE	HUSY	10	470	-	45	N ₂	93.20	4.90	1.90	<ul style="list-style-type: none"> • Both catalysts show great gas yields (95%) and high quantities of olefins and aromatics. • Coke and solids deposited on the catalyst pores were easily removed by burning them in the air at 550 °C. • The catalysts were able to recover their activities and helped produce high gas yields after regeneration. 	[103]
		Hbeta	10	470	-	45		95.70	2.40	1.90		
Fixed Bed Reactor	WEEE plastic	Y-zeolite	100	500	10	-	N ₂	14.90	68.60	16.50	<ul style="list-style-type: none"> • Higher conversion of the styrene to other aromatic products, particularly benzene and toluene. 	[104]
Fixed Bed Reactor	LDPE	H-Y zeolite	50	500	25	60	N ₂	52.35	43.90	3.75	<ul style="list-style-type: none"> • Polyolefins yielded higher percentages of gaseous products (in the range 42 wt% – 45 wt%), while PS yielded a greater amount of liquids (~72 wt%). • The catalytic cracking led to the formation of liquid products, while thermal cracking of polyolefins produced waxes rather than the liquid fraction. • In the case of PS, thermal pyrolysis produced higher yields of liquid with greater amounts of styrene. 	[43]
	HDPE							48.56	43.00	8.44		
	PP							47.60	42.10	10.30		
	PS							24.71	70.70	4.59		
Semi-continuous	WEEE plastic	HUSY	33	400	-	50	N ₂	3.00	83.00	13.20	<ul style="list-style-type: none"> • Both zeolites HUSY and high-silica HZSM-5, employed at 400°C, showed a strong cracking effect, involving a decrease of the viscous fraction and an increase of light hydrocarbons content in the oil. • HUSY enhanced particularly the production of ethylbenzene, while HZSM-5 was able to produce a high concentration of monomer styrene. 	[89]

								2.00	91.00	7.10	<ul style="list-style-type: none"> • HZSM-5 showed the best performance in terms of the oil yield, comparable to the ones obtained at higher temperatures without catalysts.
Fixed Bed Reactor	Virgin HDPE	Y-zeolite	-	600	10	30	N ₂	36.00	45.00	10.00	<ul style="list-style-type: none"> • Y-zeolite catalyst produced a reduction of oil yield compared to thermal pyrolysis, but the composition of the oil was mostly aromatic hydrocarbons (~79%). • Loading transition metals on the Y-zeolite catalyst promoted the formation of aromatic hydrocarbons (80-95%) in the product oil and increased hydrogen gas production. • The addition of metals to the Y-zeolite generates high carbon deposition on the catalyst (14-26%). Increasing the metal content enhances this behavior.
		Ni-Y-zeolite	1					36.00	36.00	22.00	
		zeolite	5					36.00	43.00	18.00	
		Mo-Y-zeolite	1					38.00	31.00	24.00	
		zeolite	5					36.00	35.00	20.00	
		Fe-Y-zeolite	1					33.00	40.00	14.00	
		zeolite	5					38.00	30.00	22.00	
		Ga-Y-zeolite	1					37.00	36.00	18.00	
		zeolite	5					37.00	29.00	24.00	
		Ru-Y-zeolite	1					42.00	34.00	18.00	
zeolite	5	31.00	43.00	20.00							
Co-Y-zeolite	1	36.00	36.00	22.00							
zeolite	5	40.00	30.00	26.00							
Micro-wave reactor	Virgin LDPE		5	500	50	20	N ₂	43.30	56.40	0.30	<ul style="list-style-type: none"> • HY zeolite promotes gasoline components formation, resulting in a higher oil yield than the thermal cases. • The optimum oil yield (56.54 %-wt) and oil quality were achieved at the pyrolysis temperature of 500 °C, the catalysis temperature of 450 °C, and the HY to LDPE ratio of 1:10.
		HY	7					39.25	60.01	0.74	
			10					41.80	56.54	1.66	
			20					38.00	59.60	2.40	
Tube reactor	Virgin LDPE	HY	100	400	-	-	N ₂	24.40	70.10	5.50	<ul style="list-style-type: none"> • HY zeolite catalyst allows increasing product yields at lower temperatures and shorter reaction times. • Light oil produced is composed mainly of C7 to C14.
				500				26.10	68.20	5.70	
				600				36.80	57.20	6.00	
Batch	Waste LDPE	Zeolite	10	500	-	-	N ₂	28.00	15.00	57.00	<ul style="list-style-type: none"> • Higher conversion is obtained using LDPE (plastic bags) and zeolite with a conversion rate of 47%. • The liquid fractions obtained from the pyrolysis of LDPE in the presence of catalyst have shown superior quality as compared to oil obtained from LDPE without catalyst.
Fixed Bed Reactor	MPW	HY	In line	500	10	60	N ₂	48.9	37.5	13.6	<ul style="list-style-type: none"> • Both catalysts increase the gas yields and lowered the recovered oil • The main drawback of catalyst application in pyrolysis is coke deposition (3.4-6.2%) • The main product in the oil fraction is gasoline (~45%)
		HZSM-5							51.5	37	

$$^a \text{Catalyst dose} = \frac{\text{mass of polymer}}{\text{mass of catalyst}} \cdot 100 [\%]$$

486 The effect of zeolite-based catalysts on the pyrolysis' yields has been widely explored (Table
487 3). They have been applied to the pyrolysis of virgin polymers, of mixed materials as mixed
488 plastic waste (MPW), of the plastic fraction of waste from electric and electronic equipment
489 (WEEE) and using different types of reactors, different temperatures (400-600 °C), heating rates
490 (if any, 10-50 °C/min), and residence times (20-60 min). The catalysts dose was usually in the
491 range 1-10%, leading to high yields and efficient cracking of the products.

492 The main type of silica-alumina catalysts are SA-1, SA-2, and ZSM-5. They present different
493 acidity (SA-1 > ZSM-5 > SA-2) [29], and have different effects on the product distribution.
494 Owusu et al. (2018) reported a 20-60 % increase in gas production and 17-33 % reduction of
495 degradation temperature during the pyrolysis of various feedstock (HDPE, PP and PS) in the
496 presence of silica-alumina catalyst (catalyst/polymer ratio 1:10), and high-quality liquid
497 products even with lower yields. Similar results have been reported for ZSM-5 [108]: an
498 increase in gas production, lower liquid yield, but better oil quality.

499 Fluidized catalytic cracking (FCC) is a process used in refinery industries to crack heavy
500 hydrocarbons into lighter fractions [29]. The transformation is carried out with a catalyst which
501 is periodically regenerated. However, after extensive use, the spent catalyst (FCC catalyst) still
502 has reactive properties and can be used in pyrolysis processes [109]. Several studies [88,110]
503 reported an increase in gas production from 4.4% to 14% for thermal and 0.6 FCC catalyst to
504 PP ratio, respectively. Using waste materials as catalysts increases the environmental
505 sustainability of pyrolysis. Studies using activated carbon [69,111], red mud [61], cement [71],
506 fly ash [64,112], and metal oxides impregnated waste brick kiln dust [113] have been found.

507 In conclusion, catalysts have been extensively researched, mostly at the lab scale, implying
508 higher process costs and their required periodic regeneration. The actual knowledge gap
509 concerns the investigation of catalysts applications at a larger scale, aimed at optimizing the
510 economic balance of the overall process (e.g., cost of the catalyst and of its regeneration vs

511 higher production and better quality of high-value pyrolysis products at lower temperatures).
512 Alternative catalysts derived from waste materials, as FCC, Red Mud, and activated carbon,
513 should be further explored.

514

515 **3.5. Products**

516 The amount and quality of the gas, condensable, and solid products derived from plastic
517 pyrolysis depend on temperature, heating rate, residence time, and catalyst. In thermal
518 pyrolysis, an increase in temperature leads to higher amounts of gas products [57]. This also
519 happens in catalytic pyrolysis, but here the main reason for yields variations is the catalyst itself
520 [114], and solid yields are higher due to inorganic fractions of the catalyst and its impurities
521 [115]. A new study investigated the correlations between the polymeric constituents of the
522 feedstock and the pyrolysis products, highlighting how a bond-based classification of the
523 carbon atom could serve as the basis for describing the impact of complex polymeric feedstocks
524 in predictive models [116].

525

526 *3.5.1. Gas products*

527 Depending on the plastic type (i.e., polymer branching and bond strength), different gas yields
528 have been observed, increasing with temperature [117,118]. Honus [49] pointed out that PET
529 and PVC are the highest gas-forming plastics, polyolefins (PE and PP) yield similar quantities
530 of gas, and PS produces the lowest amount. Akubo [73] reported that using zeolites during
531 pyrolysis increased hydrogen and C₂-C₄ hydrocarbons. In addition, an increase in
532 catalyst/polymer ratio leads to higher hydrogen and lower weight hydrocarbons [67]. Non-
533 condensable gases deriving from plastic pyrolysis are high-value products made of hydrogen,
534 methane, ethene, ethane, propene, propane, butene, and butane, with the fraction of methane,

535 hydrogen and ethylene increasing with temperature [117,119]. The higher heating value (HHV)
536 of the gas depends on the plastic type [37,95,111]. PE and PP produce gas with HHV ranging
537 46-50 MJ/kg and 45-48 MJ/kg, respectively. PS showed low gas yield, with HHV equal to 42-
538 53 MJ/kg. Dechlorinated PVC produced gas having the highest HHV (51-59 MJ/kg), while
539 PET gas exhibited the lowest HHV (5-10 MJ/kg). Considering mixed plastic waste, the typical
540 HHV range for the pyrolysis gas is 43-66 MJ/kg [120–124]. Recently, a study converting waste
541 toys via catalytic pyrolysis at the laboratory scale (1 g of feedstock) showed that direct
542 production of hydrogen and methane is feasible and attractive when using a two-stage pyrolysis
543 set up[125]. Zajemska [126] treated the discarded fraction of municipal solid waste (containing
544 60% plastic waste) in a full-scale auger reactor at 900°C. They reported a gas composition of
545 22.2% CH₄, 18.4% H₂, 19.7% CO, 17.2% CO₂, and 8.6% C₂H₄. Validating that pyrolysis
546 syngas production for plastic waste is possible.

547 In conclusion, the existing literature reports the application of pyrolysis gas for process energy
548 recovery. However, it is a valuable product; as reference, natural gas and propane have HHV
549 equal to 52 and 50 MJ/kg, respectively [127]. Considering that hydrogen technologies are fast
550 expanding, future research should be oriented towards the operation of pyrolysis processes able
551 to maximize the content of hydrogen in their gas, paving the way for the application of the
552 optimized process at the industrial scale.

553

554 *3.5.2. Condensable products*

555 The amount of condensable product depends on plastic type, reaction temperature, time, and
556 catalyst [128–130]. Generally, plastic pyrolysis is carried out at the optimum degradation
557 temperature which can be defined by thermogravimetric analysis. For the main plastic types
558 (PE, PP, PS) this temperature is approximately 450°C to obtain over 50% degradation [131],

559 with PS showing the highest liquid production while PE only producing wax[132]. This
560 difference is due to their structure, with PS having a simpler structure than other plastic types.
561 Ahmad [30] studied PE and PP pyrolysis and defined the best temperature for condensable oil
562 formation as 300°C for PP (70%-wt) and 350°C for PE (80%-wt). The difference in quantity
563 achieved is due to polyethylene's better disposition to crack into liquid oil than polypropylene,
564 which tends to form gaseous products. Moreover, residence time influences liquid yield; slow
565 pyrolysis producing more oil and char while fast pyrolysis generating more gases [133].
566 Generally, oil is analyzed with GC-MS for compounds identification. Depending on the
567 process parameters, the number of the compounds varies. Furthermore, catalytic pyrolysis can
568 narrow the product range because of its intrinsic selectivity towards some compounds.
569 Polystyrene yields styrene at around 48%-wt [131]. The main constituents of the oil are
570 aromatic hydrocarbons; their high stability and resistance to cracking and hydrogenation
571 prevents their conversion into paraffin and olefins [134]. Polyethylene terephthalate liquid oil
572 contains almost 9.6%-wt of benzoic acid [41]. The presence of this acid, specifically its
573 corrosiveness, makes it unsuitable for fuel applications [41]. Polyvinylchloride yields low
574 liquid oil quantities, mainly because during degradation dehydrochlorination takes place
575 leading to gas formation, such as hydrochloric acid and chlorine [135]. The presence of these
576 compounds makes the liquid fraction not adequate to be used as fuel [136]. Polyethylene
577 produces mainly aliphatic compounds, followed by monoaromatics and polyaromatics [137].
578 Moreover, when the temperature increases, a decrease in aliphatic content was reported with
579 higher yields of aromatics [138]. The analysis of the Benzene-Toluene-Xylene (BTX) aromatic
580 fraction showed that the main component is benzene. Also, an increase in temperature leads to
581 a higher quantity of benzene, while toluene and xylene decrease [39]. Akubo [73] reported that
582 the addition of zeolite catalyst improved the oil quality: 79 %-wt of aromatic hydrocarbons is
583 achieved with Benzene-Toluene-Ethylene-Xylene (BTEX) as main 1-ring structures and

584 naphthalenes as 2-rings compounds. An increase in BTEX and aromatic compounds are also
585 reported with fly ash as a catalyst [112]. Similarly to PE, polypropylene produces aliphatic,
586 monoaromatic, and polyaromatic compounds, but it shows higher yields of the aromatic
587 fraction [85].

588 When pyrolytic oil is used as fuel, its physical properties must be assessed and compared with
589 those of gasoline and diesel. All these characteristics are determined following ASTM tests
590 and typical values are reported in Table 4. All the oils report similar fuel characteristics which
591 are comparable to diesel [110,139–141]. Plastic pyrolysis oil needs upgrading and purification
592 to find a final application. Distillation seems to be an effective method to clean and separate
593 plastic pyrolysis oil fractions [142]. The implementation of a distillation steps allows to
594 separate the lighter fraction which presents more gasoline-like characteristics [143–145].

595

Table 4. Plastic pyrolysis oil fuel properties

Feedstock	Pyrolysis parameters	API gravity	Density @ 15 °C (g/cm ³)	Kinematic viscosity @ 40 °C (mm ² /s)	Higher heating value (MJ/kg)	Ash (%-wt)	Cetane number (min)	Octane number (MON) (min)	Octane number (RON) (min)	Pour point (°C)	Flash Point (°C)	Reference
PP	Batch pyrolysis at 300 °C	33.03	0.86	4.09	35.5	0	49.3	87.6	97.8	-9	30	[30]
HDPE	Batch pyrolysis at 350 °C	27.48	0.89	5.08	30.6	0	46.0	85.3	95.3	-5	48	
PE	CSTR at 500 °C with distillation at 160 °C	-	0.706	-	32.2	-	-	-	88.8	-	-	[143]
	CSTR at 500 °C with distillation at 220 °C	-	0.811	2.23		-	56.54	-	-	-	92	
PP	CSTR at 500 °C with at 160 °C	-	0.707	-	34.3	-	-	-	91.7	-	-	[143]
	CSTR at 500 °C with distillation at 220 °C	-	0.797	2.04		-	42.23	-	-	-	79	
PP	Catalytic batch pyrolysis (10 %-wt catalyst) at 300 °C	34	0.761	1.899	43.4	0.039	-	-	-	-10	29	[110]
	Catalytic batch pyrolysis (10 %-wt catalyst) at 350 °C	35	0.75	1.964	43.5	0.037	-	-	-	-10	28.4	
	Catalytic batch pyrolysis (10 %-wt catalyst) at 375 °C	36	0.78	2.205	44.3	0.042	-	-	-	-9	28	
MPW	Batch at 370-380 °C	-	0.865	4.3	38.5	-	-	-	-	-	53	[140]
	Rotary kiln at 450 °C	-	-	7.1	44.0	-	-	-	-	9	-	
PW	Rotary kiln at 450 °C with distillation at 120 °C	-	-	0.71	42.3	-	-	-	-	<-30	-	[145]
	Rotary kiln at 450 °C with distillation at 200 °C	-	-	1.22	43.7	-	-	-	-	<-31	-	
	Rotary kiln at 450 °C with distillation at 300 °C	-	-	3.55	44.6	-	-	-	-	12	-	
PP	Fix bed reactor at 500 °C	56.71	0.7512	0.652	47.2	0.01	45.5	83.7	97.1	-	-	[144]
MPW	Batch at 500 °C	42.85	0.809	115.36	45.6	-	74.07	-	-	-	-	[139]
PP	Fixed Bed Reactor at 500°C	-	0.776	1.69	44.7	-	-	-	-	-	<24	[146]
PS		-	0.942	1.22	40.5	-	-	-	-	-	34	
Virgin Plastic Mix	Auger at 540°C	-	0.830	1.75	44.0	-	-	-	-	-	<20	[147]

	Auger at 540°C with distillation at 170-370°C	-	0.831	2.95	43.8	-	-	-	-	-	77
	Auger at 540°C	-	0.830	1.32	45.2	-	-	-	-	-	<20
MPW	Auger at 540°C with distillation at 170-370°C	-	0.832	2.95	43.8	-	-	-	-	-	63
Gasoline	Standard values	55	0.780	1.17	42.5	-	-	81-85	91-95	-	42
Diesel	Standard values	38	0.807	1.9-4.1	43.0	0.01	<40	-	-	6	52

[30]

598 The application of plastic waste pyrolysis oil (WPO) in diesel and gasoline engines has been
599 extensively researched and nine articles have been selected applying the methodology reported
600 in section 2. The results can be summarized as follows: i) WPO properties are comparable to
601 those of biodiesel and diesel [148], ii) blending of pyrolysis oil and diesel leads to higher break
602 thermal efficiency (BTE) and mechanical efficiency, with an increase in NO_x, CO, HC
603 emissions with increasing the percentage of WPO in the blend [149–151], iii) use of WPO
604 blends generates lower torque due to lower heating value and cetane number [152], iv) up to
605 20-30% WPO blend with diesel seems to be the optimal mixing condition as it improves the
606 fuel quality and presents results comparable to pure diesel [153,154], v) pyrolysis oil fractions
607 obtained from plastic waste pyrolysis that have been tested in ignition engines decrease fuel
608 consumption but increase NO_x and CO emissions [155], vi) WPO and diethyl ether blends
609 show improvements in BTE, more significant with the increase in percentage of pyrolysis oil,
610 lower NO_x and CO emissions, due to higher cetane number, and an increase in HC emissions
611 [156], vii) exhaust gas recirculation decreases NO_x emissions (-62.28%) if compared with
612 conventional diesel, but shows higher CO (+0.85%) and HC (+48 ppm) emissions[157]
613 Another application of pyrolysis oils is bitumen production by distillation and BTEX extraction
614 [112,158].

615 Liquid products from the pyrolysis of plastic waste are the most researched due to their possible
616 application as fuels without extensive post treatment. In accordance with a circular economy
617 vision, this is not a feasible approach, whereas employment of WPO as naphtha substitute for
618 virgin plastic production is a close-loop approach to be investigated. Moreover, to maximize
619 economic profitability, separation of 'low-volume high-value products from the oil, such as
620 waxes, should be considered.

621 3.5.3. *Solid products*

622 The pyrolysis process has been widely studied mainly focusing on pyrolysis oil and gas, since
623 they are the most valuable products from an economic point of view. However, most pyrolysis
624 processes produce some solid residues. Slow heating rate and high residence time tend to
625 produce more char when compared to fast pyrolysis [133]. When using plastic waste as
626 feedstock, both processes produce low char quantities. This is due to the high volatile content
627 of plastic waste (97-99%), low moisture (0-2%) and low ash (0.3-1%) [159,160]. The feedstock
628 initial composition influences the solid residue content; as an example, the char composition
629 derived from fast pyrolysis of HDPE included 51.4% volatile matter, 46% fixed carbon, 2.4%
630 moisture, and 0.2% ash [161]. They performed elemental analysis on the residue and found out
631 that the main elements are hydrogen and carbon with lower amounts of nitrogen and sulfur, the
632 calorific value of the char of 18.84 MJ/kg, and, due to its low sulfur content, they found it
633 suitable for combustion. Char can be used as additive in epoxy composite material, resulting
634 in enhanced electrical conductivity, hardness, tensile strength, and flame retardancy behavior
635 [84]. Addition of char to concrete has also been explored, with the strength improved by the
636 char ability to adsorb water favoring hydration products formation [162]. Another end use is
637 adsorption materials; plastic pyrolysis char has been employed to adsorb arsenic from water
638 and 99.4% uptake has been reported [163]. The absorption aptitude of char makes it as a
639 valuable candidate for CO₂ absorption. A CO₂ uptake of 78 mg g⁻¹ was demonstrated for an
640 incoming gas containing 40% CO₂ and at a temperature of 15°C [164]. At the same
641 temperature, an uptake of 63 mg g⁻¹ was reported for a CO₂ flow of 50 mL min⁻¹ for 2h [165].
642 In the fight for greenhouse gas emissions, pyrolytic chars can be an environmental low-cost
643 alternative to amines for carbon dioxide capture. A new approach is to use of pyrolysis derived
644 chars as material source for graphene and carbon nanotubes (CNT) production [60]. Dai [166]

645 in their review defined plastic waste pyrolysis as the most promising method for the formation
646 of carbon precursors essential for carbon nanomaterials growth.

647 Plastic-derived chars need to be further researched and new value-added applications, such as
648 graphene and CNT production, need to be explored. The market for these products is expanding
649 and the related revenues can greatly improve the economic sustainability of an industrial scale
650 plant.

651

652 **3.6. Life Cycle Analysis (LCA) of plastic pyrolysis**

653 Following the described literature review methodology, only ten LCA studies were found.
654 Three were review articles comparing different technologies for waste-to-energy technologies
655 and chemical recycling. Dastjerdi [13] reviewed all different approaches applied in LCA
656 studies: i) unitary functional unit is the most employed, followed by a waste generation-based
657 unit; ii) treatment/use of products and residuals, and collection/transport of waste are the most
658 utilized boundary conditions; iii) Global Warming Potential (GWP), terrestrial acidification,
659 human toxicity, terrestrial eutrophication, and photochemical oxidant formation are the most
660 studied impact categories; iv) foreground and background have a great influence on the strength
661 of the results.

662 Another study [167] compared LCA application to different chemical recycling techniques and
663 pyrolysis had the highest number of articles, probably due to greater data availability.
664 Moreover, comparison with mechanical recycling should not be performed since chemical
665 treatment should be recognized as an aid for the treatment of the residual and
666 mixed/contaminated fractions which cannot be mechanically recycled [167]. Andooz [168]
667 reviewed the potential benefit and drawback of plastic waste pyrolysis from a circular economy
668 point of view. They found out that developed countries are ready for industrial scale pyrolysis
669 applications, and this would increase the circularity and environmental benefits of the industrial

670 sector. Legislations need to recognize pyrolysis products as such instead of classifying them as
671 waste [168].

672 The selected references have been categorized according to the LCA approach and boundary
673 conditions (Table 5). The key issues are the feedstock and the use of real (primary) data or
674 literature (secondary) ones. Functional units, products application, boundary conditions, and
675 context are the basis of the LCA, and greatly impact the results because they define the system
676 for the analysis: changing it can completely shift the impact/benefits obtained. Al-Salem [169]
677 compared a material recovery facility plant with reprocessing for the recycling of plastic waste
678 (i.e., waste sorting plant with production of marketable products) with a pyrolysis plant able to
679 produce petrochemicals. Different substitution scenarios of the final market products have been
680 assessed: 100% substitution, 50% substitution and 0% substitution where all the products are
681 sent to landfill. For both plants, the best results in all impact categories are achieved when
682 100% substitution of conventional products is considered. Comparing the MFR and the
683 pyrolysis plant for a substitution factor of 1, the material recovery facility shows lower burdens
684 for acidification potential (AP), eutrophication potential (EP), global warming potential
685 (GWP), while pyrolysis performs better for the photochemical ozone creation potential (POCP)
686 presenting a negative value (environmental benefit). The most impacted category for the
687 pyrolysis plant is the acidification potential (>2000 kg SO₂-Eq) [169].

688 New product applications can change the impacts. Ahamed [170] compared the environmental
689 impacts of the conversion of 1 ton of real flexible packaging waste into either pyrolysis oil or
690 pyrolysis oil and multi-walled carbon nanotubes (MWCNs). They reported insignificant
691 impacts for eutrophication, terrestrial acidification, fine particulate matter formation,
692 freshwater consumption, land use, photochemical ozone formation, and stratospheric ozone
693 depletion [170]. Both scenarios present benefit for:

- 694 • Fossil depletion category due to pyrolysis oil generation, when MWCNs are produced
695 the benefit is greater (+14%) due to the displacement of acetylene in the production.

- 696 • Ionizing radiation, the pyrolysis oil offsets the emission of mining and extraction of
697 zeolites.
- 698 • Freshwater ecotoxicity and marine ecotoxicity, there is no emission of wastewater from
699 the process.
- 700 • Human toxicity (non-cancer), pyrolysis oil production accounts as an avoided burden
701 on the environment. For this category multi-walled carbon nanotubes production shows
702 a greater benefit, 6 times higher.

703 Plastic waste pyrolysis for oil production shows environmental burdens for terrestrial
704 ecotoxicity and climate change mainly due to catalyst extraction and HCl production during
705 pre-treatment. These two categories show positive effects for MWCNs production, the
706 synthesis process offsets 76% of the impact [170]. The most impacted category is the human
707 toxicity (cancer) where both scenarios have negative effects, catalyst mining and production
708 are the highest impacts [170].

709 Jeswani [171] analyzed the environmental impacts of chemical recycling through pyrolysis,
710 mechanical recycling, and energy recovery under three different perspectives:

- 711 • Waste, where the goal is the production of pyrolysis oil as main product and char as co-
712 product for pyrolysis and it is compared to energy recovery from incineration (30%)
713 and combustion of refuse-derived fuel (RDF). Mechanical recycling is not considered
714 in this case. The functional unit is the treatment of 1 ton of municipal plastic waste.
- 715 • Product, aiming at the production of virgin grade plastic. The functional unit is defined
716 as the production of 1 ton of virgin-grade plastic granulate (LDPE). The system
717 expansion approach is used to consider the avoided burdens from the incineration of
718 the feedstock and RDF, considering the loss of energy recovered. This scenario is
719 compared to the production of low-density polyethylene from naphtha in a cradle to
720 gate approach.
- 721 • Combined, here production and end-of-life treatment of MPW are considered. As base
722 to all scenarios, virgin plastic mix is produced from naphtha and the use is disregarded.
723 MPW is treated in a pyrolysis plant for oil production and upgrade to produce virgin-
724 grade LDPE as per the waste perspective. A comparison with energy recovery from
725 incineration and combustion of RDF, and mechanical recycling (sorting, separation,
726 milling, and extrusion) for production of mixed plastic granulate is carried out. The
727 functional unit corresponds to the production of 1 ton of mixed virgin plastic and
728 treatment at the end-of-life.

729 The environmental impacts resulting from the LCA are presented in Table 6. The waste
730 perspective for pyrolysis presents a 2.3 times lower climate change impact and a 1.8 times

731 better energy use benefits than the combination of incineration and RDF. In all the other impact
732 categories chemical recycling performs worse than energy recovery, in particular for
733 freshwater eutrophication where it creates a burden on the environment (+0.1 g P eq/t). The
734 production of 1 ton of virgin-grade plastic (LDPE) by thermochemical treatment lowers the
735 climate change impact by 124% and energy use by 38% than that of producing virgin polymer.
736 All the other environmental impacts are worse for LDPE produced by chemical recycling than
737 for fossil production. For the combined product and waste case, the results suggest that
738 chemical recycling has a slightly higher climate change impact (7%) than mechanical
739 recycling, but 42% lower than the energy recovery option. For energy use plastic pyrolysis as
740 a similar impact (+53.70 GJ/t) to energy recovery (+53.20 GJ/t) which is lower than mechanical
741 recycling (+56.10 GJ/t). On the other hand, all the other parameters considered (acidification,
742 eutrophication, photochemical ozone formation, and human toxicity) present greater impacts
743 for pyrolysis than the other alternatives.

744 Benavides [172] carried out a life cycle assessment on greenhouse gas emission of pyrolysis
745 of non-recycled plastics (NRP) for fuel production. In the study pyrolysis oil is allocated to
746 displace conventional low-sulfur naphtha, and char is either landfilled or sold as energy
747 product. Pyrolysis gas environmental impact is analyzed under two different scenarios: only
748 the amount of gases needed to heat the system is combusted and the remaining is sold as energy
749 product, or all the gas is combusted to generate heat and electricity for all the plant (no external
750 consumption) and the excess of electricity is sold to the national (USA) grid. The analysis
751 shows that selling char as energy product instead of landfilling it results in lower GHG
752 emissions for every scenario considered. Combustion of flue gases to cover the heat
753 requirements of the process and sell the remaining as energy product produces lower
754 greenhouse gases emissions, in this case when char is sold as market product then the lowest
755 impact is achieved (78 gCO₂ eq/MJ diesel). This translates into a 14% reduction in emissions

756 if compared to conventional low-sulfur diesel production (91 gCO₂ eq/MJ diesel). Das [173]
757 compared the two previously described studies [171,172] aiming to assess if circularity (e.g.,
758 chemical recycling) is the better option for plastic waste pyrolysis. Plastic to plastic and plastic
759 to fuel life cycle assessment of greenhouse gasses emission is carried out in two different
760 geographical areas: USA and Europe [173]. They reported that, based on a cradle-to-gate
761 approach, fuel production by pyrolysis emits lower GHGs compared to plastic production in
762 the U.S. Under this approach, the opposite is true for the European scenario, plastic to plastic
763 shows lower emissions than plastic to fuel. A reason for this is the twice as high LDPE yield
764 obtained by the pyrolysis plant in this case which accounts for more credits resulting in
765 environmental benefit. In contrast, if a cradle-to-grave boundary is implemented, then plastic
766 production has lower emissions than fuel production for both geographical contexts. The reason
767 for this is the increase in emission from diesel combustion that exceed the one associated with
768 chemical recycling [173].

769 The application of LCA during the design and scale-up of a pyrolysis plant has shown great
770 importance in accounting for the environmental impacts of the process. Performing an LCA
771 study in an iterative way on a pilot plant aiming to improve its environmental performance for
772 the industrial scale application showed that i) replacing electrical heaters with flue gas heat
773 exchangers; ii) using the light hydrocarbons produced in the preheat burner and generator; iii)
774 using the flue gas as heat source to dry the feedstock; reduces all the environmental impacts
775 [174]. The reductions compared to the base system are substantial; Abiotic Depletion Potential
776 (ADP) - fossil (65.8%), AP (52.2%), EP (48.8%), Freshwater Aquatic Eco-Toxicity Potential
777 (FAETP) (44.8%), GWP (44.3%), POCP (56.6%) [174].

778 The low number of available references reporting LCA studies is self-explanatory. The results
779 show that plastic waste pyrolysis creates environmental benefits for the energy use/fuel
780 depletion category. Climate change is impacted but the burdens are lower than energy recovery

781 from plastic waste and fossil plastic production. All the other impact categories are negatively
782 affected by pyrolysis, in particular eutrophication potential and human toxicity score the worse
783 impacts in all studies. Catalyst use in pyrolysis seems to aggravate all the environmental
784 impacts due to mining and preparation. More research in this area is urgently needed to
785 understand the environmental impacts of a full-scale plastic pyrolysis plant located in a specific
786 geographical context, to lessen negative impacts and draw attention to hotspots.

Table 5. Life Cycle Assessment model parameters

Feedstock	Data	Functional unit	Process	Products' application	Boundary conditions	Study context (country, energy mix, target market)	Reference
Plastic as a fraction of MSW	Secondary	Input based - 1000 tons per annum of PSW	Low-Temperature Pyrolysis on MRF site	Petrochemicals and energy	Foreground comprises of the plant and background considers transportation, energy, water, fuels, etc.	United Kingdom, UK energy mix (NG 44%, Hard coal 28%, Nuclear 22%), Greater London Area (Greenwich, Lewisham, Westminster, Bromley, and Exeter in Devon)	[169]
Mixed plastic rich in PE	Secondary	Unitary - 1 ton of feedstock	Thermal cracking - RT7000 designed model	Petrochemicals and fuels	Cradle to grave	United Kingdom, UK energy mix, not defined	[174]
Real flexible packaging plastic waste	Primary and secondary for missing data	Unitary - 1 ton of feedstock	Pyrolysis and catalytic reforming of the vapors	Oil upgrade to fuel quality, multi-well carbon nanotubes production from non-condensable gases and by-products uses for energy	The Boundary system starts at the waste treatment facility	Not specified	[170]
Mixed plastic waste	Primary and secondary for missing data	Unitary - 1 ton of feedstock	Pyrolysis	The oil used as naphtha alternative	Collection and transportation of MPW from households, sorting, pyrolysis, purification, and product use. The remaining MSW is incinerated for energy recovery.	Germany, EU energy mix in 2013 (48% renewables)	[171]
		Output-based - 1 ton of plastic granulate of virgin-grade quality (LDPE)		Production of virgin-grade quality plastic pellets	No incineration, expansion approach used to define avoided impacts.		
Mixed plastic waste	Secondary	Combined - 1 ton of mixed virgin plastics and their end-of-life treatment	Pyrolysis	Different applications depending on the scenario	Three scenarios for end-of-life treatment: 1) pyrolysis, 2) mechanical recycle + MSW incineration, 3) Refuse derived fuel + MSW incineration	Not specified	[175]
		Unitary - 1 ton of feedstock		Three scenarios: 1) high-value chemicals production, 2) energy recovery, 3) monomer production	Foreground comprises of the plant and background considers raw material extraction, transportation, energy, water, fuels, etc.		

Mixed plastic waste	Primary and secondary for missing data	Unitary - 1 ton of feedstock <hr/> Output-based - 1 ton of fuel	Pyrolysis	Two scenarios: 1) flue gas is combusted, and the electricity is sold as a product; 2) the excess flue gas is sold as product. In both cases, liquid products replace naphtha and char is sold as energy product.	The boundary system starts at the waste treatment facility	United States of America, U.S energy mix in 2016 (33% renewable)	[172]
Recyclable materials rich in plastics	Primary and secondary for missing data	Input based - 3928 million tons per year	Pyrolysis	Petrochemicals and fuels	Foreground comprises of waste treatment, chemical production, utility processes, and background considers transportation, energy, water, fuels, etc.	Germany	[176]

788

789 **Table 6** - LCA results of chemical recycling, mechanical recycling, and energy use for three different scenarios. The waste perspective results are
790 referred to 1 ton of MPW, the product prospective results are referred to 1 ton of virgin grade LDPE, and the combine perspective results refer to
791 1 ton of mixed virgin plastic production and treatment at the end-of-life. Positive values = environmental burdens, negative values = environmental
792 benefits [171]

Environmental impacts	Unit	Waste perspective		Product perspective			Combined perspective	
		Chemical recycling	Energy recovery	Chemical recycling	Virgin plastic (from naphtha)	Chemical recycling	Mechanical recycling	Energy recovery
Climate change	kg CO2 eq./t	+739.00	+1716.00	-450.00	+1890.00	+2130.00	+1990.00	+3650.00
Energy use	GJ/t	-26.60	-15.00	+42.90	+69.40	+53.70	+56.10	+53.20
Acidification	Moles of H+ eq./t.	-6.00	-23.00	+6.70	+3.00	+2.79	2.30	0.56
Freshwater eutrophication	g P eq./t	+0.10	-20.00	+45.10	+3.40	+3.70	-0.20	-18.30
Marine eutrophication	kg N eq./t	-0.04	-0.86	+2.30	+0.80	+0.73	0.55	-0.13
Photochemical ozone formation	kg NMVOC eq./t	-0.40	-1.90	+5.60	+2.50	+2.60	+2.10	+0.80
Human toxicity	1'000'000 Tox pts/t	-5.80	-8.30	+5.05	+0.19	+1.94	+0.96	+0.16

793

794 **3.7. Techno-economic assessment**

795 The largest knowledge gap resulting from the literature review concerns the exploration of the
796 economical sustainability of the scale up of a plastic pyrolysis process to the commercial scale.
797 In fact, only seven papers have been found in the literature following the methodology reported
798 in section 2. Fivga [177] modeled a baseline and three scale-up scenarios for a pyrolysis plant
799 treating mixed plastic waste in a fluidized bed reactor at 530°C to produce heavy oil substitute.
800 Pyrolysis gases and char are used as heat source to sustain the process. The baseline case
801 considers a throughput of 100 kg/h, the scaleups have capacities of 1,000, 10,000, and 100,000
802 kg/h, respectively. Aspen HYSYS is used to run the techno-economic assessment (TEA) and
803 the following results are obtained: i) the base case scenario is not economically sustainable
804 during its life (20 years) and it never reaches profitability; ii) the three scale-ups present
805 positive Net Present Value (NPV) at four years, one year, and within the year, respectively, for
806 increasing size; iii) the main parameter affecting the overall economic feasibility is the fuel
807 production rate which is directly linked to the fuel production cost. On the other hand, Lubongo
808 [178] reported payback periods of 9.58 years and 7.31 years for plant size of 2,500 kg/h and
809 4,167 kg/h, respectively. The basic plant operating conditions are comparable to the previous
810 one: mixed plastic waste (50% PS, 25% PP, 15% LDPE, and 10% HDPE), 500°C, 1
811 atmosphere, residence time 30 min, and fluidized bed reactor technology. The desired product
812 is pyrolysis oil which undergoes separation for production of light and heavy oil, pyrolysis gas
813 is used as heat source for the plant, and the char is landfilled. However, the different locations
814 (England and New York State) and the use of silica alumina as catalysts in the latter study led
815 to longer payback periods.

816 Another study compared two different product applications to study their influence on the
817 economics of the plant [179]. Open-loop and closed-loop pyrolysis configuration of mixed
818 polyolefins were considered: in the first case, waxes are the final product while naphtha is the

819 target in the second case. For both cases, non condensable gases as used as energy source to
820 sustain the process, char is not included in the analysis. The TEA showed that wax production
821 result in a shorter break-even period, even if further purification of the exiting stream is
822 required, due to the higher product value (34% increase income). This scenario has more robust
823 results since the price fluctuation are lower when compared to the oil market [179]. The cut off
824 sizes are 70 kton/year and 115 kton/year for open-loop and close-loop, respectively. Riedewald
825 [180] preformed a techno-economic assessment for a MPW pyrolysis process using a molten
826 zinc bed reactor (PyroPlast). The goal is pyrolysis oil production, while the gas is used as
827 energy source for the process and the char is disposed in landfill. They considered Belgium as
828 the plant location and a processing capacity of 40,000 ton/year of plastic waste. Under these
829 conditions they obtained a positive NPV starting from the first year of operation.

830 Pacheco-López [181] compared the economic feasibility of pyrolysis and incineration of MPW
831 (40% PE, 35% PP, 18% PS, 4% PET, and 3% PVC). The aim of the pyrolysis plant is the
832 production of light a heavy oil, the gas is used for heat production and the char is not accounted
833 for. Pyrolysis with oil separation and upgrade is more profitable (198.18 €/t MPW) than
834 incineration (177.79 €/t MPW), and it presents a lower capital cost 42.8 M€ compared to 45.6
835 M€. Another study evaluated the effect of pre-sorting of the feedstock on the economic
836 feasibility of the process [182]. Feeding sorted HDPE/PP bales to the pyrolysis plant results in
837 a feedstock price of 240 \$/t which leads to an unprofitable system. The feedstock price needs
838 to be lower than 200 \$/t for pyrolysis to be economically sustainable [182].

839 A new study aimed to evaluate the use of pyrolysis oil as energy source in China [183]. The
840 predicted increasing future plastic waste production results in an increase pyrolysis oil
841 production from 24.66 Mt/y in 2020 to 37.01 Mt/y in 2028. The results show that using the
842 upgraded pyrolysis oil for electricity generation is economically profitable with a net present

843 value of 8.8 M\$ after 20 years, a payback period of 10.6 years, and levelized energy cost of
844 0.0752 \$/kWh.

845 When considering difficult waste materials, a simulation on PET pyrolysis was found. Shahbaz
846 [184] simulated pyrolysis using Aspen plus and performed a techno-economic and
847 environmental analysis of the process. The results showed that a temperature of 750°C results
848 in the highest operating cost 2.255 M\$/y against 2.249 M\$/y for an operating temperature of
849 150°C, but it presents the best environmental conditions with CO₂ emissions of 86 kg/h
850 compared to 160 kg/h for the lower temperature.

851 In conclusion, the limited literature studies available show the feasibility of plastic waste
852 pyrolysis scale up to the industrial size. They need to be based on real experimental data and
853 use unsorted contaminated waste to clearly understand the future of plastic pyrolysis.
854 Simulations are the base of the advancement since they can estimate the best operating
855 conditions and related impacts in a time efficient manner. More studies are required, since the
856 economic sustainability of the process is greatly influenced by the location, operating
857 conditions, products application, market availability, and products price.

858

859 **4. Applications at industrial scale**

860 Commercial plastic pyrolysis plants are starting to appear all over the world. Companies are
861 very protective of their technologies. Scarce data can be found on the process itself and no
862 information is typically available on products quality and composition.

863 Plastic Energy™ has two operating plants in Spain. They can treat 5,000 t/y of a PE/PP/PS
864 mixture with maximum 10% contamination [185]. Their patented technology [186] melts the
865 plastic feed which is then feed to a semi-batch mechanically fluidized reactor at a temperature
866 higher than 350°C. The pyrolysis vapours are condensed in a two-stage condensation system.

867 The plant yields 72-75% TACOIL™, 18% syngas, and 8-10% char [185]. The oil is sold to
868 petrochemical industries, the gas is used as a heat source for the process and the char is sold to
869 construction industries.

870 Regenyx is a pyrolysis plant located in Tigard, Oregon, USA, able to chemically recycle
871 between 50-100 ton per day of polystyrene [187]. According to the patent [188] the pyrolysis
872 plant receives a chipped feedstock which is then mixed, compacted, and densified prior to
873 entering the reactor. The pyrolysis vessel is a twin-screw reactor that allows for the movement
874 of the liquid plastic mix at the beginning and extraction of the solid residue in the end. The
875 pyrolysis vapours are then quenched and condensed. Oil yields of 81% are claimed [189].

876 The US based company Brightmark has developed a pyrolysis system (Plastics Renewal™)
877 able to convert all post-consumer plastic waste (#1-7) [190]. Very limited technical information
878 is available on the plant. Plastic waste is collected, shredded, pelletized and pyrolyzed. The
879 Ashley facility can treat 100,000 ton/y of plastic waste and produces 18 million gallons of ultra-
880 low sulfur diesel fuel and naphtha and 6 million gallons of wax [191].

881 Recycling Technologies patented a pyrolysis plant for mixed plastic waste (7,000 ton/y)
882 conversion into a chemical feedstock (5,250 ton/y) called Plaxx [192]. According to the patent,
883 the pyrolysis technology is based on a laboratory scale fluidized bed reactor with defined a
884 height to width ratio - operating at 400-600°C with a bed mass of 2.5-3.5 kg [193]. The
885 technology is not implemented at the commercial scale yet, but the construction of the plant at
886 Binn Eco Park in Perth, Scotland has already started [194].

887 Klean Industries [195] is an international company focused on products recovery from
888 hydrocarbon-rich wastes. Thanks to the collaboration with other companies, they were able to
889 develop pilot scale pyrolysis plants in both USA and Thailand. Both projects demonstrated that
890 plastic pyrolysis at the large scale is feasible and sustainable, but more public engagement and
891 process engineering are required.

892 The industrial data shows that (a) polyolefins mixes or single materials are the most recurring;
893 (b) mixing is essential in large scale operations; (c) the explored temperature range is typically
894 350-600°C; and (d) the achieved oil yields vary between 72 and 81%. All these results agree
895 with the literature analyzed in this work.

896 The trajectory for the advancement of plastic pyrolysis technology necessitates a transition
897 from the generation of fuels to the production of chemicals. Direct synthesis of monomers,
898 requisite for the fabrication of new plastics through pyrolysis, presents economic challenges.
899 However, a circular economy framework can be realized through the production and
900 subsequent refinement of pyrolytic oil. This refined output can serve as a viable feedstock in
901 naphtha cracking processes, enabling the synthesis of new plastics from non-recyclable plastic
902 waste. It is imperative for researchers, pyrolysis enterprises, waste management entities,
903 recycling organizations, and the petrochemical sector to initiate and sustain dialogue and
904 collaboration. This collective effort is essential to establish a closed-loop system, thereby
905 facilitating the transition towards a more sustainable and circular approach in the management
906 and utilization of plastic waste.

907 **5. Conclusions**

908 This work represents a critical review of the literature appeared between 2011 and 2023 dealing
909 with the pyrolysis of plastic waste, with the aim of performing a comprehensive analysis by
910 considering at the same time the technical aspects (feedstocks, process parameters, reactors,
911 products, catalysts, kinetics, and pollutants), the economic feasibility of the scale-up, and the
912 environmental assessment through LCA. The results can be summarised as follows:

- 913 • Plastic waste pyrolysis has seen an exponential increase in interest in the last 12 years.
914 The articles published in the 2023 are 3.5 times the ones found in 2016 and 7 times the
915 number published in 2011.

- 916 • Virgin polymers and artificial mixtures of virgin plastics mimicking real mixed plastic
917 waste are the most explored. PE, PP, and PS are the polyolefins most studied since they
918 are the main components of post-consumer plastic waste. There is lack of experimental
919 data obtained with real plastic waste, containing fillers, additives, and contaminations.
- 920 • Most literature studies refer to lab scale (62%) and unmixed batch reactors (54%), with
921 an increase in interest in pilot scale experimentation in the last three years (52% of
922 research references).
- 923 • Mixing is fundamental to increase heat transfer and obtain constant and uniform
924 composition of products.
- 925 • Temperature is a critical parameter affecting yields and product composition. The most
926 used value is 500°C (46% of the references).
- 927 • Pressure affects degradation mechanisms, influencing oil production and composition.
928 It is crucial for oil quality and cost efficiency: better oil quality requires less post-
929 treatment which lower its production costs.
- 930 • Catalysts are mostly studied at the lab scale, and the most explored are zeolites, silica-
931 alumina and multi-function catalysts.
- 932 • Pyrolysis gas did not attract much research interest. Gases are considered as a by-
933 product that can be used for energy recovery as heating source for the process.
- 934 • Liquid products from pyrolysis are mostly researched for fuels applications. However,
935 aiming at a circular economy approach, this is not sustainable. The separation of 'low-
936 volume high-value' products, sch as waxes, need exploration.
- 937 • Plastic-derived chars have potential for value-added applications, like graphene and
938 CNT production.

- 939 • Limited LCA studies are available. The studies report lower environmental burdens for
940 plastic waste pyrolysis if compared to incineration and landfill disposal. The location
941 greatly influences the results.
- 942 • Feasibility of plastic waste pyrolysis scale-up is demonstrated by all the techno-
943 economic analysis reviewed.
- 944 • Industrial plants are starting to appear all over the world, but companies do not release
945 information about technologies, yields, and quality of products.

946 The knowledge gaps identified are:

- 947 • Real plastic waste needs to be studied to understand the effect of the contamination on
948 the process (catalytic or inhibitor effect), the products (enhanced or worsened), but
949 mainly to identify the processing challenges that need to be overcome to achieve full-
950 scale implementation.
- 951 • Sacrificial catalysts, possibly produced from residues and wastes, need to be
952 investigated because they could optimize the overall economic balance at the
953 commercial scale.
- 954 • Considering the growing interest in hydrogen technologies, future research on plastic
955 waste pyrolysis should be oriented towards processes able to maximize the content of
956 hydrogen in the gas products.
- 957 • Considering the liquid products, further research is needed to investigate their
958 perspectives as “non-fuel” chemical products.
- 959 • Life cycle assessments and process simulations need to be implemented at the
960 design/lab scale to define the best operating conditions and setup to minimize
961 environmental burdens, maximise yield and quality of products and, consequently, the
962 economic potential of commercial operations.

- 963 • Plastic pyrolysis products need to stop being considered as wastes. New legislations are
964 needed to categorize them as marketable products. This will allow to set quality
965 standards for pyrolysis products that will help the research community to identify
966 promising setups and operating conditions to tackle this problem of plastic waste
967 management.
- 968 • LCA and techno-economic assessment studies are necessary to support the
969 technological transfer and commercial implementation, and to understand the
970 environmental impacts of a full-scale pyrolysis plant fed by “real” plastic waste.

971 Plastics are not going to disappear from our lives due to their application in crucial sector such
972 as health, construction, automotive, electric and electronics to name a few. Currently, there is
973 a lack of functional sustainable alternatives. Plastic pyrolysis can be a crucial technology to
974 tackle the plastic waste problem due its adaptability and wide range of products that can be
975 integrated in existing systems. The research world needs to switch from lab scale
976 experimentation to larger scale so that technological challenges can be addressed. Feeding
977 problems due to non uniform size, melting and clogging of feeding lines, heat transfer troubles
978 due to both poor mixing and particle size, equipment erosion, corrosion, and wear are just a
979 few of the challenges encountered in larger plastic pyrolysis plants. When the
980 practical/mechanical tests are solved than pyrolysis can became an integrated part of the waste
981 management system handling non-recycled, heavily contaminated plastic waste. The future of
982 plastic pyrolysis lies in shifting focus from fuel to chemical production, requiring collaboration
983 across sectors to achieve a sustainable circular economy.

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990

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