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# Oil production from waste polyethylene and polystyrene co-pyrolysis: Interactions of temperature and carrier gas flow rate

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22 oil components and fractions. The results revealed that low temperature, high PS mass  
23 fraction, and low carrier gas flow rate were conducive to a light WPE/WPS co-pyrolysis oil  
24 production. The findings could guide the industrial process of waste plastic pyrolysis in  
25 different regions. Moreover, ANN-GA coupled with central-composite design can be used to  
26 regulate different target products under more complex conditions due to its good robustness.

27

28 **Keywords:** *Waste Plastic; Co-pyrolysis; Oil Production; Artificial neural network; Genetic*  
29 *algorithm.*

30

## 31 **1. Introduction**

32 The production of plastic has grown promptly from 1.5 million tons in 1950 to 368  
33 million tons in 2019 [1]. The European plastic production was up to 57.9 million tons in 2019  
34 [1], and almost half of the produced plastic became waste [2]. Moreover, nearly 25% of waste  
35 plastic winds up in landfills [3]. Consequently, waste plastic accumulates rapidly due to the  
36 low environmental degradability [4]. Plastic pollution destroys ecosystems [5] and causes  
37 harm to living creatures [6]. Pyrolysis is considered a promising alternative to landfill  
38 disposal of waste plastic, which can simultaneously produce liquid oil similar to commercial  
39 fuels (gasoline and diesel) [7-10]. Polyethylene (PE) and polystyrene (PS) are the first and  
40 third-most components in waste plastic due to their wide range of uses [11-12]. Therefore,  
41 many studies have investigated waste polyethylene (WPE) and polystyrene (WPS) pyrolysis  
42 for oil production. Park et al. [13] studied the pyrolysis of WPE at 653–736°C and recovered

43 15–38wt% oil in a tandem two-stage reactor. Al-Salem et al. [7,14] conducted the low-density  
44 PE pyrolysis experiments at 500–800°C. They found that the maximum wax yield was  
45 64.5wt% at 500°C [14], and the highest oil yield was 29wt% at 600°C [7]. The WPS pyrolysis  
46 oil has been reported to have a higher yield than the WPE pyrolysis oil. Nisar et al. [15]  
47 recovered 44.9–86.1wt% oil from the pyrolysis of WPS at 340–420°C. Quesada et al. [11]  
48 also obtained a high yield of 80wt% WPS thermal pyrolysis oil at 500°C in a fixed bed reactor.  
49 It can be concluded that temperature is a critical parameter determining pyrolysis oil  
50 production.

51 WPE and WPS in waste plastic have different contents in different regions [3]. Therefore,  
52 the effect of WPE/WPS mixture composition on oil production has also been studied by  
53 researchers. Quesada et al. [11] investigated the WPE/WPS mixture (25wt% PS) co-pyrolysis  
54 at 500°C and obtained ~70wt% of oil yield. Siddiqui and Redhwi [16] investigated the high-  
55 density-PE/PS (50–75wt% PS) co-pyrolysis at 440°C in a batch reactor. It was found that as  
56 the PS mass fraction increased, the oil yield decreased from 72.8wt% (under 50wt% PS) to  
57 25.6wt% (under 66.7wt% PS), while it increased to 56.1wt% when the PS mass fraction  
58 continuously increased to 75wt% PS. Williams and Williams [17] obtained 67.3wt% oil from  
59 PE/PS mixture (50wt% PS) co-pyrolysis at 700°C. While the predicted oil yield, calculated by  
60 the PE and PS sole pyrolysis oil yields, was 81.7wt%. They concluded that an interaction  
61 between PE and PS led to a lower-than-expected oil yield. Klaimy et al. found that the co-  
62 pyrolysis of plastic mixtures promoted the production of lightweight components (C6 and C7)  
63 in the oil [18]. Saad et al. [19] suggested that PE and PS could be fully decomposed before

64 500°C with a heating rate of 10°C/min. However, the plastic compositions affected the  
65 activation energies of mixed plastics. Moreover, co-pyrolysis of other types of plastics [20]  
66 and co-pyrolysis of plastics with biomass [21] also found the complex interactive effects of  
67 the mixture compositions.

68 On the other hand, the carrier gas flow rate is a significant parameter during the pyrolysis  
69 of plastic [22-23]. Muhammad et al. [23] investigated the effect of carrier gas flow rate on the  
70 pyrolysis of linear low-density PE. They found that increasing the flow rate from 0mL/min to  
71 1mL/min could dramatically enhance the oil yield from 54.0wt% to 61.5wt%. In contrast, the  
72 oil yield decreased to 45.0wt% under the flow rate of 60mL/min. Pan et al. [22] concluded  
73 that the WPE pyrolysis oil yield would decrease from 82.43wt% to 65.32wt% when the flow  
74 rate increased from 20mL/min to 100mL/min.

75 It can be concluded that the temperature and the carrier gas flow rate are two vital  
76 parameters during plastic pyrolysis. The WPE/WPS mixture composition also has a non-linear  
77 effect on the co-pyrolysis oil yield. The artificial neural network (ANN) is a promising  
78 method to formulate the dependent variable in terms of independent variables with complex  
79 interactions [24]. Neshat et al. [25-26] adopted ANN to predict wind speed and wind turbine  
80 power output. The findings revealed that ANN could accurately establish the expressions of  
81 the dependent variables. ANN can also be competent for detection works, e.g., plate's  
82 structural damage [27] and COVID-19 [28]. An optimization algorithm is required to  
83 determine the extrema of complex mathematical expressions established by the ANN. Genetic  
84 algorithm (GA), inspired by the evolution theory, is a widely used optimization algorithm [29].

85 Furthermore, GA has been proven to be qualified to determine the extrema of ANN-  
86 established expression [30].

87 The primary aim of this study is to investigate the interactions of operating conditions  
88 (temperature and flow rate of carrier gas) on the WPE/WPS (with different mixture  
89 compositions) co-pyrolysis oil yield. A hybrid model of the artificial neural network coupled  
90 with a genetic algorithm (ANN-GA) is adopted to quantitatively describe the complex  
91 interactions of operating conditions and WPE/WPS mixture composition on the product yields  
92 based on the experimental results. The oil samples are analyzed by gas chromatography/mass  
93 spectrometry (GC/MS) to determine the specific components. The ANN-GA is also adopted to  
94 predict and optimize the oil components and fractions under different operating conditions  
95 and WPE/WPS mixture compositions. The findings can guide the regional industrialization of  
96 waste plastic pyrolysis.

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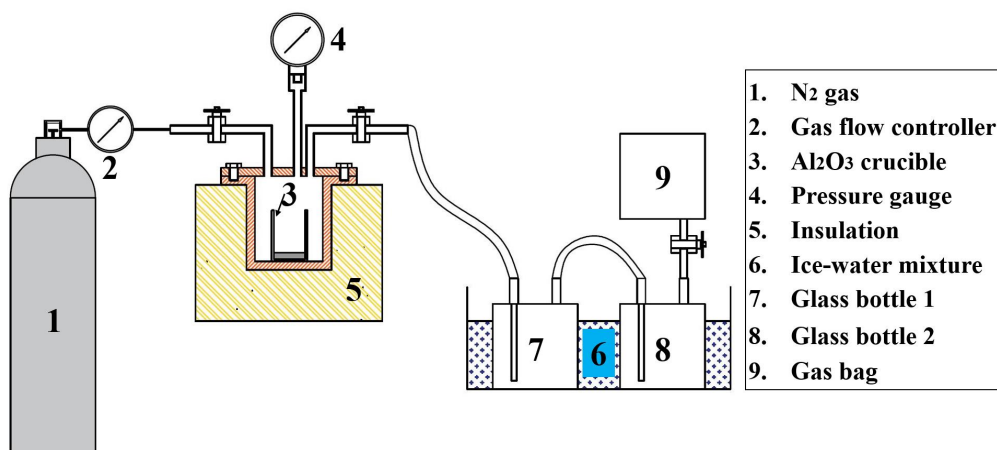
## 98 **2. Experiments and method**

### 99 *2.1. Pyrolysis experiments*

100 WPE and WPS were recycled from municipal solid waste (MSW) and provided by  
101 Zhoushan Jinke Renewable Resources Co., China. The waste plastics were first collected and  
102 sorted, then washed and dried, and finally made into pellets of about 3mm. The total mass of  
103 the WPE/WPS mixture used in each pyrolysis experiment was ~5g. As shown in Fig. 1, the  
104 WPE/WPS co-pyrolysis experiments were carried out in a 200mL semi-batch reactor. The  
105 WPE/WPS samples were mixed well and placed in the Al<sub>2</sub>O<sub>3</sub> crucible. The reactor was

106 purged with nitrogen (under 100mL/min) for 30min to ensure an inert atmosphere. The  
107 reactor was heated to the target temperature at 6°C/min and stayed at the target temperature  
108 for 20min. Two in-tandem glass bottles collected the WPE/WPS pyrolysis oil in a mixture of  
109 ice and water. A gas bag collected the pyrolysis gas. The char remained in the Al<sub>2</sub>O<sub>3</sub> crucible  
110 and was collected after cooling the reactor to 20°C.

111 The temperature, PS mass fraction, and flow rate of carrier gas were studied in the ranges  
112 of 425–525°C [19,22,31], 10–30wt% PS [3], and 0–60mL/min [23] based on the plastic  
113 pyrolysis characteristics, regional WPE/WPS composition, and reactor size. The WPE/WPS  
114 co-pyrolysis experiments were designed according to the central-composite design [32] and  
115 tabulated in Table A.1. 17 sets of experiments (R1–R17) were conducted to obtain the training  
116 set data of ANN-GA. 5 sets of experiments (R18–R22) were carried out to get the testing set  
117 data of ANN-GA. Moreover, the WPE/WPS co-pyrolysis oil samples were characterized by  
118 GC/MS to determine the specific composition of the oil.



119  
120 **Fig. 1.** The WPE/WPS co-pyrolysis experimental setup.

121  
122 *2.2. Method*

123 A hybrid artificial neural network model coupled with a genetic algorithm (ANN-GA)

124 was adopted to predict and optimize the WPE/WPS co-pyrolysis product yields, oil

125 components, and fractions. The simulation platform of the ANN-GA is Matlab R2016a. Fig. 2

126 demonstrates the flow chart of ANN-GA. The mathematical expressions of ANN-GA have

127 been detailed described in the previous study [30]. The arithmetic expressions between the

128 independent (temperature, PS mass fraction, and flow rate of carrier gas) and dependent

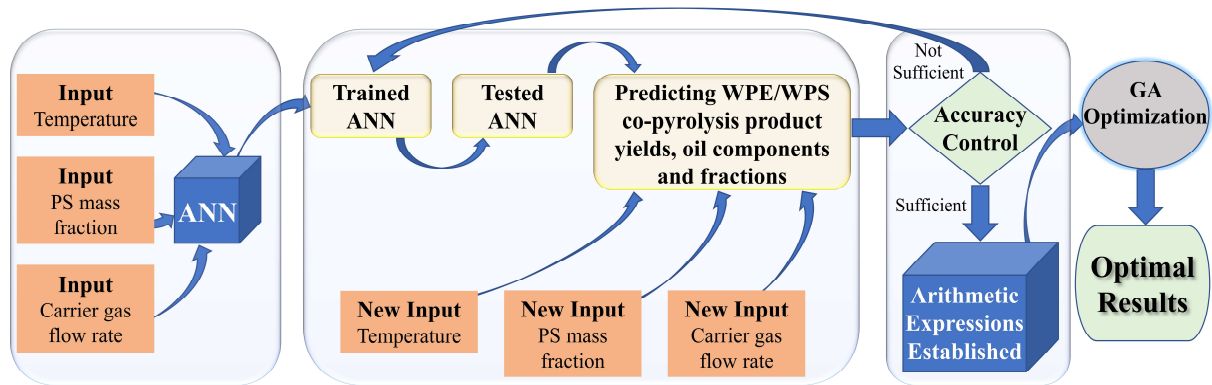
129 variables (co-pyrolysis product yields, oil components, and fractions) were established by

130 ANN based on the experimental results [33]. Subsequently, GA was used to optimize the

131 objective functions built by ANN. Consequently, the conditions (temperature, PS mass

132 fraction, and carrier gas flow rate) for the highest oil yield and the lightest oil production

133 could be determined by ANN-GA.



134

135 **Fig. 2.** The flow chart of ANN-GA.

136

137 **3. Results and discussion**

138 *3.1. Accuracy of ANN predicted co-pyrolysis product yields*

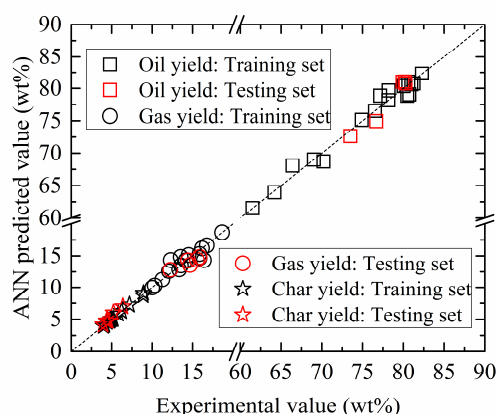
139 Fig. 3 shows the experimental and ANN predicted WPE/WPS co-pyrolysis oil, gas, and

140 char yields. It can be seen that the experimental oil yield varied from 61.51wt% (Sample R4)



141 to 82.28wt% (Sample R13), the experimental gas yield fluctuated from 10.01wt% (Sample R1)  
142 to 18.65wt% (Sample R17), and the experimental char yield oscillated from 3.93wt% (Sample  
143 R21) to 8.92wt% (Sample R5). It was reported that the WPE thermal pyrolysis oil and gas  
144 yields were 65.32–83.50wt% and 11.50–18.75wt% in the temperature range of 425–525°C  
145 [22]. The PS thermal pyrolysis oil yields were 80wt% under 450°C [34] and 71wt% under  
146 500°C [35]. Moreover, the WPS pyrolysis gas and char yields were ~16wt% and ~4wt% under  
147 500°C, respectively [11]. The WPE/WPS co-pyrolysis product yields obtained in the present  
148 work were within the ranges of individual WPE and WPS pyrolysis ones. Quesada et al. [11]  
149 studied the WPE/WPS co-pyrolysis under 500°C and 25wt% PS, obtaining ~70wt% of oil  
150 yield, ~22wt% of gas yield, and ~8wt% of char yield, values highly close to that obtained in  
151 the present work. Williams and Williams [17] obtained 64.0–67.3wt% oil yield and 1.2–  
152 2.2wt% char yield from PE/PS co-pyrolysis, which were also very close to the results in the  
153 present work.

154 On the other hand, Table 1 lists the root mean square error (RMSE), mean relative error  
155 (MRE), mean absolute error (MAE), standard deviation (SD), and coefficient of  
156 determination ( $R^2$ ) values between the experimental and ANN predicted co-pyrolysis product  
157 yields in the training and testing sets. The high  $R^2$  values and low RMSE, MRE, MAE, and  
158 SD values [36] exhibited the high accuracy of the ANN predicted WPE/WPS co-pyrolysis  
159 product yields.



160

161

**Fig. 3.** Experimental and ANN predicted co-pyrolysis product yields.

162

163

**Table 1.** Errors in the ANN predicted co-pyrolysis product yields.

	RMSE`	MRE	MAE	SD	R2
Training set	0.76%	-0.14%	2.06%	3.23%	0.9994
Testing set	0.87%	-1.60%	5.07%	4.03%	0.9993

164

165 *3.2. Interactions of temperature, PS mass fraction, and carrier gas flow rate on product*  
 166 *yields*

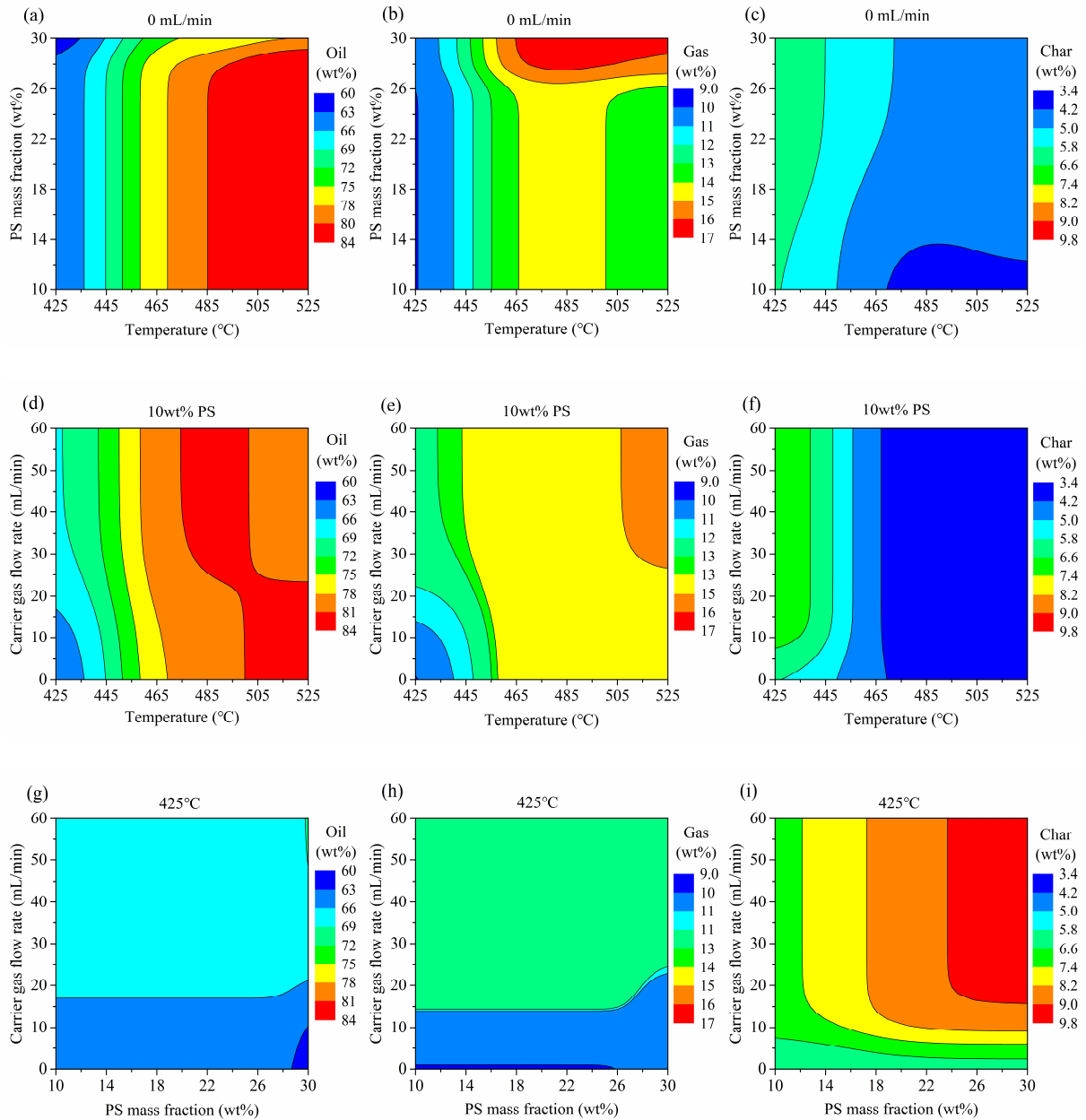
167 Fig. 4 demonstrates the interactions of temperature, PS mass fraction, and carrier gas flow  
 168 rate on the distributions of oil, gas, and char productions under 0mL/min, 10wt% PS, and  
 169 425°C. It is worth noting that due to the pressure difference generated during the pyrolysis of  
 170 plastics, oil and gas can flow out of the reactor spontaneously under 0mL/min. Moreover, Figs.  
 171 A.1–3 illustrate the ANN predicted oil, gas, and char yields under 30mL/min and 60mL/min,  
 172 20wt% PS and 30wt% PS, and 475°C and 525°C, respectively. It could be seen that the  
 173 temperature had a more significant impact on the oil yield than the PS mass fraction under all  
 174 flow rates of carrier gas (Fig. 4a, Fig. A.1a, and Fig. A.1d). Quesada et al. [11] also reported

175 that the WPE/WPS co-pyrolysis oil yield only hovered in a narrow range of ~70–73wt%  
176 within the range of 25–75wt% PS. As the temperature increased, the oil yields were  
177 significantly enhanced by 18.35wt% (under 10wt% PS) and 16.65wt% (under 30wt% PS)  
178 under the non-sweeping atmosphere (0mL/min, Fig. 4a). Higher temperatures could deepen  
179 the random scissions of WPE/WPS long chains [37] and the secondary cracking of char [38-  
180 39], thereby enhancing oil and gas yields and reducing char yield. Increasing the temperature  
181 further would cause the secondary reactions of oil cracking [15,40], and gas recondensation  
182 and repolymerization [17]. Therefore, when the temperature exceeded 480°C and continued to  
183 rise, the decrease in gas yield under 0mL/min (Fig. 4b) could be ascribed to the gas  
184 recondensation and repolymerization (gas consumption) were more intense than the oil  
185 cracking (gas generation). However, as the temperature increased from 488°C, the decrease in  
186 oil yield under 30mL/min (Fig. A.1a) and 60mL/min (Fig. A.1d) might be attributed to the oil  
187 cracking (oil consumption) was more violent than the gas recondensation and  
188 repolymerization (oil formation) [14].

189 Improving the flow rate of carrier gas under low temperatures would enhance the oil and  
190 gas yields, regardless of the variation of PS mass fraction. For instance, the oil and gas yields  
191 were enhanced by 4.76wt% and 2.90wt% under 425°C and 10wt% PS (Fig. 4d). However,  
192 increasing the carrier gas flow rate decreased the oil yield by ~2wt% under the highest  
193 temperature (525°C). The reduction of the oil yield might be ascribed to the shorter residence  
194 time of volatile products in the reaction zone under higher flow rates of carrier gas, thereby  
195 inhibiting the Diels-Alder reactions of gaseous olefin for oil formation [41]. Simultaneously,

196 the gas yield was enhanced by 1.80wt% (13.50wt%–15.30wt%) as the flow rate of carrier gas  
197 increased under 525°C (Fig. 4e). On the other hand, the carrier gas flow rate's increase led to a  
198 decrease in char yield under high temperatures. As the flow rate of carrier gas increased, the  
199 char yields reduced by 0.09wt%, 0.15wt%, and 0.17wt% under 10wt% PS (Fig. 4f), 20wt%  
200 PS (Fig. A.2c), and 30wt% PS (Fig. A.2f), respectively. The decrease in char yield could be  
201 ascribed to the inhabitation of secondary reactions of the co-pyrolysis volatiles for char  
202 generation [42]. However, enhancing the carrier gas flow rate increased the char yield under  
203 lower temperatures. The flow rate change would affect the heat transfer efficiency in the  
204 reaction zone [43], which might enhance the char yield.

205 Enhancing the PS mass fraction in WPE/WPS led to a decrease in oil yield and an  
206 increase in gas and char yields under the non-sweeping atmosphere (0mL/min). For instance,  
207 the enhancement of PS mass fraction led to a reduction of 2.46wt% in oil yield (Fig. 4g), an  
208 enhancement of 0.35wt% in gas yield (Fig. 4h), and an enhancement of 0.40wt% in char yield  
209 (Fig. 4i) under 425°C and 0mL/min. Similar trends were reported by Quesada et al. [11]: a  
210 decrease of ~3.5wt% in oil yield, an increase of ~1.5wt% in gas yield, and an increase of  
211 ~2wt% in char yield were observed when the PS mass fraction increased from 50wt% to  
212 75wt% in WPE/WPS. The increase in char yield might be ascribed to the abundant aromatics  
213 presented in the PS pyrolysis oil, which could further generate char [15].



214

215

216

217 **Fig. 4.** Interactive effects of temperature, PS mass fraction, and carrier gas flow rate on the distributions of

218 WPE/WPS co-pyrolysis products: (a) Oil yield under 0mL/min; (b) Gas yield under 0mL/min; (c) Char

219 yield under 0mL/min; (d) Oil yield under 10wt% PS; (e) Gas yield under 10wt% PS; (f) Char yield under

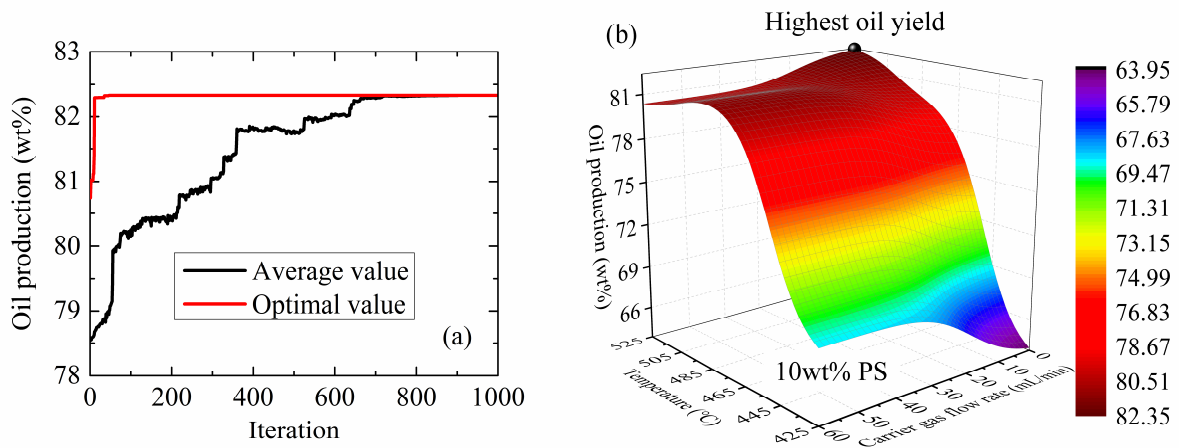
220 10wt% PS; (g) Oil yield under 425°C; (h) Gas yield under 425°C; (i) Char yield under 425°C.

221

222 **3.3. Optimization of oil yield by ANN-GA**

223 Fig. 5a shows the optimal and average values of the ANN-GA objective function (oil  
 224 yield). The ANN-GA predicted the highest oil yield was 82.33wt% under 525°C, 10wt% PS,  
 225 and 0mL/min (Fig. 5b). Meanwhile, the experimental oil yield under the ANN-GA optimized  
 226 conditions was 82.28wt%. The relative error between the experimental and ANN-GA  
 227 predicted oil yields was merely 0.06%, revealing the ANN-GA optimized value's high  
 228 accuracy.

229 It is also noteworthy that the highest oil yield under 60mL/min was obtained at 485°C.  
 230 The optimal temperature of 485°C was close to the optimal WPE pyrolysis temperatures  
 231 reported by Quesada et al. [31] (500°C) and Pan et al. [22] (488°C). Carrier gas could  
 232 accelerate the purging of the pyrolysis volatiles and carry heavier volatile molecules out of the  
 233 reactor. The heavy pyrolysis oil components remained in the reactor under the non-sweeping  
 234 atmosphere (0mL/min). Higher temperatures were required for the thermal cracking of oil's  
 235 heavy components into shorter-chain hydrocarbons, which then could volatilize and  
 236 spontaneously leave the reactor [44]. Therefore, WPE/WPS co-pyrolysis oil yield optimized  
 237 temperature exceeded 500°C, generally considered the optimal temperature for the pyrolysis  
 238 of waste plastics with carrier gas [45].



239

240 **Fig. 5.** Optimization of oil yield by ANN-GA: (a) Representation of optimization process; (b) Highest oil  
241 yield.

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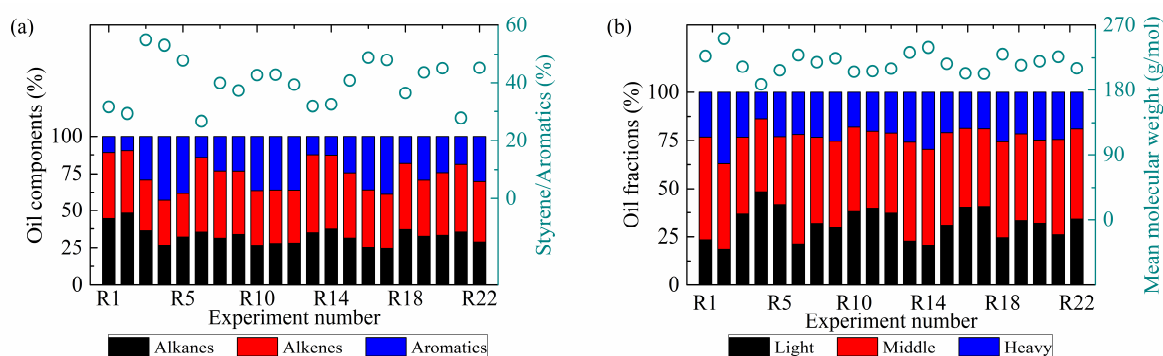
### 243 3.4. GC/MS analysis

244 The components of WPE/WPS co-pyrolysis oil samples (R1–R22) were analyzed by  
245 GC/MS. Moreover, the WPE/WPS co-pyrolysis oils were divided into light (C7–C11), middle  
246 (C12–C20), and heavy fractions (>C20) based on the carbon numbers [22]. Fig. 6a indicates  
247 that the oils were composed of alkanes (24.55–48.25%), alkenes (29.38–52.57%), and  
248 aromatics (9.68–42.89%) [11]. Meanwhile, styrene accounted for the highest proportion of  
249 aromatics in oils, which varied from 26.73% to 54.87%. High styrene proportion might be  
250 ascribed to aromatic hydrocarbons that take up the main component of WPS thermal pyrolysis  
251 oil, of which styrene occupies 33–40% [46].

252 Fig. 6b shows that the light, middle, and heavy oil fractions fluctuated from 18.49–  
253 48.49%, 35.32–56.78%, and 13.64–37.05%, respectively. The WPE/WPS co-pyrolysis oils  
254 had more light and middle fractions than WPE pyrolysis oils [22]. Moreover, the mean  
255 molecular weight of the WPE/WPS co-pyrolysis oil oscillated from 187.73g/mol to  
256 251.01g/mol, which was between the mean molecular weights of individual PS (176g/mol)  
257 and PE (306g/mol) thermal pyrolysis oils [17].

258 The operating conditions of temperature and carrier gas flow rate had complex  
259 interactions on the WPE/WPS co-pyrolysis oil components and fractions [47]. Moreover, the  
260 styrene/aromatics ratio and the mean molecular weight of WPE/WPS co-pyrolysis oil did not

261 linearly change as the PS mass fraction increased [16]. The non-linear relationship indicated  
 262 that the reaction species of WPE and WPS had undetermined interactions on the distributions  
 263 of co-pyrolysis oil components and fractions [17]. Therefore, ANN-GA was adopted to  
 264 investigate the interactions of temperature, PS mass fraction, and carrier gas flow rate on the  
 265 oil components and fractions; and determine the optimal conditions for each component and  
 266 fraction.



267  
 268 **Fig. 6.** WPE/WPS co-pyrolysis oil components and fractions: (a) Distributions of alkanes, alkenes, and  
 269 aromatics in oils, and the styrene/aromatics ratio variations; (b) Distributions of light, middle, and heavy  
 270 fractions in oils, and the mean molecular weight variations.

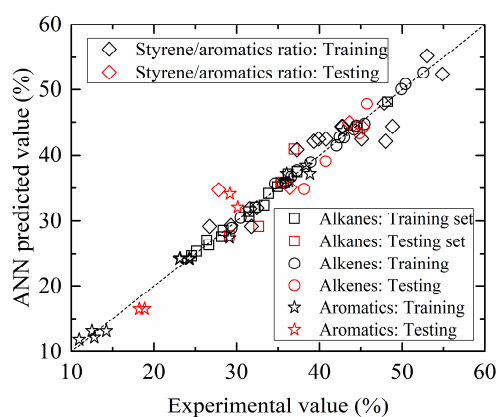
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### 272 3.5. WPE/WPS co-pyrolysis oil components

#### 273 3.5.1. Interactions of temperature, PS mass fraction, and carrier gas flow rate

274 Fig. 7 demonstrates the experimental and ANN predicted oil components in the training  
 275 and testing sets. Besides, Table 2 tabulates the RMSE, MRE, MAE, SD, and R2 values  
 276 between the experimental and ANN predicted oil components. The high R2 values and low  
 277 RMSE, MRE, MAE, and SD values verified the accuracy of predicted results.





278

279

**Fig. 7.** Experimental and ANN predicted oil components.

280

281

**Table 2.** Errors in the ANN predicted oil components.

	RMSE`	MRE	MAE	SD	R2
Training set	1.44%	-0.16%	2.60%	3.38%	0.9798
Testing set	2.96%	-0.53%	7.97%	3.39%	0.8762

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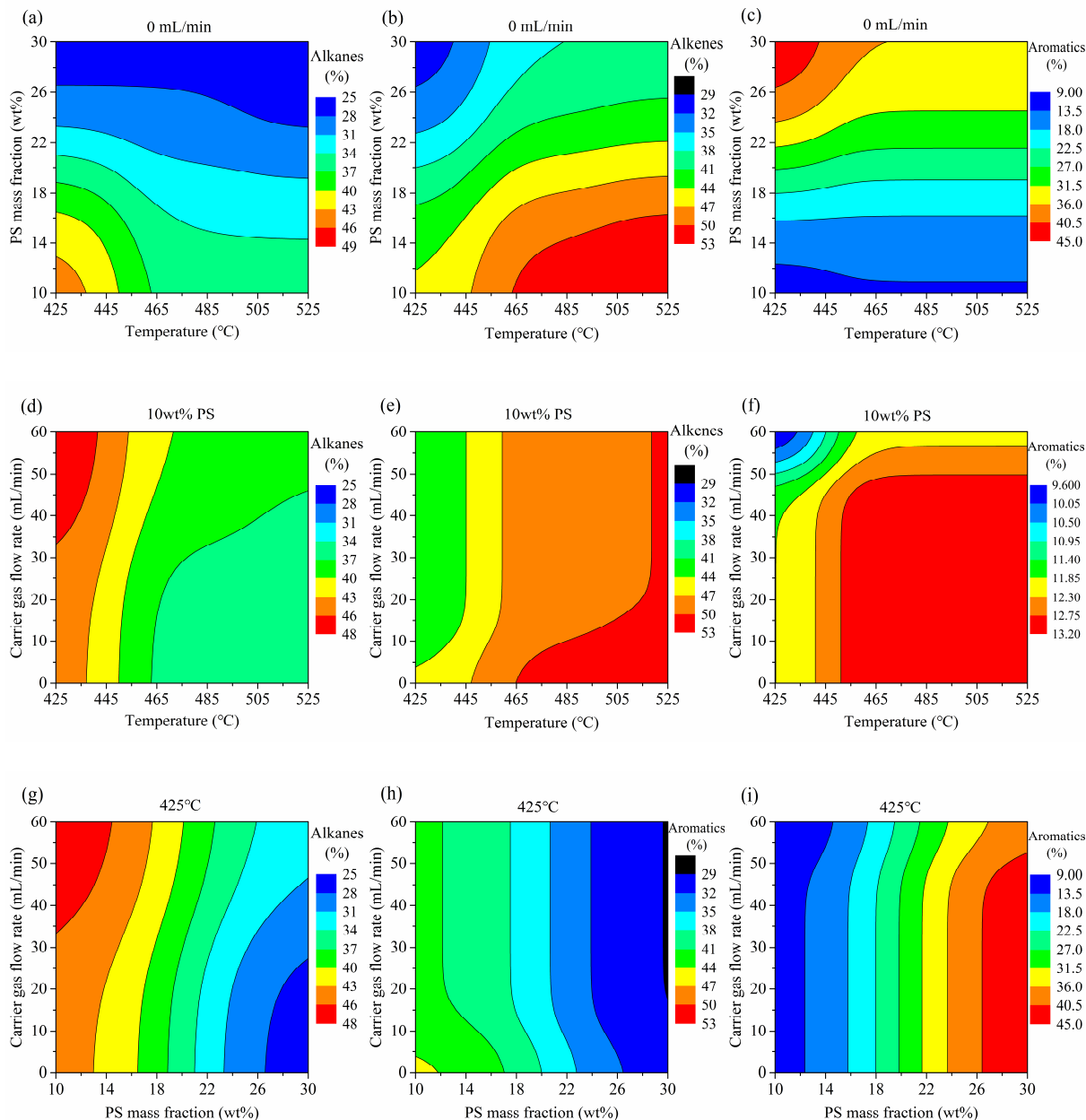
292

Fig. 8 shows the interactions of temperature, PS mass fraction, and carrier gas flow rate on the distributions of oil components under 0mL/min, 10wt% PS, and 425°C. The interactions of triple conditions under 30mL/min, 20wt% PS, and 475°C (Fig. A.5); and 60mL/min, 30wt% PS, and 525°C (Fig. A.7) are similar to the conditions under 0mL/min, 10wt% PS, and 425°C. It could be observed that the temperature had a more significant influence on the alkanes proportion under lower PS mass fractions (Fig. 8a). As the temperature increased under 10wt% PS, the alkanes proportion reduced by 9.10% (44.31%–35.21%, Fig. 8a), while the alkanes proportion enhanced by 8.12% (44.45%–52.57%, Fig. 8b). Increasing temperature contributed to a decrease of 7.87% (43.73%–35.86%) in aromatics proportion under 30wt% PS (Fig. 8c). Aromatics were mainly generated from the thermal

293 decomposition of PS in WPE/WPS. The polycyclic PS could be decomposed under a lower  
294 temperature than PE [48], thereby a high aromatics proportion of 43.73% was obtained under  
295 425°C, 30wt% PS, and 0mL/min (Fig. 8c). Enhancing the temperature aggravates the  
296 secondary cracking reactions of alkanes and aromatics for the generation of alkenes, leading  
297 to a decrease in alkanes and aromatics proportions and an increase in alkenes proportion [49].

298 Figs. 8d–f demonstrate the interactions of carrier gas flow rate and temperature on the  
299 proportions of alkanes, alkenes, and aromatics under 10wt% PS, respectively. It could be seen  
300 that improving the flow rate of carrier gas would enhance the alkanes proportion and reduce  
301 the alkenes and aromatics proportions under all temperatures. For instance, as the flow rate of  
302 carrier gas increased under 525°C and 10wt% PS, the alkanes proportion increased by 2.25%  
303 (35.21%–37.46%), while the alkenes and aromatics proportions decreased by 2.51%  
304 (52.57%–50.06%) and 0.94% (13.11%–12.17%), respectively. Higher carrier gas flow rates  
305 could purge the co-pyrolysis volatiles out of the reaction zone faster [50], thereby suppressing  
306 the secondary cracking of alkanes for alkenes formation and the Diels-Alder reactions of  
307 alkenes for aromatics formation [41].

308 On the other hand, as the PS mass fraction increased, the alkanes and alkenes proportions  
309 reduced by ~16–18% (Fig. 8g) and ~12.6–14% (Fig. 8h), while the aromatics proportion  
310 significantly enhanced by ~29–32% (Fig. 8i) under 425°C. The principal components of PS  
311 pyrolysis oil were aromatics [51], thereby enhancing the PS mass fraction led to an increase in  
312 aromatics proportion and a decrease in alkanes and alkenes proportions.



313

314

315

316 **Fig. 8.** Interactive effects of temperature, PS mass fraction, and carrier gas flow rate on the distributions of

317 WPE/WPS co-pyrolysis oil components: (a) Alkanes proportion under 0mL/min; (b) Alkenes proportion

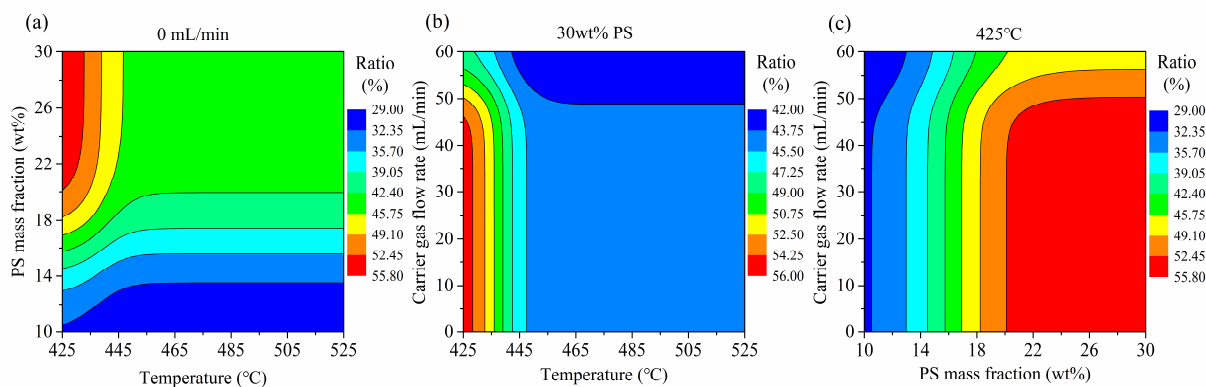
318 under 0mL/min; (c) Aromatics proportion under 0mL/min; (d) Alkanes proportion under 10wt% PS; (e)

319 Alkenes proportion under 10wt% PS; (f) Aromatics proportion under 10wt% PS; (g) Alkanes proportion

320 under 425°C; (h) Alkenes proportion under 425°C; (i) Aromatics proportion under 425°C.

321

322 Styrene was the dominant component in the WPE/WPS co-pyrolysis oil's aromatics  
323 proportion, which could be ascribed to the primary product of styrene in the PS pyrolysis oil  
324 [16,46]. Fig. 9 depicts the interactions of temperature, PS mass fraction, and carrier gas flow  
325 rate on the styrene/aromatics ratio under 0mL/min, 30wt% PS, and 425°C. Moreover, Fig. A.6  
326 and Fig. A.8 demonstrate the interactions of triple conditions under 30mL/min, 20wt% PS,  
327 and 475°C; and 60mL/min, 10wt% PS, and 525°C, respectively. The enhancement of PS mass  
328 fraction contributed to the increases of ~26%, ~26%, and ~19% in styrene/aromatics ratio  
329 under 0mL/min (Fig. 9a), 30mL/min (Fig. A.6a), and 60mL/min (Fig. A.8a), respectively. The  
330 temperature had a more significant impact on the styrene/aromatics ratio under 20wt% (Fig.  
331 A.6b) and 30wt% (Fig. 9b) PS mass fractions compared to the one under 10wt% PS mass  
332 fraction (Fig. A.8b). Besides, the increase in temperature under 20wt% and 30wt% PS mass  
333 fractions resulted in a reduction of styrene/aromatics ratio, which might be ascribed to the  
334 secondary cracking of styrene under higher temperatures [52]. On the other hand, improving  
335 the flow rate of carrier gas led to a decrease in styrene/aromatics ratio under 425°C (Fig. 9c),  
336 regardless of the variation of PS mass fraction. Higher carrier gas flow rates would shorten  
337 the volatiles residence time in the reaction zone, thereby resulting in the incomplete pyrolysis  
338 of PS for styrene formation.



339

340

**Fig. 9.** Interactive effects of temperature, PS mass fraction, and carrier gas flow rate on the

341

styrene/aromatics ratio: (a) Under 0mL/min; (b) Under 30wt% PS; (c) Under 425°C.

342

### 343 3.5.2. ANN-GA optimization

344

Fig. 10a shows that the highest alkanes proportion was 48.09% under 425°C, 10wt% PS,

345

and 60mL/min, which indicated that low temperature and high carrier gas flow rate could

346

suppress the secondary cracking of alkanes in oil [53-56]. On the other hand, the highest

347

alkenes proportion was 52.57% under 525°C, 10wt% PS, and the non-sweeping atmosphere

348

(0mL/min). High temperature and low carrier gas flow rate resulted in the over-cracking of

349

the oil, which might be conducive to the formation of alkene.

350

The highest aromatics proportion of 43.73% was obtained under 425°C, 30wt% PS, and

351

0mL/min. Increasing temperature can affect the aromatics proportion in three aspects: i)

352

promoting the alkanes' dehydro-aromatization [57] for aromatics formation, ii) facilitating the

353

alkenes' Diels-Alder reactions [41] for aromatics formation, and iii) exacerbating the over-

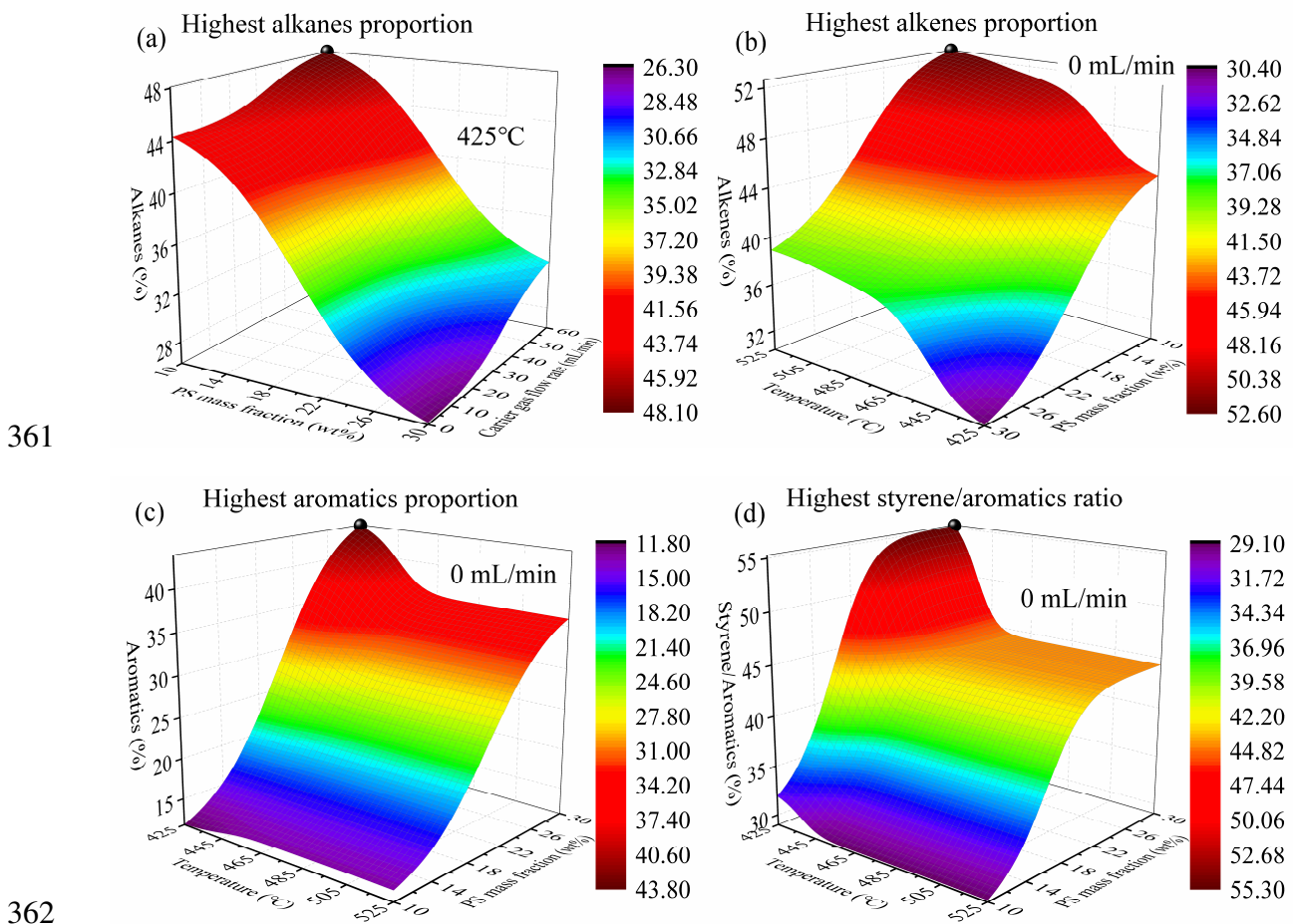
354

cracking for aromatics consumption [49]. The highest aromatics proportion achieved under

355

low temperature indicated that the inhibition of aromatics over-cracking dominated the

356 aromatics formation. High PS mass fraction in the WPE/WPS mixture could enhance the  
 357 proportion of the aromatics; thereby, the highest aromatics proportion was gained under  
 358 30wt% PS. Meanwhile, the highest styrene/aromatics ratio was 55.22% under the same  
 359 conditions (425°C, 30wt% PS, and 0mL/min). This was because styrene was the dominant  
 360 component in the PS pyrolysis oil.



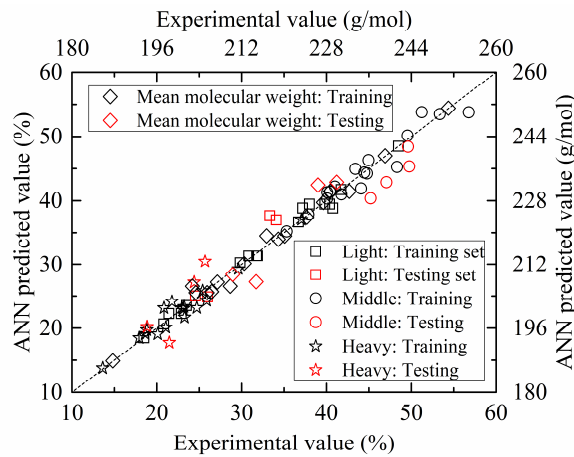
363 **Fig. 10.** ANN-GA optimized proportions of the oil components and styrene/aromatics ratio: (a) Highest  
 364 alkanes proportion; (b) Highest alkenes proportion; (c) Highest aromatics proportion; (d) Highest  
 365 styrene/aromatics ratio.

366

367 *3.6. WPE/WPS co-pyrolysis oil fractions*

368 3.6.1. Interactions of temperature, PS mass fraction, and carrier gas flow rate

369 Fig. 11 shows the experimental and ANN predicted oil fractions in the training and testing  
 370 sets. Moreover, Table 3 lists the RMSE, MRE, MAE, SD, and R2 values between the  
 371 experimental and ANN predicted oil fractions. The high R2 values and low RMSE, MRE,  
 372 MAE, and SD values exhibited high accuracy of the predicted results.



373  
 374 **Fig. 11.** Experimental and ANN predicted oil fractions.

375  
 376 **Table 3.** Errors in the ANN predicted oil fractions.

	RMSE`	MRE	MAE	SD	R2
Training set	1.38%	-0.09%	2.00%	2.91%	0.9995
Testing set	3.38%	-1.77%	6.88%	3.22%	0.9970

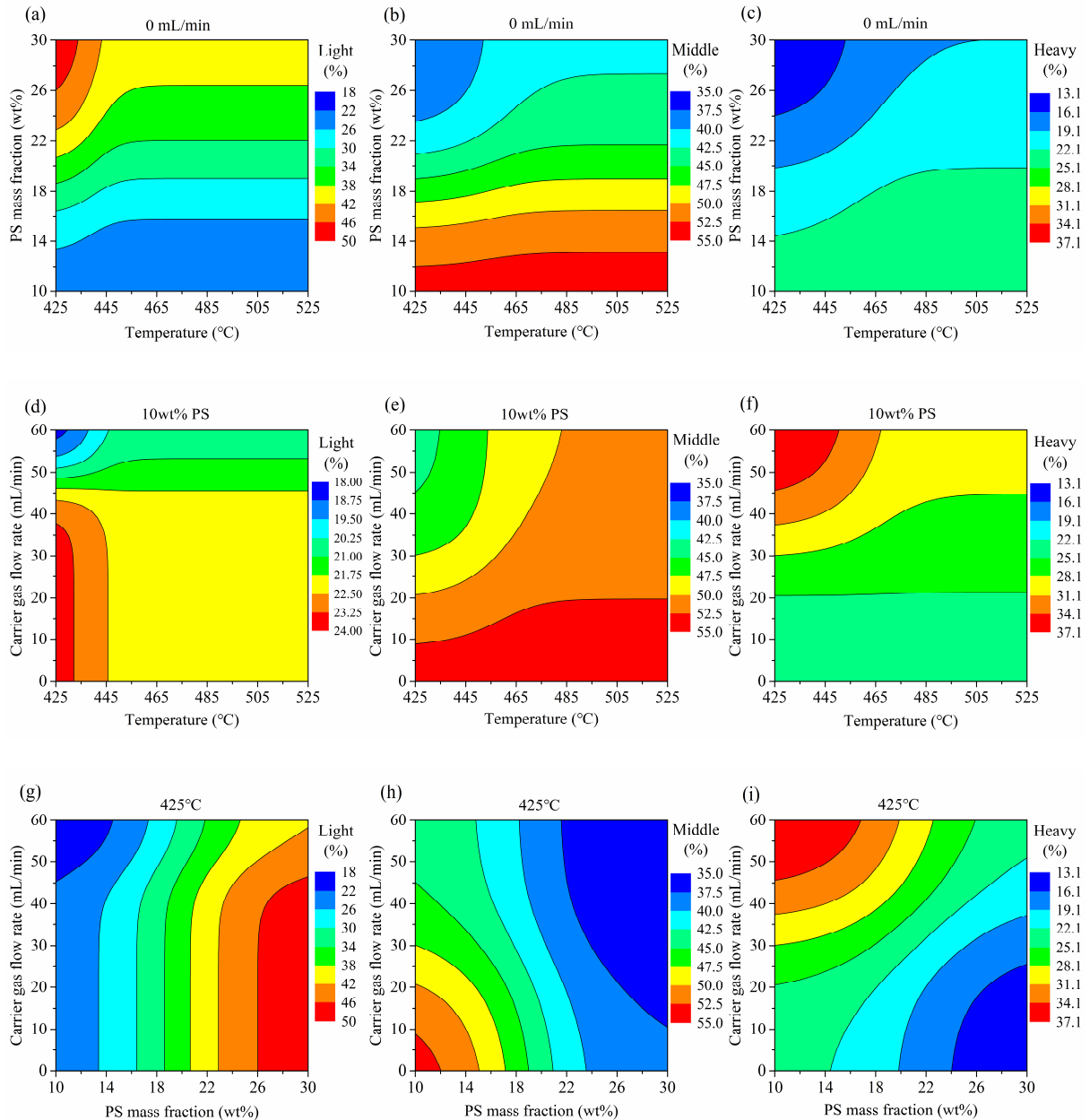
377  
 378 Fig. 12 depicts the interactions of temperature, PS mass fraction, and carrier gas flow rate  
 379 on the distributions of oil fractions under 0mL/min, 10wt% PS, and 425°C. The interactions of  
 380 triple conditions under 30mL/min, 20wt% PS, and 475°C (Fig. A.11); and 60mL/min, 30wt%  
 381 PS, and 525°C (Fig. A.13) are similar to the conditions under 0mL/min, 10wt% PS, and 425°C.  
 382 Figs. 12a–c demonstrate that the temperature significantly impacted the oil fractions under

383 higher PS mass fractions. As the temperature increased under 10wt% PS, the light fraction  
384 decreased by 1.24%, and the middle and heavy fractions increased by 0.31% and 1.30%,  
385 respectively. However, the light fraction significantly reduced by 9.03%, and the middle and  
386 heavy fractions enhanced by 4.32% and 5.35% when the temperature increased under 30wt%  
387 PS. Higher temperatures could aggravate the secondary cracking of light fraction in oil,  
388 thereby reducing the light fraction and enhancing the middle and heavy fractions [58].

389 It is noteworthy that the light and middle fractions would decrease, and the heavy fraction  
390 would increase as the flow rate of carrier gas increased, regardless of the variations of  
391 temperature and PS mass fraction (Figs. 12d–f). For instance, the carrier gas flow rate's  
392 increase reduced the light (by 5.02%) and middle (by 9.10%) fractions, and enhanced the  
393 heavy fraction (by 14.05%) under 425°C and 10wt% PS, respectively. It might be ascribed to  
394 the suppression of gas recondensation and repolymerization for generating the light and  
395 middle fractions in oil under higher carrier gas flow rates.

396 On the other hand, enhancing the PS mass fraction could considerably increase the light  
397 fraction and decrease the middle and heavy fractions in oil. For example, as the PS mass  
398 fraction enhanced under 425°C and non-sweeping atmosphere (0mL/min), the light fraction  
399 increased by 24.99% (23.51%–48.50%), whereas the middle and heavy fractions decreased by  
400 15.62% (53.48%–37.86%) and 9.23% (22.99%–13.76%), respectively. It might be ascribed to  
401 the high proportion of styrene in the PS pyrolysis oil [11], which belonged to the light fraction.  
402 Consequently, a higher PS mass fraction could contribute to an increase in the light fraction.





403

404

405

406 **Fig. 12.** Interactive effects of temperature, PS mass fraction, and carrier gas flow rate on the distributions of

407 oil fractions: (a) Light fraction under 0mL/min; (b) Middle fraction under 0mL/min; (c) Heavy fraction

408 under 0mL/min; (d) Light fraction under 10wt% PS; (e) Middle fraction under 10wt% PS; (f) Heavy

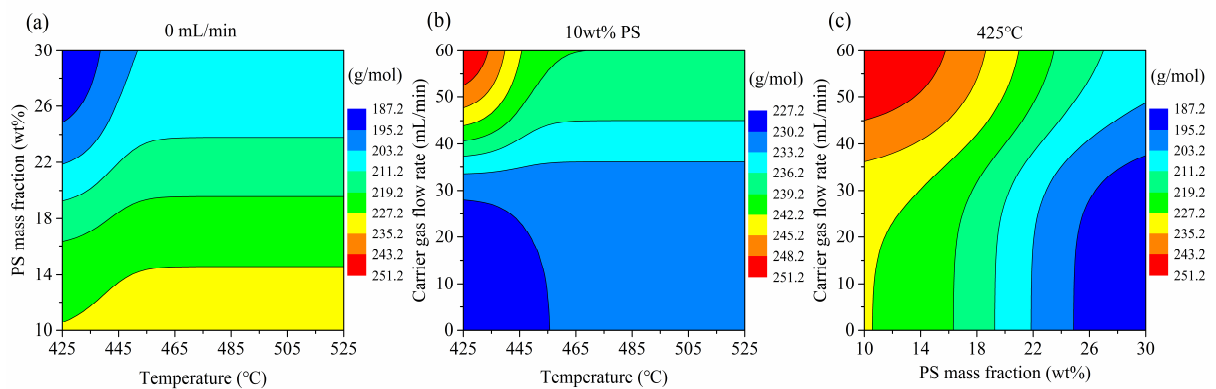
409 fraction under 10wt% PS; (g) Light fraction under 425°C; (h) Middle fraction under 425°C; (i) Heavy

410

fraction under 425°C.

411

412 Fig. 13 depicts the interactions of temperature, PS mass fraction, and carrier gas flow rate  
 413 on the oil's mean molecular weight under 0mL/min, 10wt% PS, and 425°C. Moreover, Fig.  
 414 A.12 and Fig. A.14 demonstrate the interactions of triple conditions under 30mL/min, 20wt%  
 415 PS, and 475°C; and 60mL/min, 30wt% PS, and 525°C, respectively. Fig. 13a shows that the  
 416 oil's mean molecular weight increased with the increasing temperature under all PS mass  
 417 fractions. Moreover, the temperature significantly impacted the oil's mean molecular weight  
 418 under higher PS mass fractions. For instance, as the temperature increased under 0mL/min,  
 419 the oil's mean molecular weight was enhanced by 2.77g/mol and 16.98g/mol under 10wt% PS  
 420 and 30wt% PS, respectively. The enhancement might be due to the more intense over-  
 421 cracking reaction of the oil's light fraction [59], producing a heavier oil with a higher mean  
 422 molecular weight. A higher carrier gas flow rate increased the oil's mean molecular weight  
 423 under all temperatures (Fig. 13b). The increase was because the light and middle fractions  
 424 decreased, whereas the heavy fraction increased with the increasing flow rate of carrier gas  
 425 (Figs. 12d–f). Besides, the increase in PS mass fraction contributed to a significant decrease  
 426 of ~40g/mol in the oil's mean molecular weight under 425°C (Fig. 13c).



427

428 **Fig. 13.** Interactive effects of temperature, PS mass fraction, and carrier gas flow rate on the oil's mean

429 molecular weight: (a) Under 0mL/min; (b) Under 10wt% PS; (c) Under 425°C.

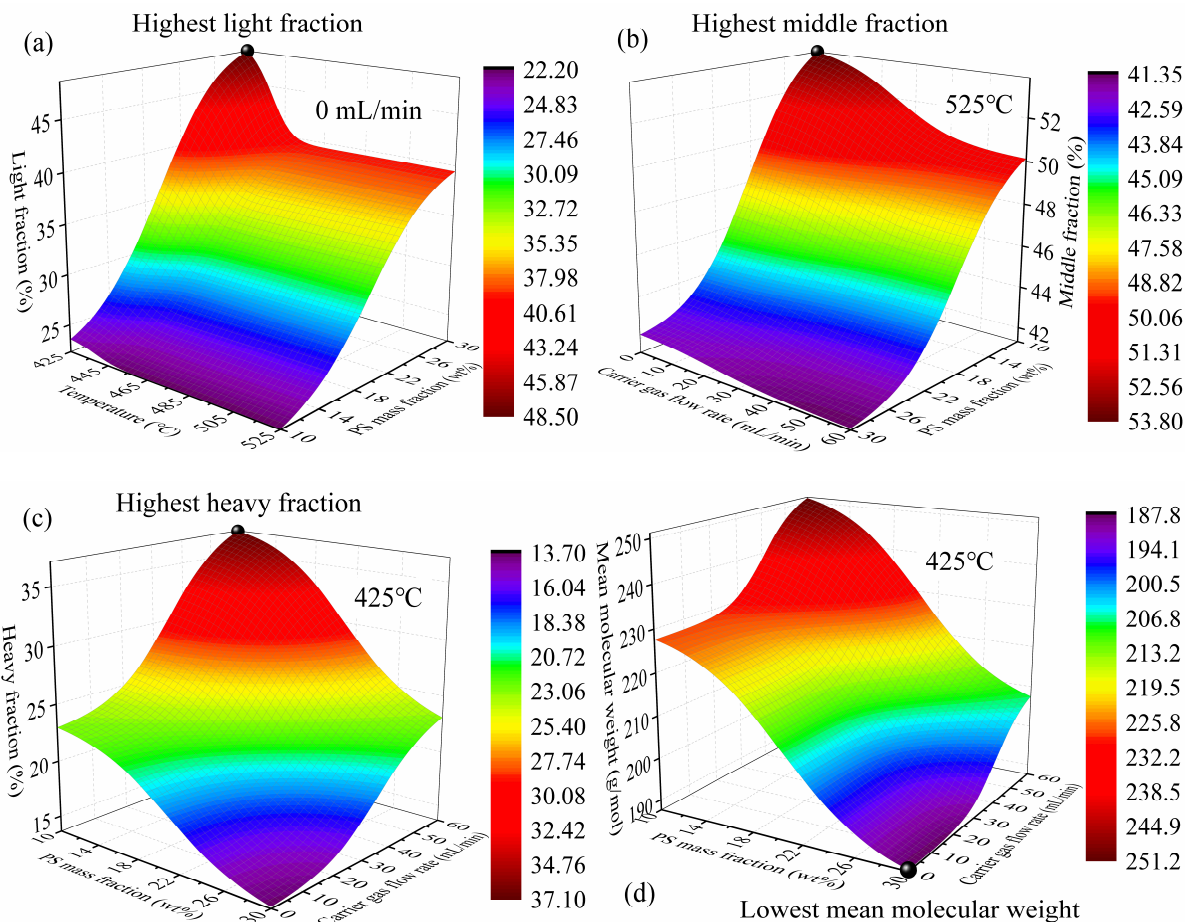
430

### 431 3.6.2. ANN-GA optimization

432 As discussed in Section 3.6.1, the temperature, PS mass fraction, and carrier gas flow rate  
433 had complex interactions on the distributions of WPE/WPS co-pyrolysis oil fractions and the  
434 oil's mean molecular weight. Therefore, the ANN-GA was adopted to determine the highest  
435 oil fractions and the lightest co-pyrolysis oil with the lowest mean molecular weight.

436 Fig. 14a shows that the highest light fraction was 48.50% under 425°C, 30wt% PS, and  
437 0mL/min. Low temperature and non-sweeping atmosphere could suppress the secondary  
438 cracking of light fraction and promote gas recondensation for light fraction formation. The  
439 highest middle fraction of 53.79% was obtained under 525°C, 10wt% PS, and 0mL/min (Fig.  
440 14b). Higher temperatures might be conducive to the secondary cracking of heavy fraction for  
441 middle fraction generation. On the other hand, the highest heavy fraction was 37.04% under  
442 425°C, 10wt% PS, and 60mL/min (Fig. 14c).

443 The lightest WPE/WPS co-pyrolysis oil with the lowest mean molecular weight of  
444 187.85g/mol was obtained under 425°C, 30wt% PS, and 0mL/min (Fig. 14d). It could be  
445 concluded that low temperature, high PS mass fraction, and non-sweeping atmosphere were  
446 conducive to the light WPE/WPS co-pyrolysis oil production.



447

448

449 **Fig. 14.** ANN-GA optimized oil fractions and mean molecular weight: (a) Highest light fraction; (b)

450 Highest middle fraction; (c) Highest heavy fraction; (d) Lowest mean molecular weight.

451

#### 452 4. Conclusions

453 This study investigated the co-pyrolysis of WPE/WPS for oil production under different  
 454 temperatures and carrier gas flow rates. The ratio of the real-life WPE and WPS is different in  
 455 different regions. Therefore, the effect of WPE/WPS mixture composition on oil production  
 456 was also studied. It was found that the WPE/WPS mixture composition and operating  
 457 conditions had complex interactions on the co-pyrolysis oil yield. Therefore, a hybrid model  
 458 of ANN-GA was adopted to optimize the conditions for oil production based on the central-

459 composite design. It is noteworthy that the optimized conditions in this study have limitations  
460 because the experiments were performed in a bench-scale reactor. Nevertheless, the findings  
461 could provide qualitative guidance for the regional industrialization of waste plastic pyrolysis.  
462 ANN-GA can also be used to regulate different target products under more complicated  
463 conditions due to its good robustness. This study's principal conclusions are outlined as  
464 follows.

- 465 ● The highest oil yield was 82.33wt% under 525°C, 10wt% PS, and 0mL/min. The findings  
466 revealed that high temperature, low PS mass fraction, and low carrier gas flow rate were  
467 conducive to a higher oil yield.
- 468 ● The highest alkanes, alkenes, and aromatics proportions were 48.09%, 52.57%, and  
469 43.73%, respectively. Additionally, the highest styrene/aromatics ratio was 55.22% under  
470 the same conditions as the highest aromatics proportion (425°C, 30wt% PS, and 0mL/min).
- 471 ● The lightest WPE/WPS co-pyrolysis oil with the lowest mean molecular weight of  
472 187.85g/mol was obtained under 425°C, 30wt% PS, and 0mL/min. It could be concluded  
473 that low temperature, high PS mass fraction, and low carrier gas flow rate were conducive  
474 to the light WPE/WPS co-pyrolysis oil production.

475

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#### 478 **Competing Interest**

479 The authors declare no competing financial interest.

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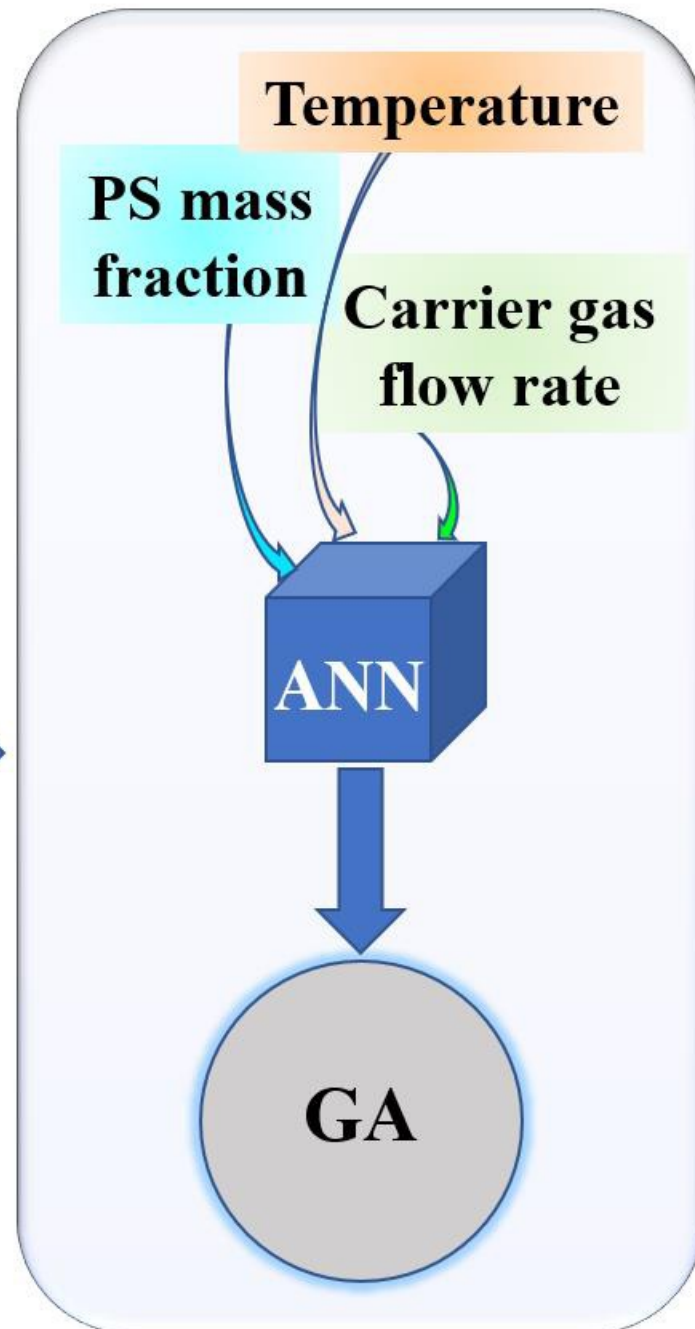
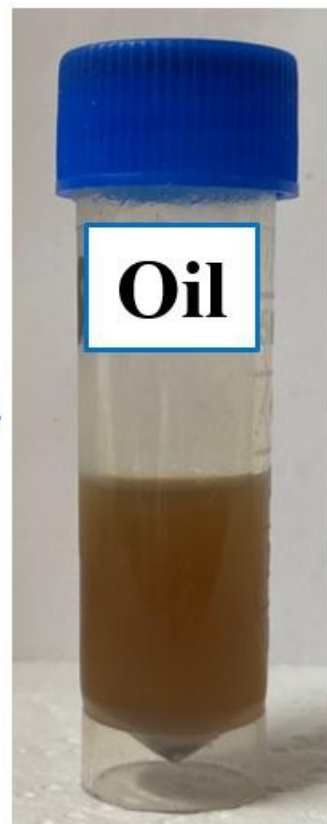
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**Waste plastics recovered from MSW**

**Pyrolysis**



**WPE/WPS co-pyrolysis for oil production**

