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1	Oil production from waste polypropylene and polystyrene co-
2	pyrolysis: Interactions of temperature and carrier gas flow rate
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10	Abstract: This study investigated the co-pyrolysis of waste polyethylene (WPE) and
11	polystyrene (WPS) for oil production under different operating conditions (temperature and
12	carrier gas flow rate). The ratio of the real-life WPE and WPS is different in different regions.
13	Therefore, the interactions of operating conditions were investigated under different
14	WPE/WPS mixture compositions. A hybrid model of artificial neural network-genetic
15	algorithm (ANN-GA) was adopted to predict and optimize the co-pyrolysis oil yield due to
16	the complex interactions of the WPE/WPS mixture composition and operating conditions.
17	Consequently, the highest oil yield was 82.33wt% under 525°C, 10wt% PS, and a non-
18	sweeping atmosphere (0mL/min). The results indicated that high temperature, low PS mass
19	fraction, and low carrier gas flow rate led to a higher oil yield. The WPE/WPS co-pyrolysis
20	oils were composed of alkanes, alkenes, and aromatics. Meanwhile, styrene accounted for the
21	highest proportion of aromatics in oils. ANN-GA was also adopted to predict and optimize the

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 million tons in 2019 [1]. The European plastic production was up to 57.9 million tons in 2019 33 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 low environmental degradability [4]. Plastic pollution destroys ecosystems [5] and causes 36 harm to living creatures [6]. Pyrolysis is considered a promising alternative to landfill 37 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 third-most components in waste plastic due to their wide range of uses [11-12]. Therefore, 40 many studies have investigated waste polyethylene (WPE) and polystyrene (WPS) pyrolysis 41 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 PE pyrolysis experiments at 500-800°C. They found that the maximum wax yield was 44 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. It can be concluded that temperature is a critical parameter determining pyrolysis oil 49 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 continuously increased to 75wt% PS. Williams and Williams [17] obtained 67.3wt% oil from 58 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 copyrolysis of plastic mixtures promoted the production of lightweight components (C6 and C7) 62 63 in the oil [18]. Saad et al. [19] suggested that PE and PS could be fully decomposed before 500°C with a heating rate of 10°C/min. However, the plastic compositions affected the
activation energies of mixed plastics. Moreover, co-pyrolysis of other types of plastics [20]
and co-pyrolysis of plastics with biomass [21] also found the complex interactive effects of
the mixture compositions.

On the other hand, the carrier gas flow rate is a significant parameter during the pyrolysis of plastic [22-23]. Muhammad et al. [23] investigated the effect of carrier gas flow rate on the pyrolysis of linear low-density PE. They found that increasing the flow rate from 0mL/min to 1mL/min could dramatically enhance the oil yield from 54.0wt% to 61.5wt%. In contrast, the oil yield decreased to 45.0wt% under the flow rate of 60mL/min. Pan et al. [22] concluded that the WPE pyrolysis oil yield would decrease from 82.43wt% to 65.32wt% when the flow 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 parameters during plastic pyrolysis. The WPE/WPS mixture composition also has a non-linear 76 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 structural damage [27] and COVID-19 [28]. An optimization algorithm is required to 82 determine the extrema of complex mathematical expressions established by the ANN. Genetic 83 84 algorithm (GA), inspired by the evolution theory, is a widely used optimization algorithm [29]. Furthermore, GA has been proven to be qualified to determine the extrema of ANN-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

WPE and WPS were recycled from municipal solid waste (MSW) and provided by Zhoushan Jinke Renewable Resources Co., China. The waste plastics were first collected and sorted, then washed and dried, and finally made into pellets of about 3mm. The total mass of the WPE/WPS mixture used in each pyrolysis experiment was ~5g. As shown in Fig. 1, the WPE/WPS co-pyrolysis experiments were carried out in a 200mL semi-batch reactor. The WPE/WPS samples were mixed well and placed in the Al₂O₃ 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 Al₂O₃ 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.



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123 A hybrid artificial neural network model coupled with a genetic algorithm (ANN-GA) was adopted to predict and optimize the WPE/WPS co-pyrolysis product yields, oil 124 125 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 yield, ~22wt% of gas yield, and ~8wt% of char yield, values highly close to that obtained in 150 151 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.

On the other hand, Table 1 lists the root mean square error (RMSE), mean relative error (MRE), mean absolute error (MAE), standard deviation (SD), and coefficient of determination (R2) values between the experimental and ANN predicted co-pyrolysis product yields in the training and testing sets. The high R2 values and low RMSE, MRE, MAE, and SD values [36] exhibited the high accuracy of the ANN predicted WPE/WPS co-pyrolysis product 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 425°C. It is worth noting that due to the pressure difference generated during the pyrolysis of 169 170 plastics, oil and gas can flow out of the reactor spontaneously under 0mL/min. Moreover, Figs. A.1-3 illustrate the ANN predicted oil, gas, and char yields under 30mL/min and 60mL/min, 171 172 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 flow rates of carrier gas (Fig. 4a, Fig. A.1a, and Fig. A.1d). Quesada et al. [11] also reported 174

175 that the WPE/WPS co-pyrolysis oil yield only hovered in a narrow range of ~70-73wt% within the range of 25-75wt% PS. As the temperature increased, the oil yields were 176 177 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 repolymerization (oil formation) [14]. 188

Improving the flow rate of carrier gas under low temperatures would enhance the oil and gas yields, regardless of the variation of PS mass fraction. For instance, the oil and gas yields were enhanced by 4.76wt% and 2.90wt% under 425°C and 10wt% PS (Fig. 4d). However, increasing the carrier gas flow rate decreased the oil yield by ~2wt% under the highest temperature (525°C). The reduction of the oil yield might be ascribed to the shorter residence time of volatile products in the reaction zone under higher flow rates of carrier gas, thereby 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 reaction zone [43], which might enhance the char yield. 204

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].



Fig. 4. Interactive effects of temperature, PS mass fraction, and carrier gas flow rate on the distributions of
WPE/WPS co-pyrolysis products: (a) Oil yield under 0mL/min; (b) Gas yield under 0mL/min; (c) Char
yield under 0mL/min; (d) Oil yield under 10wt% PS; (e) Gas yield under 10wt% PS; (f) Char yield under
10wt% PS; (g) Oil yield under 425°C; (h) Gas yield under 425°C; (i) Char yield under 425°C.



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



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

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

The operating conditions of temperature and carrier gas flow rate had complex interactions on the WPE/WPS co-pyrolysis oil components and fractions [47]. Moreover, the 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.



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

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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

Fig. 7 demonstrates the experimental and ANN predicted oil components in the training

and testing sets. Besides, Table 2 tabulates the RMSE, MRE, MAE, SD, and R2 values

- 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.





Fig. 7. Experimental and ANN predicted oil components.

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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

282

283 Fig. 8 shows the interactions of temperature, PS mass fraction, and carrier gas flow rate 284 on the distributions of oil components under 0mL/min, 10wt% PS, and 425°C. The 285 interactions of triple conditions under 30mL/min, 20wt% PS, and 475°C (Fig. A.5); and 286 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 287 288 influence on the alkanes proportion under lower PS mass fractions (Fig. 8a). As the 289 temperature increased under 10wt% PS, the alkanes proportion reduced by 9.10% (44.31%-290 35.21%, Fig. 8a), while the alkanes proportion enhanced by 8.12% (44.45%–52.57%, Fig. 8b). 291 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 292

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



Fig. 8. Interactive effects of temperature, PS mass fraction, and carrier gas flow rate on the distributions of
WPE/WPS co-pyrolysis oil components: (a) Alkanes proportion under 0mL/min; (b) Alkenes proportion
under 0mL/min; (c) Aromatics proportion under 0mL/min; (d) Alkanes proportion under 10wt% PS; (e)
Alkenes proportion under 10wt% PS; (f) Aromatics proportion under 10wt% PS; (g) Alkanes proportion
under 425°C; (h) Alkenes proportion under 425°C; (i) Aromatics proportion under 425°C.

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.



340

Fig. 9. Interactive effects of temperature, PS mass fraction, and carrier gas flow rate on the styrene/aromatics ratio: (a) Under 0mL/min; (b) Under 30wt% PS; (c) Under 425°C.

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343 3.5.2. ANN-GA optimization

Fig. 10a shows that the highest alkanes proportion was 48.09% under 425°C, 10wt% PS, and 60mL/min, which indicated that low temperature and high carrier gas flow rate could suppress the secondary cracking of alkanes in oil [53-56]. On the other hand, the highest alkenes proportion was 52.57% under 525°C, 10wt% PS, and the non-sweeping atmosphere (0mL/min). High temperature and low carrier gas flow rate resulted in the over-cracking of the oil, which might be conducive to the formation of alkene.

The highest aromatics proportion of 43.73% was obtained under 425°C, 30wt% PS, and 0mL/min. Increasing temperature can affect the aromatics proportion in three aspects: i) promoting the alkanes' dehydro-aromatization [57] for aromatics formation, ii) facilitating the alkenes' Diels-Alder reactions [41] for aromatics formation, and iii) exacerbating the overcracking for aromatics consumption [49]. The highest aromatics proportion achieved under low temperature indicated that the inhibition of aromatics over-cracking dominated the aromatics formation. High PS mass fraction in the WPE/WPS mixture could enhance the proportion of the aromatics; thereby, the highest aromatics proportion was gained under 30wt% PS. Meanwhile, the highest styrene/aromatics ratio was 55.22% under the same conditions (425°C, 30wt% PS, and 0mL/min). This was because styrene was the dominant component in the PS pyrolysis oil.





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.



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Fig. 11. Experimental and ANN predicted oil fractions.

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



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

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

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



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.

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 OmL/min, 419 the oil's mean molecular weight was enhanced by 2.77g/mol and 16.98g/mol under 10wt% PS and 30wt% PS, respectively. The enhancement might be due to the more intense over-420 421 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 (Figs. 12d-f). Besides, the increase in PS mass fraction contributed to a significant decrease 425 426 of ~40g/mol in the oil's mean molecular weight under 425°C (Fig. 13c).





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

430

431 3.6.2. ANN-GA optimization

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

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

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



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

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

451

452 **4.** Conclusions

This study investigated the co-pyrolysis of WPE/WPS for oil production under different temperatures and carrier gas flow rates. The ratio of the real-life WPE and WPS is different in different regions. Therefore, the effect of WPE/WPS mixture composition on oil production was also studied. It was found that the WPE/WPS mixture composition and operating conditions had complex interactions on the co-pyrolysis oil yield. Therefore, a hybrid model of ANN-GA was adopted to optimize the conditions for oil production based on the central459 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.

The highest oil yield was 82.33wt% under 525°C, 10wt% PS, and 0mL/min. The findings
revealed that high temperature, low PS mass fraction, and low carrier gas flow rate were
conducive to a higher oil yield.

The highest alkanes, alkenes, and aromatics proportions were 48.09%, 52.57%, and 43.73%, respectively. Additionally, the highest styrene/aromatics ratio was 55.22% under the same conditions as the highest aromatics proportion (425°C, 30wt% PS, and 0mL/min).
The lightest WPE/WPS co-pyrolysis oil with the lowest mean molecular weight of 187.85g/mol was obtained under 425°C, 30wt% PS, and 0mL/min. It could be concluded 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.

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478 **Competing Interest**

479 The authors declare no competing financial interest.

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