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Influences of temperature, waste size and residence time on the generation of polycyclic aromatic hydrocarbons during the fast pyrolysis of medical waste

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ABSTRACT

Mismanagement of Medical wastes can lead to human health and environmental risks. Currently, new pyrolysis technologies are being used to treat medical waste that can reduce the amount of landfilled waste, make it safe, and eventually convert it to a hydrocarbon fuel. Polycyclic aromatic hydrocarbons (PAHs) are pyrolysis by-products and major environmental pollutants. In this study, hazardous medical wastes were pyrolyzed using a semi-industrial pilot scale fast pyrolysis reactor with the purpose of improving the quality of the char for its recovery or use as fuel. The generation of total 4-6 rings PAHs was studied in char product from hazardous medical waste fast pyrolysis under different pyrolysis conditions variables including a vast temperature range (300-700°C), different residence times (100-190 s) and various waste particle sizes (1-3 cm). GC analyzer coupled with a FID detector was used to detect and measure the PAH compounds in char residues. The results demonstrated that the PAHs are present in significant concentrations in char product (54-1184 mg kg^{-1}). Generation of total 4-6 rings PAHs varied by temperature, residence time and waste size. Significant interaction was observed between residence time and temperature that influenced the PAHs generation. By optimizing the pyrolysis operating conditions it is possible to minimize the amount of PAHs generation in the char.

Keywords: Fast pyrolysis, Polycyclic aromatic hydrocarbons (PAHs), Char, Medical waste.

INTRODUCTION

Health care can generate variety of wastes. The major sources of medical wastes (MW) are hospitals and other health care activities such as laboratories or dialysis (Stanković *et al.* 2008). Around 75-90% of MW categorized non-hazardous waste, whereas the remaining 10-25% are hazardous (Ali *et al*. 2017). Inappropriate management of these wastes can increase environmental and health risks. Health care activities in developing countries suffer from inappropriate waste management which can lead to health and environmental risks (Delmonico *et al.* 2018; Alavi *et al.* 2018). The handling and processing of healthcare wastes are further exacerbated in developing countries by several factors, including lack of technological and economical capacities, social problems, and inadequate training of responsible staff (Mmereki *et al.* 2017).

Currently, several process equipment, including autoclaves, hydroclaves, pyrolysis reactors and incineration chambers are being used to suppress the medical waste pollutions and hazards (Chartier 2014). Based on several studies in the United States, 49-60% of medical wastes are incinerated, 20-37% are autoclaved, and 4-5% are treated by other technologies, (Windfeld & Brooks 2015). The use of thermal solutions such as incineration and

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high temperature pyrolysis inevitably generates toxic substances such as dioxin and furans, mercury and PAHs (Pacyna *et al.* 2006; Verma 2014). In the recent years, much more attention is given to the promising pyrolysis technology. Currently, pyrolysis technologies are being used to treat medical wastes that can reduce the amount of landfilled waste, make it safe to dispose, and eventually convert most of it to fuel such as char and pyrolytic oil. The process is divided into three types depending on the operating conditions exploited: slow, fast, and flash pyrolysis. Fast pyrolysis occurs at a moderate temperature range, short residence time of the vapor and high heating rate. This technology further is characterized by relatively low investment costs and high energy efficiencies as compared to other processes (Guedes *et al.* 2018).

Application of pyrolysis in treating various kinds of wastes has been widely discussed in literature as among which there are studies over municipal sludge (Zhou *et al*. 2015; Chang *et al.* 2016), waste tires (Martínez *et al.* 2014) and biomass wastes (Tripathi *et al.* 2016; Arabyarmohammadi *et al.* 2018). However, few studies have specifically focused on the effects of pyrolysis on medical waste considering its remarkable diversity which may affect the pyrolytic activity (Deng *et al.* 2008). Despite the lack of proper evidences in hand, pyrolysis may lead to thermal cracking, with high temperature and long residence time functioning as precursors towards the formation of long-lasting toxic organics, such as polycyclic aromatic hydrocarbons. There are still argues if these species could be considered as molecular markers of incomplete combustion processes (Cass 1998; Ravindra *et al.* 2008). Given the mutagenic, teratogenic and carcinogenic activity of some of these PAHs, they have been classified as priority pollutants for humans (Kim *et al.* 2013) The toxicity of PAHs is backed by their molecular structure, which contain nitro, chlorinated and oxy groups (Lundstedt *et al.* 2007). 4-6 rings PAHs, containing four or more benzene rings and more potent carcinogens, teratogens or mutagens than PAHs with lower numbers of aromatic rings, can cause higher risks to human health and environmental ecology. 4-6 rings-PAHs are highly stable, high potential for accumulation in the environment, lipophilic and hydrophobic (Shi *et al.* 2018). The available literature conjointly confirm the presence of PAHs in the products of pyrolysis plants. However, none of the authors has considered the mixed matrix of medical waste. Studies carried out mostly on incineration chambers, contain reports of the formation of PAHs during waste burning and the impact of operating conditions on PAH derivatives (Horii *et al.* 2008; Ravindra *et al.* 2008).Various factors including the particle size, temperature and time of pyrolysis as well as moisture content inluence the pyrolysis efficiency and the generation of unwanted by-products, namely PAHs. Zielinska & Oleszczuk studied the effect of pyrolysis operating temperatures (500-700°C) on PAHs concentrations in sewage sludge-derived biochar's, reporting that the production rate of these chemical compounds would increase at elevated temperatures(Zielińska & Oleszczuk 2016). The production of PAHs in pyrolysis is also dependent on the reactor residence time, feedstock properties, particle size, moisture and temperature. Recently, it has been reported that process conditions such as temperature and residence time could affect the formation of PAHs from the pyrolysis of cellulose and low molecular weight (LMW) PAHs first formed and passed through a maximum at lower temperature than the high molecular weight PAHs (McGrath *et al.* 2003). Wolfarm Buss (2016) examined the role of feedstock types (wood and straw, as the most common feedstocks) on total concentrations of USEPA- recognized PAHs in resulting biochars at a typical pyrolysis process and noted the impact of particle size on PAHs formation (Buss *et al.* 2016). Most recently, increasing the particle diameter is reported to be strongly correlated with the PAHs formation at high pyrolysis temperature ranges (Stark & Ghoniem 2017). These studies have all been carried out on a laboratory or pilot scale reactors.

Therefore, in this study we applied the fast pyrolysis process as a solution for medical waste treatment. We have studied the influence of different operating factors of fast pyrolysis process (particle size, temperature, and resident time) on the concentration and generation of total 4-6 rings of USEPA PAHs, i.e. fluoranthene, benzo(a)anthracene, chrysene, pyrene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, dibenz(a,h)anthracene, benzo(g,h,i)perylene, and indeno(1,2,3-cd)pyrene in the char product from the hazardous medical waste.

MATERIALS AND METHODS

Waste sampling and characterization

Hazardous medical waste (HMW) samples for this study were collected from Mofid children's hospital in Tehran (the capital city of Iran). The hospital is under the auspices of Shahid Beheshti University of Medical Sciences with a capacity of 261 beds. The mean rate of infectious and pharmaceutical-chemical waste production in this

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hospital are 315 kg day^{-1} and 10 kg day^{-1}, respectively. Waste samples were collected from a random mixture of 120 kg infectious and 40 kg pharmaceutical-chemical wastes. The collected samples were first crashed by an industrial grade crasher into smaller pieces and then sieved into three particle sizes of 1, 2 and 3 cm using a mesh screen. The sieved waste composition (Wt.%) included plastic (39.3); paper (11.15); textile (23.8); glass / sharp particles (22.7); about 3.05% (Wt.%) was also recorded as unrecognizable medical waste. The mean moisture content of the waste was 30 (Wt.%).

Experimental set-up

Fast pyrolysis experiments were conducted in a semi-industrial pilot scale apparatus. The fast pyrolysis setup was consisted of the following main components.

A stainless-steel (Grade ASTM A-316) pipe was employed for preparation of the main chamber. The reaction chamber was equipped with a screw of the same material as a solid propellant. Total length of the reaction tube was 150 cm, of which 110 cm is heated by electrical heater bands. A screw hopper feeder, shown as weight loss feeder in Fig. 1. Total length of this section was 40 cm. Split tube conductive electrical furnace installed around the main reaction pipe which covers the last 110 cm of reactor length. A collection chamber at the bottom outlet of the reactor, was used to collect solid char produced during the feedstock pyrolysis. A water-cooled condenser was employed for condensing the exhaust gases which exit from the top outlet of reactor. Also a chamber used for collecting the pyrolytic-oil condensate and helicoid screw driving occurred using an electric motor. The main design dimensions of the pyrolysis reactor are presented in Table 1.

Table 1. Dimensions of the pyrolysis reactor and screw.

Item	Dimensions (cm)
Heated Tube Length (L)	110
D (Tube Inner Diameter)	10
d_{out} (Screw Diameter)	7.5
d_{in} (Shaft Diameter)	3
p_c (Screw Pitch)	5

The furnace temperature varied in the range of 300-700 °C. The residence time inside the reactor, which is controlled by the rotation rate of the screw, varied a range between 90 to 600 seconds. The maximum feeding rate to the reactor was 5 kg h^{-1}. The pyrolysis process was performed in absence of oxygen through continuous flush of nitrogen gas (N_2). As for 1 kg of feed to the reactor, 150 L nitrogen was injected to meet this goal. Since any sudden temperature change in the top outlet of the reactor could lead to the unwanted phenomenon of condensation inside the outlet tubes (especially in presence of heavy hydrocarbons in the gas stream) the top outlet tubes were also coiled by an electrical heater and the tube external temperature was maintained at 350 °C. Through experiments with different feedstock compositions and in various runtimes in our study, the corresponding temperature was found optimum as it avoids the condensation of heavy organic molecules in the outlet. The schematic diagram of pyrolysis depicted in Fig. 1.

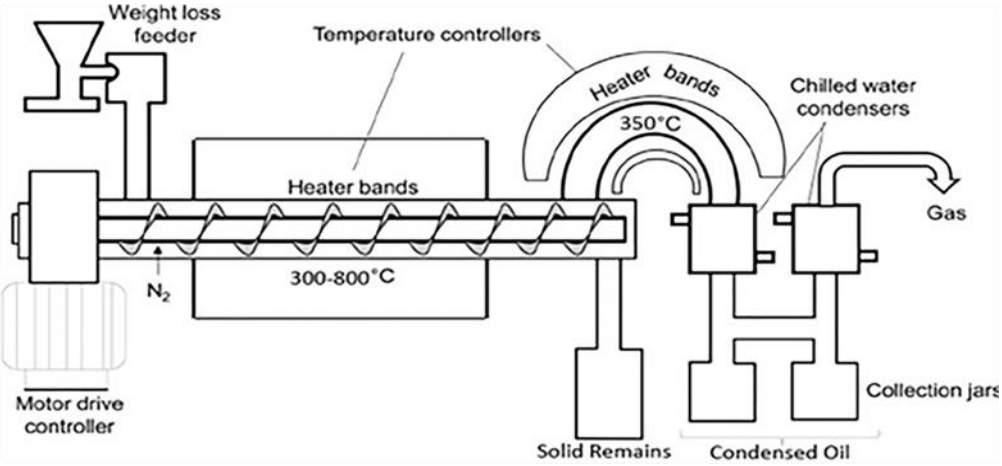

Fig. 1. The schematic diagram of fast pyrolysis reactor.

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Experimental design and data analysis

Like any typical pyrolysis experiment, prior to the commencement, a nearly homogeneous volume of mixed medical waste (feedstock) was prepared. The Design Expert Software (version 7.0) was employed for the statistical design of experiments and data analysis. RSM has been demonstrated to be a powerful statistical tool for obtaining optimum conditions for complex processes and evaluating the interactions of mutually influencing parameters with a limited number of experiments. The relative significance of several affecting factors even in the presence of complex interactions could indeed be assessed through employing this technique. Experiments were initiated as a preliminary study for determining the most influencing parameters before designing the experimental runs. Accordingly, three factors including temperature, residence time, and feedstock particle size were selected as key effective independent design variables, designated as (X_t), (X_r), and (X_s), respectively; whereas, the generation of total 4-6 rings PAHs was considered as the response of the system (dependent variable, Y). Table 2 depicts the variables and their levels established by CCD. Statistical calculations were carried out with coded values according to the following relationship:

$$Z_i = \frac{X_i - X_0}{\Delta x}$$

where Z_i is the coded (dimensionless) value of the variable, X_i represents the actual value of parameter, X_0 stands for the real value of independent variable in center point, and Δx refers to the difference between the high and the median values of the variable. Accordingly, the studied levels were designated as -2, -1, 0, +1, and +2 as are given in these tables.

Table 2. Independent variables and their levels for the CCD experiment.

Independent variables	Limits and levels				
	-2	-1	0	+1	+2
Temperature (°C) (X_T)	262.5	350	525	700	787.5
Residence time (s) (X_r)	77.5	100	145	190	212.5
Waste size (cm) (X_s)	0.5	1	2	3	3.5

Given the general function describing the interaction between the independent and dependent variables, the data obtained were fitted to a second-order polynomial equation:

$$Y = b_0 + \sum b_i X_i + \sum b_{ii} X_i^2 + \sum b_{ij} X_i X_j + \varepsilon$$

where *Y* is the response; b_0 refers to the constant, b_i represents the linear effect of the factor X_i (i=1, 2 and 3), b_{ii} stands for the quadratic effect of the factor X_i, b_{ij} represent the linear interaction effect between the input factors X_i and X_j, and the residual term is shown by ε. Variance analysis (ANOVA) was used for graphical analyses and for evaluating the significance of interactions between the process variables as well as the responses considering the Fisher,s F-test. Model terms were evaluated by the p-value (probability) and correlations were considered significant at α=0.05. Adequacy of various model test and the quality of fit of the regression model was assessed through sequential model sum of squares, model summary statistics, coefficient of determination (R^2) and adjusted R^2 (R^2_{adj}).

Analytical measurements

Sampling of the char residue was followed while processing the feedstock in the designated runs, as described in the next section. The collected samples were then subjected to PAHs analysis via a GC analyzer coupled with FID.

Char residue sample preparation

Char samples were initially homogenized within a homogeneous masonry. One gram of the sample was precisely weighed, moved to a 10 mL volumetric balloon, and then acetone was added to the sample and subjected to

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ultrasonic radiation for 30 min. The WiseClean Ultrasonic Bath, WUC-D10H was used. Finally, the sample was centrifuged and about 1 µL of clear supernatant solution was injected, into the GC analyzer. A Varian CP 3800 GC analyzer coupled with a FID detector was used to detect and measure the PAH compounds in both solid and liquid residues from waste pyrolysis runs aforementioned in this study. Nitrogen gas with a high purity (99.999%) was used as the carrier gas. The capillary used as GC separation column was CP-Sil8 (30 m × 0.32 mm × 0.25 µm). The GC/MS program was set-up as follows: injector temperature, 280°C; split ratio 7:1; initial pressure, 6psi; volume of injected sample, 1µL; FID detector temperature 300°C; initial column temperature, 120 °C, hold for 3 min; final temperature, 300°C, hold for 5 min. The measurement was tripled and the mean of the three measurement results was reported in this paper.

RESULTS AND DISCUSSION

Analysis of variance (ANOVA)

ANOVA test was applied to assess the adequacy of the quadratic model for prediction of ∑4-6 rings PAHs concentrations in the residue-char (Table 3). The statistical significance of factors was evaluated according to p-value and f-value. The significance of the model was confirmed by a high F-value. As shown in Table 3, the *F*-value of 73.05 for char, implies that the models are accurate and that there is a low chance of 0.01% for this large "model *F*-value" to occur due to noise. The p-value for char models are less than 0.0001. In order for the model to enhance, the insignificant terms can be further eliminated. According to ANOVA, the lack of fit tests were insignificant (3.82) which implies that the model sufficiently fitted the experimental data. To well present the quality of the model, the coefficient of determination R^2 and adjusted coefficient of determination R^2 were used to control the fitness of the models. Based on the ANOVA results, the value of the determination coefficient, R^2 = 0.9771, for the char confirms the compatibility of the model. The value of adjusted R^2 (i.e. 0.9637) shows a substantial agreement between the model-predicted and the lab-based experimental data. The generation of ∑4-6 rings PAHs in char can be expressed by the following equation:

$$Y\ (mg\ kg^{-1}) = -3612.89+4.099X_t+1409.12X_s+19.4X_r+0.026X_tX_r-6.26X_t^2-350.86X_s^2-0.096X_r^2$$

where Y is the total 4-6 rings PAHs generation and X_t, X_s, X_r, are temperature, waste particle size and residence time, respectively. It can be deduced from the variance analysis that for the first-order and second-order main effects, temperature, waste particle size and residence time exhibited primary importance. Comparison of experimentally-measured levels versus the predicted values for PAHs generation is presented in Fig. 2.

Table 3. ANOVA for Response Surface Reduced Quadratic Model of ∑4-6 rings PAHs in char.

Source	Sum of squares	df	Mean square	F value	p-value Prob > F	
Model	3.869E + 006	7	5.527E + 005	73.05	< 0.0001	significant
A-tem	7.161E + 005	1	7.161E + 005	94.65	< 0.0001	
B-size	401.29	1	401.29	0.05	0.8217	
C-DT	6.922E + 005	1	6.922E + 005	91.49	< 0.0001	
AC	3.490E + 005	1	3.490E + 005	46.13	< 0.0001	
A^2	3.793E + 005	1	3.793E + 005	50.13	< 0.0001	
B^2	1.269E + 006	1	1.269E + 006	167.68	< 0.0001	
C^2	3.966E + 005	1	3.966E + 005	52.42	< 0.0001	
Residual	90793.62	12	7566.14			
Lack of Fit	76504.54	7	10929.22	3.82	0.0794	not significant
Pure Error	14289.08	5	2857.82			
Cor Total	3.960E + 006	19				

Analysis of pyrolysis product

Char is a solid carbon-based (91-63%) byproduct of fast pyrolysis process (Guedes *et al.* 2018). It includes significant proportion of PAHs due to the inevitable nature of its production (Hilber *et al.* 2012; Kwon *et al.*

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2015); PAHs can be formed on the surfaces of char (Wang *et al.* 2017). The maximum acceptable concentration of Σ16 EPA PAHs in a safe pyrolytic char is 6–20 mg kg^{-1} according to the International Biochar Initiative (IBI) (Initiative 2012). Table 4 lists the central composite design matrix and the corresponding values of PAHs production (as total 4-6 rings PAHs) in the char products derived from the fast pyrolysis of hazardous medical waste samples. The concentration of 4-6 rings PAHs ranged from 54 to 1148 mg kg^{-1} that was remarkably higher as compared to other feedstock in similar studies and IBI threshold. The high value of total PAHs reported by Zhurinsh *et al*. (2005) was 146 mg kg^{-1} for pinewood charcoal produced during slow pyrolysis (Zhurinsh *et al.* 2005) and 2945 mg kg^{-1} total PAHs in a wood gasifier system (Schimmelpfennig & Glaser 2012). The results showed that generation of PAHs is a function of feedstock type and the processing conditions.

Table 4. The central composite design matrix for char derived from RSM.

Run	Temperature (°C)	Waste size (cm)	Residence time (s)	∑4-6 rings PAHs (mg kg^{-1})
1	525	2	145	1018
2	525	2	212.5	855
3	525	2	145	1008
4	350	3	100	100
5	262.5	2	145	106
6	525	2	145	1148
7	700	3	190	1021
8	350	1	100	54
9	700	1	100	102
10	350	1	190	62
11	525	0.5	145	217
12	525	2	145	1031
13	350	3	190	207
14	700	1	190	962
15	700	3	100	95
16	525	2	145	1031
17	525	2	77.5	161
18	525	3.5	145	102
19	787.5	2	145	929
20	525	2	145	1010

Effect of each operating variable (waste particle size, residence time and temperature) and their interaction on PAHs in residue-char derived from the medical waste fast pyrolysis are shown in Figs. 3a-3c. Pyrolysis temperature is one of the main factors affecting PAHs concentration in the char. It can be observed in Fig. 3a that the concentration of 4-6 PAHs is increased with the rise of pyrolysis temperature from 300 to 700°C which is in good agreement with other reports (Nakajima *et al.* 2007; Keiluweit *et al.* 2012). Fig. 3b represents the interaction profile of process temperature and waste particle size during the pyrolysis of medical waste. A significant increase can be found in the formation of aromatic hydrocarbons by elevating temperature, such that the maximum amount of 4-6 rings PAHs generation has occurred in 525°C with waste particle size of 2 cm, in agreement with Devi and Saroha (2015) who quantified PAHs content in the biochar. They have found that biochar derived at about 500°C had significantly higher PAHs concentration than that produced at other temperatures (Devi & Saroha 2015). The interaction between the temperature and residence time are shown in Fig. 3a, exhibiting that elevating in temperature and residence time influence on 4-6 rings PAHs generation, similar to that observed in previous studies (Sánchez *et al.* 2012a). A pattern of different behaviors is observed for total PAHs formation. PAHs concentration arise up to a residence time of 145s. Above this residence time, a slight decrease is observed which depends on the temperature. There is a statically significant difference in the total 4-6 rings PAHs produced by elevating in residence times and temperature. One of the main reasons for this increase, is that at elevated temperatures, most of the PAHs are adsorbed on char (Viteri *et al.* 2016). Fig. 3c displays the relationship between

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waste particle size and residence time of pyrolysis and 4-6 rings PAHs generation, exhibiting that the production rate has been elevated in general, but has slightly been reduced from 2 to 3 cm.

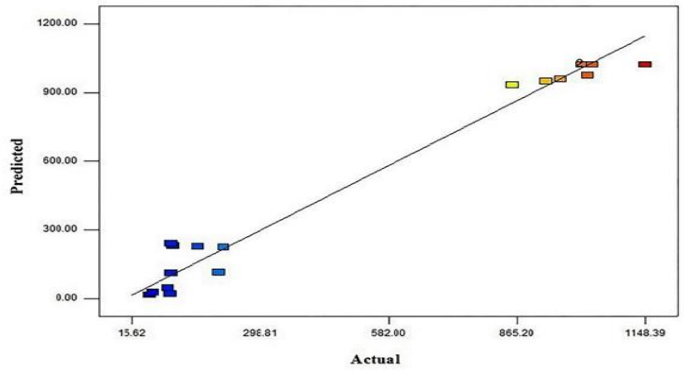

Fig. 2. The graph of predict vs. actual of PAHs generation in the residue-char.

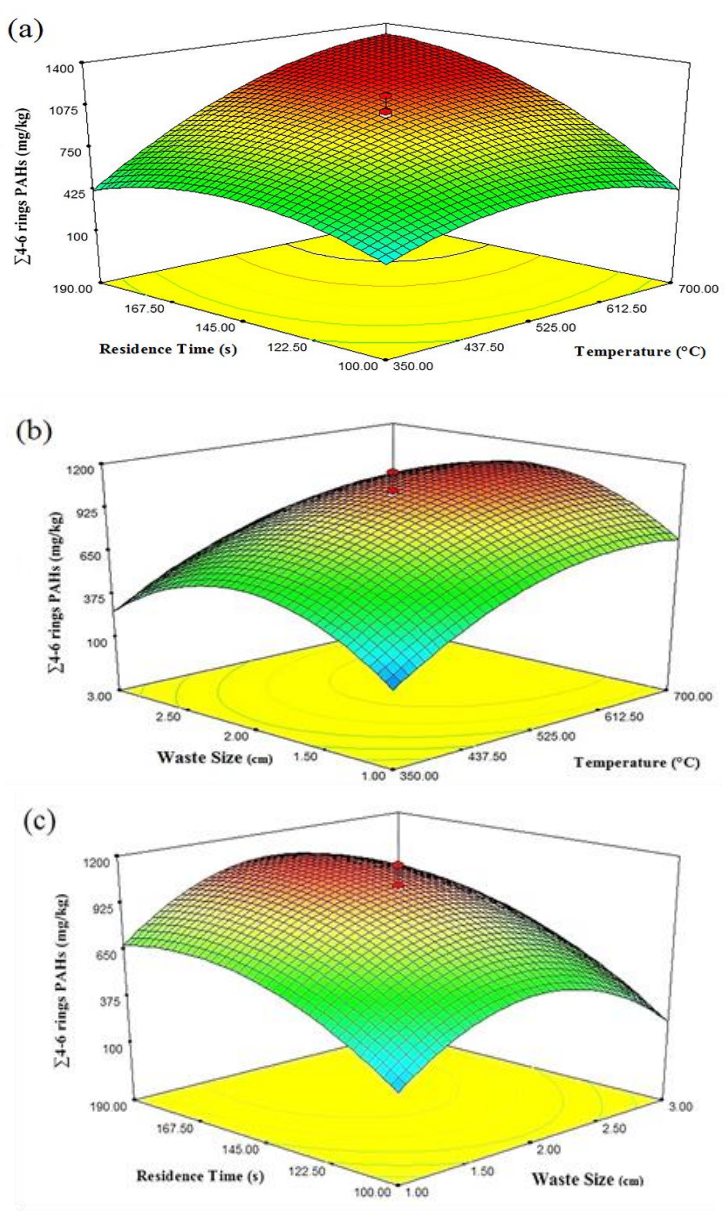

Fig. 3. The 3D plots of Total PAHs vs. pyrolysis variable operation factor interactions.

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By increasing in residence time, the 4-6 rings PAHs generation has been raised. This is consistent with the observations indicating that PAHs raise by prolonged residence time (Cunliffe & Williams 1998; Sánchez *et al.* 2012b). Based on these two factors, the maximum PAHs production has taken place at 2 cm and 145 s. Increased particle diameter has been shown to be positively correlated with the PAH generation at elevated pyrolysis residence times. Influence of the particle diameter on PAH yields is proposed as follows: larger particles undergo pyrolysis at lower effective particle devolatilization temperatures, which was found with a high degree of statistical significance to result in an elevated fraction of synapoyl aldehyde in the primary tar content, and ultimately in increased PAH formation (Stark & Ghoniem 2017).

CONCLUSIONS

In conclusion, we investigated the relationship between the different variable of medical waste fast pyrolysis factor (temperature, waste size and residence time) on 4-6 rings PAHs generation. PAHs are main byproducts in char. The interaction of temperature and residence time significantly influenced the total amount of 4-6 rings PAHs. Furthermore, by increased waste particle size from 1 to 2 cm, the ∑4-6 rings PAHs generation was elevated in the char, while reduced from 2 to 3 cm. The results of this study provide a better understanding of the medical waste fast pyrolysis and the influence of operation factors on the release of 4-6 rings PAHs during pyrolysis. This can help to control the emission of PAHs and improve the efficiency of medical waste pyrolysis.

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بررسی تاثیر دما، اندازه ذرات و زمان ماند بر میزان تولید هیدروکربن‌های آروماتیک چندحلقه‌ای در پیرولیز پسماندهای خطرناک پزشکی

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چکیده

مدیریت نامناسب پسماندهای پزشکی منجر به خطر افتادن سلامتی انسان و محیط زیست می‌شود. در حال حاضر، از فن-آوری‌های جدید پیرولیز برای تصفیه پسماندهای پزشکی استفاده می‌شود که می‌تواند علاوه بر بی‌خطرسازی و کاهش میزان دفن پسماندها، آنها را به سوخت هیدروکربنی تبدیل کند. یکی از آلاینده‌های حاصل از پیرولیز هیدروکربن‌های آروماتیک چند حلقه‌ای (PAHs) است. در این مطالعه، پسماندهای خطرناک پزشکی با استفاده از یک پایلوت نیمه صنعتی پیرولیز شد و تولید PAHs ۶-۴ حلقه‌ای در چار حاصل از پیرولیز پسماندهای پزشکی در شرایط متغیر پیرولیز از جمله دامنه دمای (۷۰۰-۳۰۰ درجه سانتیگراد)، زمان ماند (۱۹۰-۱۰۰ ثانیه) و اندازه ذرات (۳-۱ سانتیمتر) با هدف بهبود کیفیت چار برای بازیابی آن یا امکان استفاده به عنوان سوخت بررسی شد. برای شناسایی و اندازه‌گیری ترکیبات PAHs در چار از دستگاه GC با آشکارساز FID استفاده شد. نتایج نشان داد که PAHs در دامنه غلظت‌های متفاوت در چار وجود دارد ($mg\ kg^{-1}$ ۱۱۸۴-۵۴). میزان تولید PAHs ۶-۴ حلقه‌ای با تغییر دما، زمان ماند و اندازه ذرات پسماند متفاوت است و اثر متقابل معناداری بین زمان ماند و دما در تاثیر بر تولید PAHs مشاهده شد. با بهینه سازی شرایط بهره‌برداری پیرولیز امکان به حداقل رساندن میزان PAHs در چار وجود دارد.

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