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Substitution of Garden and Polyethylene Terephthalate (PET) Plastic Waste as Refused Derived Fuel (RDF)

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Abstract. The generation of polyethylene terephthalate (PET) plastic and garden waste must be recycled to support the circular economy. An alternative way to reduce the plastics waste is to reduce this waste by converting it into energy such as Refused Derived Fuel (RDF) as an alternative for processing waste. Substitution of plastic and garden waste is an opportunity to be analyzed. Hence, This study aimed to investigate the potential for converting material substitution from PET and garden waste into RDF. The RDF characterized test method was carried out by proximate, water content, ash content, and analysis. At the same time, the calorific value. was tested by bomb calorimetry. Substitution of the mixture of plastic and garden waste affects each parameter of RDF pellet quality including water, ash, and caloric value (sig.< 0.05). The increase of plastic waste in pellets consistently increases the calorific value of RDF from 18.94 until 25.04 MJ/kg. The RDF pellet water and ash content also invariably affect the rate of increase in the calorific value of RDF in the multilinearity model (sig.<0.05; R² is 0.935). The thermal stability of the pellets occurred at a temperature of 500°C decomposition of hemicellulose, cellulose, and lignin in mixed garden waste with plastic in RDF pellets. The decrease in the decomposition of PET into terephthalic acid monomer from the thermal stability of raw materials and waste PET plastic pellets occurs at a temperature of 4500° C. This potential finding can be used as a basis for consideration in regions or countries that have the generation of garden waste and plastic, especially the type of PET to be used as an environmentally friendly fuel.

Keywords: Garden waste, polyethylene terephthalate, refused derived fuel, waste to energy, caloric value

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1. Introduction

The increasing volume of waste and the limited land disposal has led to waste management innovations, namely making waste alternative energy (Rajmohan *et al.*, 2019; Raksasat *et al.*, 2021; Sarwono *et al.*, 2021). Therefore, the waste processing into energy has become a concern for the Depok City Government. This can be seen with implementing the Refused Derived Fuel (RDF) pilot project implemented by the Indonesia Government (Damayanti *et al.*, 2021; Mustia *et al.*, 2021; Qonitan *et al.*, 2021; Rachman *et al.*, 2020). RDF is a technology for processing urban waste into an alternative fuel to replace coal (Hajinezhad *et al.*, 2016). Waste used as raw material for RDF must have a high calorific value (Hwang *et al.*, 2014). Based on Arifianti's research (2018), RDF made from domestic waste has a calorific value of 21.68 MJ/kg to be used as a substitute fuel for coal (Arifianti *et al.*, 2018). Therefore, urban waste has a high calorific value and can be used as raw material for RDF (Kara *et al.*, 2009). So,

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plastic, wood, and paper waste have calorific values of 33.5 MJ/kg, 20 MJ/kg, and 17 MJ/kg, respectively. (Cheremisinoff, 2003).

Using plastic waste raw materials is expected to increase the caloric value of RDF. However, plastic waste that accumulates will disturb the environment because it is non-biodegradable (Sintim *et al.*, 2019). In addition, plastic waste can be used as raw material for RDF because it has a high calorific value than other urban waste (R Darmawan *et al.*, 2021; Syguła *et al.*, 2021). The addition of plastic waste can increase the calorific value of organic waste by mixing the two components of the waste.

One of the most common types of plastic waste in urban areas is PET plastic (Aryan *et al.*, 2019; Haque, 2019; Ma *et al.*, 2020; Sari *et al.*, 2022; Septiariva *et al.*, 2022). PET waste in the combustion process does not produce toxic exhaust gases, so it is safe for the environment as fuel (Alfahdawi *et al.*, 2019). Furthermore, based on previous increasing the calorific value of RDF made from the biomass can be done by mixing it with PET plastic (Hastiawan *et al.*, 2018). Plastic waste is considered a potential economic source of chemicals and energy (Mastellone, 2020; Meys *et al.*, 2020; Solis & Silveira, 2020).

Besides plastic, the high amount of biomass such as garden waste in the composition of waste in Indonesia makes RDF production an alternative for municipal solid waste treatment. The composition of municipal solid waste varies from place to place and lifestyle. Municipal solid waste that has not been treated has a high-water content, low heating value and very heterogeneous particle size like this garden waste. This causes the use of garden waste to be balanced with the substitution of high calorific value fuels such as PET plastic.

Many of us have come across various products that use plastic materials today. As a result of the increasing private consumption of plastic materials, large amounts of waste are discharged into the environment. Catalytic cracking is a process that converts plastic waste into valuable liquid hydrocarbon products that can be utilized as an energy source for various purposes such as diesel engines, generators, vehicles, etc. Furthermore, the gas byproduct obtained in the process can be used for domestic use. For example, filling in cylinders and running gas turbines. Thus, the cracking process can be considered as another unconventional energy source. This study aimed to analyze the potential for material substitution from PET and garden waste into RDF.

2. Materials and Methods

2.1 Sample

The pellet production carried out at TPSS Merdeka 3, Depok City, Indonesia, is equipped with a chopper, a rotary dryer that functions to dry raw materials, a hammer mill machine to reduce raw materials, and pellet printing. RDF pellets have essential characteristics: moisture content, ash, and calorific value. In this research, RDF pellets were made using raw materials from various garden waste and PET plastic with an interval of 25%. Nasir has done a combination of raw materials at 25% intervals (2015). Tapioca added as an adhesive (binder) was used to increase the bond between the material particles in the manufacture of pellets (Table 1).

Table 1

Variations in garden and PET compositions in the manufacture of
RDF pellets

ID	Garden Waste	PET Waste	Binder
	(gram)	(gram)	(gram)
PP-0%	2000	0	200
PP-25%	1750	250	200
PP-50%	1000	1000	200
PP-75%	250	1750	200
PP-100%	0	2000	200

The tapioca flour adhesive used is about 10% has a good density which produces firm RDF pellets and is not easy to crack. The addition of more than 10% adhesive will affect the decrease in density so that the level of pellet homogeneity will be lower (Damayanti, 2017). The percentage of raw material for RDF pellets from garden waste and PET plastic can be seen in Table 1.

2.2 Pelletizing Process

The raw materials used are garden waste and PET plastic collected in separate containers (Figure 1). Garden waste consists of wood twigs and leaves, where the wooden twigs are cut about 10 cm to be put into the chopping machine. Meanwhile, PET plastic waste is separated from PET plastic bottles with brand labels and then washed to clean the bottles. After that, the PET plastic bottles are cut to reduce the size inserted into the chopping machine.

The available raw materials are carried out in the next step, namely the enumeration process. This study used different machines to enumerate garden waste and PET plastic. The enumeration process reduces garden waste and PET plastic to a smaller size using a shredding machine. The raw materials that have been chopped are then subjected to a drying process to reduce the water content. This is because the water content that is too high will make it difficult to burn (Suryawan, Septiariva, *et al.*, 2022). In the garden waste drying process using a rotary dryer machine. PET plastic waste does not carry out a drying process because the chopper machine reduces and makes PET plastic waste dry. Therefore, the drying machine for garden waste uses a rotary dryer.

The sieving process is carried out so that the size of the waste is not too large to be put into the next machine. In addition, the sieve machine is designed with a magnet. The other component, such as nails or metal objects found in the garbage, is not filtered but are attached to the magnet. Next, garden waste and PET are put into a hammer mill machine that reduces the size of the raw material to powder particles. After the flouring process, weighing the garden waste and PET plastic with a predetermined percentage variation composition. In the weighing process using a digital scale.

In the next stage, the process of mixing garden waste and PET plastic with variations at intervals of 25% has been carried out by previous research (Wiyono *et al.*, 2021). Before mixing the pellet, add 10% starch as an adhesive. The addition of starch as an adhesive so that RDF is not easily brittle (Rati *et al.*, 2020; Saputro *et al.*, 2021).

After printing the pellets according to variations with 25% intervals, the pellets are dried in the sun for 1-2 hours. The drying process aims to reduce the water content in the

pellets. The results of making pellets from garden waste and PET plastic are then tested in the laboratory to determine the energy characteristics of RDF fuel.

There is a mechanical process involving heat and pressure in the molding process, which aims to solidify the materials through the holes in the pelletizing machine. The pellet molding machine has an optimum temperature of around $60-80^{\circ}$ C, making the pellets dense, dry, and shiny. If the pellet machine temperature exceeds 80 C and 300 MPa, pellet strength increases substantially (Huang *et al.*, 2017). After the pellet molding process with 25% interval variation, the pellets were dried under the sun. The drying process aims to reduce the water content to protect the pellets from fungal growth; the pellets are denser and do not break easily (Suryawan, Fauziah, *et al.*, 2022). In addition, the dried pellets are packaged in plastic packaging/airtight containers to prevent increased water content.

RDF pellets have a 6-8 mm size, which indicates the more significant the pellet size (Figure 2), the denser the pellet density. Figure 2 shows the shape of the RDF pellets in this study which had a length of \pm 3.2 cm and a diameter of \pm 7 mm for RDF. Figure 2 shows that PP-100% has a transparent color from PET. At PP-0%, it has a brownish color because it comes from Garden lignin. Meanwhile, PP-75%, PP-50%, PP-25% have a brownish and clear color which comes from mixing garden lignin and PET (Figure 3).

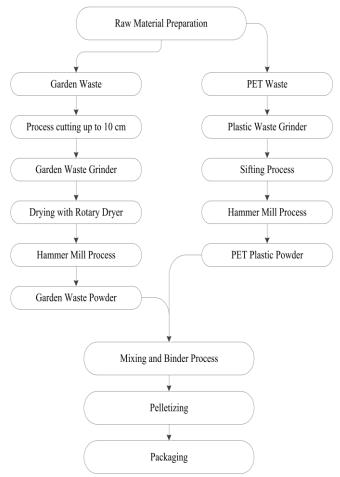


Fig. 1 Process Diagram for Making Mixed Garden and PET Waste into RDF Pellets



Fig. 2 Measurement of Dimensional of Garden and PET Waste into RDF Pellets

2.3 RDF Quality Measurement

Analysis of the moisture content of raw material for garden waste and PET plastic is carried out as raw material for RDF manufacture. The water content was analyzed by following the American Standard Test Method for Residual standards in an RDF Analysis Sample E790-87 (2004). The empty porcelain cup is heated and cooled in a desiccator for 15-20 minutes. The sample was stored in a porcelain cup as 10 grams and weighed. The sample was put into an oven at a temperature of 107 ± 30 C for 1 hour. The sample is removed from the oven and cooled in a desiccator at room temperature, then the sample is weighed, and the sample weight is recorded after heating

Analysis of the moisture content of the pellet samples was carried out by following the standards of SNI 1965-2008. First, the empty cup is heated in an oven at a temperature of 105° C for ± 2 hours until the cup's weight becomes constant. The weight of the cup and sample was weighed, then the sample was stored in the cup and heated for ± 24 hours. After that, the sample was weighted to ensure a constant weight, and the sample was reheated for 24 hours. The sample was constantly cooled in a desiccator at room temperature. Finally, the samples were weighed, and the weight recorded to get the results of the water content value based on weighing before and after heating.

Ash content is the residue of the combustion process at high temperatures. Calculation of ash content is based on the standards of the American Standard Test Method for Ash in the Analysis Sample of Refused Derived Fuel Analysis Sample E830-87(2004). The sample used is the sample at the previous water content. Then the sample was stored in a porcelain dish and heated in a furnace at a temperature of $700\pm25^{\circ}$ C. After that, the sample is removed from the furnace and cooled in a desiccator at room temperature.

The calorific value analysis was tested using a bomb calorimeter. The caloric value analysis is based on the standard of the American Standard Test Method for Gross Calorific Value D5865-11a. Samples were taken as much as \pm 0.5-gram and put into the Bomb calorimeter. After that, the tool will automatically analyze the sample, and the results will come out when the analysis has been completed.

The TGA/DTG analysis was analyzed at a temperature of 30 to 800°C, and the gas used was nitrogen with a temperature rate of 10°C/minute. The TGA/DTG used is a type of TGA/DTG instrument SDT 650. The TGA/DTG test is carried out utilizing a pellet sample in powder as much as not less than 5 mg is inserted into an alumina container. The sample entered will automatically be analyzed by the tool by producing a graph of the decrease in mass against temperature. In addition, the GC-MS (Gas Chromatography-Mass Spectroscopy) is also

used to measure RDF chemical compounds' type and content.

2.3 Data Analysis

Analysis of data on variations in garden waste and PET plastic as raw material for RDF pellets was analyzed with several variables, including analysis of moisture content, ash content, calorific value, TGA/DTG, and GC-MS. Analysis of the data obtained will be compared with the RDF standard to determine the potential variation of garden waste and PET plastic into RDF pellets.

3. Result and Discussion

3.1 RDF Quality Measurement

Mixing variations in garden waste and plastic pellets affect the percentage value of the moisture content of RDF pellets (Table 2). Garden waste contains water in the pores, so it is difficult to evaporate. This is different if the water content on the surface will be more easily evaporated than plastic waste. The increasing percentage of plastic indicates that the lower the water content is because the plastic does not have a lot of water content (Montejo *et al.*, 2011), so when mixed with garden waste, it will affect the water content produced. The water content value increased in the sample PP-100% to sample PP-0% due to adding garden waste to the formation of pellets. In addition, the addition of starch affects the pellet formation process. Starch flour acts as an adhesive that has more waterbinding properties than other types of bonds (N. Singh *et al.*, 2004). All garden waste and PET plastic variations samples can be used as RDF because they have a moisture content value that meets Indocement standards below 20%. The decreased water content will cause an increase in calorific value (Wang *et al.*, 2020). The ash content of the garden waste and plastic pellets that had the highest value was in PP-25% pellets at 3.57%, while the sample had the lowest water content, namely in PP-75% pellet variations at 1.33%.

On the other hand, the higher the percentage of garden waste composition in the mixture of garden waste and PET plastic variation, the lower the calorific value. This is because the design of garden waste has high water content, decreasing the calorific value (Yildiz et al., 2013). In this study, the results of the calorific value test were compared with several standards, namely the British, Italian, Finnish, and Indocement RDF standards, to determine whether they were suitable for RDF. The calorific value of garden waste and PET plastic is above Indocement standards and British, Italian, Finnish RDF standards. Therefore, all garden waste and PET plastic samples have the potential to be used as RDF, with samples PP-100%, PP-75%, PP-50% having relatively higher heating values than the others. The RDF pellet shows the calorific value is relatively high because PET plastic waste affects the calorific value (Table 3).

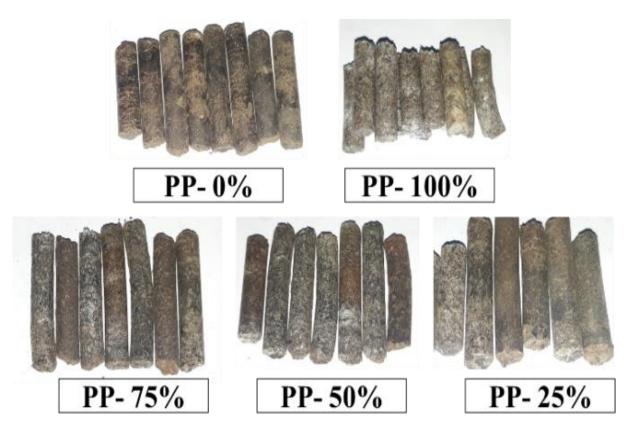


Fig. 3 RDF Pellets Visual from Garden and PET Waste

Table 2

\mathbf{C}	Characteristics of RDF Pellet Samples from Garden and PET Waste							
	No	Sample ID	Water Content (%)		Ash Content (%)		Caloric Value (MJ/kg)	
	1	PP-0%	15.21	± 0.16	4.39	± 0.22	18.94	± 0.25
	2	PP-25%	13.91	± 0.71	3.47	± 0.14	19.69	±0.33
	3	PP-50%	9.03	± 0.69	2.18	± 0.07	20.88	± 0.76
	4	PP-75%	6.98	± 0.85	1.37	± 0.06	23.77	± 0.76
	5	PP- 100%	3.83	± 0.43	0.48	± 0.21	25.04	± 0.66
	One-Sample T-Test							
	t 8.586		6.315		34.0	19		
	Sig. (2-tailed) 0.000		0.000		0.000			

Table 3

Results of ANOVA Analysis of Water Content, Ash Content, and Caloric Value Parameters on Variations in RDF Pellet Composition

Parameters		Sum of Squares	df	Mean Square	F	Sig.
Water Content	Between Groups	271.23	4	67.808	354.38	0
	Within Groups	1.913	10	0.191		
	Total	273.144	14			
Ash Content	Between Groups	29.635	4	7.409	611.021	0
Ash Content	Within Groups	0.121	10	0.012		
	Total	29.756	14			
Caloric Value	Between Groups	83.375	4	20.844	117.582	0
	Within Groups	1.773	10	0.177		
	Total	85.148	14			

Table 4

Results of Pearson Correlation Analysis of Water Content, Ash Content, and Caloric Value Parameters

	Caloric	Water	Ash
Caloric	1	-0.965	-0.966
Water	-0.965	1	0.993
Ash	-0.966	0.993	1

3.2 Statistic and Model Analysis

The value of water content affects the ignition speed of RDF fuel (Faujiah, 2016). Table 3 shows the results of the ANOVA test on the variations used in this study. The calorific value is an important parameter determining RDF pellets' quality as fuel (Białowiec et al., 2017; Kara et al., 2009; Ozyuguran et al., 2018). The calorific value of variations in garden waste pellets and PET plastic which has the highest heating value, is found in the PP-75% sample, 24.77 MJ/Kg, while the lowest is in the PP-25% sample, which is 20.32%. The addition of plastic waste to garden waste increases the calorific value (Naryono & Soemarno, 2013). The higher the percentage of PET plastic waste, the greater the calorific value produced. Pearson correlation is a form of the formula used to find the relationship between two, which in this study consists of calorific value, moisture content, and ash content. It can be seen that the relationship between water content and ash content is very high in increasing the calorific value (Table 4). At the same time, the increase in water content affects the ash content. Multicollinearity is a condition with a perfect linear relationship between water content and ash variables to the caloric value. Multicollinearity includes looking at the Variance Inflation Factor (VIF) value and its tolerance. If the value of VIF < 10 and Tolerance > 0.1, it

means that there is no multicollinearity. This study shows that they have met the established criteria (Table 3, Figure 4).

3.3 TGA/DTG Analysis

Fig 5 shows the curve of % mass against temperature in thermogravimetry testing to produce TGA and DTG. In testing the thermogravimetric analysis results (TGA), the decrease in the percentage of sample weight is related to the increase in temperature. The TGA/DTG curve process has three stages, namely: the drying stage, which is a slow mass reduction process with a temperature of 100°C, the second stage is devolatilization, which is a drastic mass reduction process and evaporation of the sample compounds tested.

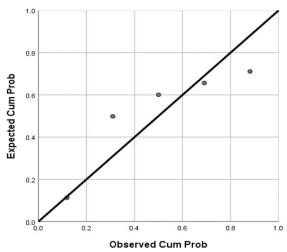


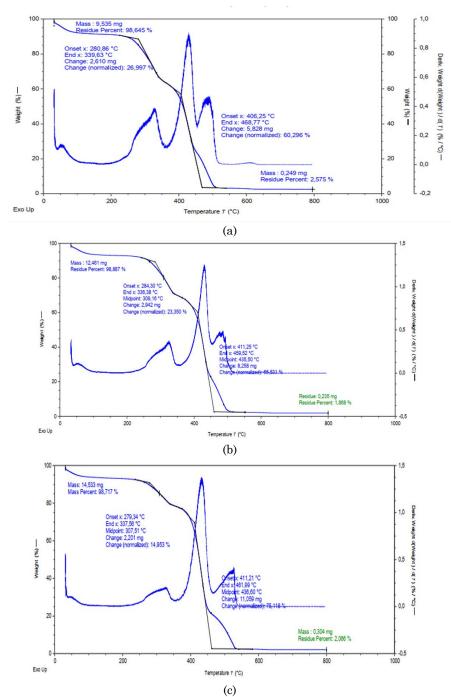
Fig. 4 Probability Plot of Variations in RDF Pellet Composition

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Table 5

Multicollinearity Analysis Water Content, Ash Content, and Caloric Value Parameters on Variations in RDF Pellet Composition

Model	Unstandardized Coefficients	Collinearity Statistics	
	В	Tolerance	VIF
(Constant)	26.152		
Water	-0.241	0.014	73.103
Ash	-0.893	0.014	73.103
R	0.967		
R Square	0.935		
Adjusted R Square	0.871		
Sig. F Change	0.065		



(c) Fig. 5 TGA Result for RDF Pellet Samples from Garden and PET Waste with composition PP-25% (a), PP-50% (b), and PP-75% (c)

The sample experienced a reduction in mass caused by the evaporation of water bound to the sample and gas release due to hemicellulose, cellulose, and lignin (Shotorban *et al.*, 2018). Gardens have the main chemical components with the decomposition of hemicellulose in the temperature range 200-260°C, cellulose in the temperature range 240-340°C, and lignin in the temperature range 280-500°C (R. Singh *et al.*, 2015). The last stage is the carbonization stage, a decrease in mass that slows down again and tends carbonized at 700°C after thermal stabilization at 250°C (Zhong *et al.*, 2022).

PP-75% is declining on a steep chart. The decomposed sample in the second stage has a decreasing peak temperature in the PP-100%, PP-75% samples with a temperature of 541.70° C to 582.55° C mindset; onset temperature at 405.270C to 459.17 0C mindset, and onset temperature at 411.21° C to 461.99° C mindset. Mass reduction occurs at a temperature of 390 0C-420 0C due to the thermal decomposition of PET because of depolymerization. PET was depolymerized completely in 60 min at 230° C and in 30 min at 240° C (Liu *et al.*, 2012).

An endothermic process will appear in the temperature range of $350-452^{\circ}$ C. Because the water molecules evaporate, the PET will decrease into a terephthalic acid monomer. In addition, the long-chain structure of PET causes a decrease in mass at high temperatures (Yang *et al.*, 2018). This occurs due to the breakdown of carbon chains followed by the release of CO₂, hydrocarbons, and hydrogen gas.

The third stage is the carbonization process with a peak temperature range of 450 0C - 550 0C. The Garden sample has a rapid carbon formation process because it has a lot of organic content. The garden contains lignin which decomposes at $280-500^{\circ}$ C (Ahmed *et al.*, 2018). In the temperature range 600-800°C, it shows that the line that starts to flatten or the rate of mass decrease is more stable

(constant). Loss of mass weight does not occur again because the sample has reached the maximum loss/decomposition point.

3.4 GC-MS Analysis

PET plastic can decompose into gas, liquid, and solid phases. At 600 °C pyrolysis with PET material, gas dominated with CO_2 , benzene, vinyl benzoate, benzoic acid, and divinyl terephthalate (Brems *et al.*, 2011; Dimitrov *et al.*, 2013). PET plastic at low temperatures is dominated by TPA (Terephthalic Acid) (Yoshioka *et al.*, 2003). TPA will decompose into benzene, CO_2 , and benzoic acid at high-temperature conditions. TPA is a benzene (CH) molecule and a carboxylic group (COOH). Therefore, the gas content of PET consists of these substances. However, in this study, the most common compounds found in the GC-MS test were phenol (Figure 6). This compound is a member of phenols, a dimethoxybenzene and a phenylpropanoid.

Dimethoxybenzene is an organic compound with the formula C_6H_4 (OCH₃). It is one of the three isomers of dimethoxybenzene. It is a white solid produced by some plant species. Dimethoxybenzene and a phenylpropanoid may be found in garden waste. In addition, one of the easiest fatty acids to obtain is palmitic acid or hexadecanoic acid. Plants from the Palmaceae family are this fatty acid's primary source, such as coconut and oil palm.

Isomers in the GC-MS test results consist of molecules with the chemical formula phenol and often with the same type of bond but have a different arrangement of atoms (can be likened to an anagram). More complete chemical elements obtained from a mixture of garden waste and PET can be seen in Table 6.

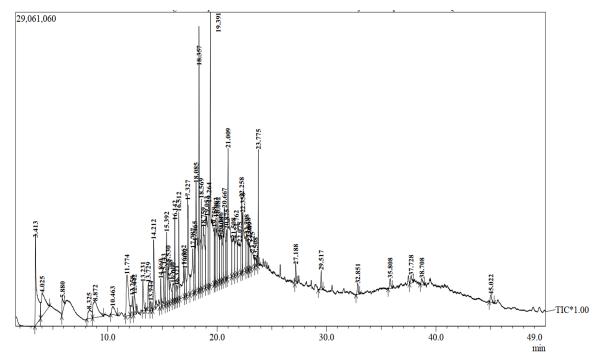


Fig. 6 GC-MS Result for RDF Pellet Samples from Garden and PET Waste with composition PP-50%

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Table 6

No	Name	%Conc
1	Phenol, 2,6-dimethoxy-4-(2-propenyl)- (CAS) 4-Allyl-2,6-dimethoxyphenol	7.7
2	Hexadecanoic acid (CAS) Palmitic acid	5.65
3	2-Azetidinone, 3,3-dimethyl- (CAS) 3-PIVALOLACTAM	4.5
4	Cyclopropane, 1,1-dibromo-2-chloro-2-fluoro- (CAS) 1,1-DIBROMO-2-CHLORO	3.73
5	1,4-Benzenedicarboxylic acid, methyl ester (CAS) Methyl terephthalate	3.6
6	Nonanoic acid (CAS) Nonoic acid	3.44
7	Phenol, 4-(3-hydroxy-1-propenyl)-2-methoxy- (CAS) Coniferyl alcohol	3.32
8	Octadecanoic acid (CAS) Stearic acid	3.19
9	MYRTENSAEUREBROMID	3.01

6. Conclusion

Substitution of the mixture of plastic and garden waste affects each parameter of RDF pellet quality. RDF pellet parameters also consistently affect the rate of increase in the heating value of RDF. The thermal stability of the pellets occurred at a temperature of 500°C decomposition of hemicellulose, cellulose, and lignin in garden waste mixed with plastic RDF pellets. Thermal stability of the raw material and RDF pellets of PET plastic waste occurs at a temperature of 450°C decreasing decomposition of PET into terephthalic acid monomer. The endothermic process occurs due to water bond b and the bond between the PET contained in the plastic. Therefore, all garden waste and PET plastic samples have the potential to be used as RDF, with samples PP-100%, PP-75%, PP-50% having relatively higher heating values than the others. It can be seen that the relationship between water content and ash content is very high in increasing the calorific value. With this potential, it can be used as a basis for consideration in waste reduction potential from the generation of garden waste and plastic PET to be used as environmentally friendly energy.

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