Kinetics of Gas and Liquid Smoke from Catalytic Pyrolysis of Human Hair Cut Waste and Its Characterization

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Abstract. Human haircut waste (HHW) is categorized as waste that is difficult to decompose by microorganisms in water and soil and has high stability to temperature. HHW disposal carelessly will cause environmental pollution problems. Incineration of HHW produces gas with a pungent odor that can interfere with the health of humans and other living organisms. Therefore, there is a need for a solution to handle HHW to prevent environmental pollution. The main objective of this research is to study the reaction kinetics of gas and liquid smoke formation resulting from the catalytic pyrolysis of HHW and its characterization. Pyrolysis is the method used in this study. The catalytic pyrolysis of HHW resulting in the gas and liquid smoke formation follows a first-order reaction, with the R-value for gas and liquid smoke being $R^2 = 0.9609$ and 0.9652, respectively. The relationship between the constant value of the reaction rate for the formation of gaseous products and temperature can be expressed in the Arrhenius equation $k = Ae^{-2,985.2/T}$ with an activation energy value (E_a) of 24.818×10^3 kJ/mol, while the liquid smoke is expressed in the Arrhenius equation $k = Ae^{-1.297.5/T}$ with an E_a of 10.787×10^3 kJ/mol. The characteristics of physical properties of liquid smoke from HHW are not yet suitable for use as fuel oil. Thereby, a method for further purification is needed so that the products can be used as fuel or other chemicals. Liquid smoke resulting from HHW pyrolysis is a mixture of 30 chemical compounds, and some of them are unstable compounds which could be indicated by a change in the color of the liquid smoke from pale yellow to dark brown in a relatively short time. Therefore, HHW liquid smoke must be stabilized with chemical stabilizers for long-term storage.

Keywords: Kinetics · gas · liquid smoke · pyrolysis · catalytic · hair · characterization

1   Introduction

Human haircut waste (HHW) generally comes from the activities of cutting men's or women's hair in a barbershop or salon. In contrast to organic waste that comes from plants, HHW is not easily digested by microorganisms in soil and water. The main constituent of human hair [1, 2] consists of 51% carbon, 21% oxygen, 17% nitrogen, 6% hydrogen, and 5% sulfur, as well as small amounts of minerals, 1% melanin and pheomelanin. In general, human hair fiber contains 50–60% keratin, 20–30% protein matrix, various pigments such as melanin, up to 30% water, and lipids [3]. The strength of human hair is due to the accumulation of keratin which forms fibrous microstructures known as filaments. A sulfide bond is a very strong cross-link (disulfide bond) that can be found, for example, between the two amino acid groups of cystine in two adjacent helical chains. Reports explained that matrix proteins that provide disulfide bonds and bind the two chains together are referred to as keratin-bound proteins [4, 5]. From the chemical composition of human hair, almost all keratin and its derivatives are difficult to degrade in nature and difficult to be processed by microorganisms in the soil. Meanwhile, urban areas with high population density, HHW often accumulates in large quantities in solid waste streams and clogs drainage systems leading to environmental problems [6]. Elimination of HHW by incineration produces a foul odor and toxic gases such as ammonia, carbonyl sulfide, hydrogen sulfide, sulfur dioxide phenol, nitrile, pyrrole, and pyridine which can cause some respiratory problems [7]. In many of these units, improper HHW effluent and other waste disposal has led to pollution and legal disputes [8, 9]. Workers at several HHW processing facilities in India created hair dust and rotting hair, which led to a rise in tuberculosis cases and respiratory illnesses [6].

Efforts to reduce the bad nature of HHW, which is difficult to degrade in nature, have been carried out by hair scavengers to be sold to the industry. Several salons collect hair waste and then process it themselves into hair extensions [10]. Frequently, the waste hair is sold to several home industries for the manufacture of buns or wigs. The management of human haircut waste necessitates systemic thinking due to the significance of protecting both the environment and human health, especially in regard to how to potentially treat HHW so that it can be used for the benefit of people and the environment or can be converted into useful and economically valuable products. In the field of technology and science development, everything can be processed and benefited. Introduction of new materials and technology has several sociocultural and economic aspects associated with it, which often determine the adaptability of these uses and technologies [11]. Therefore, it is essential to create the proper use of technology to treat HHW in the context of the necessity to take into account all potential uses and technologies as well as their effects on society, the environment, and the economy.

Human hair is a common biological waste, claim Bhandari and colleagues [3]. To create various kinds of useful materials, it can be pyrolyzed. Pyrolyzed hair has been applied in a variety of traditional medical procedures. Toxic metals (Ba, Cd, Pb, Sr) are among the numerous elements present in adult human hair [12].

One approach that has been described previously to process HHW is pyrolysis technology. The pyrolysis method is an alternative to treating municipal solid waste and is thought to have a good chance of being developed [13]. Since its mechanism of action

is simple and the pyrolysis process does not generate new waste that can contaminate the environment, the pyrolysis approach is one way to treat solid waste, especially HHW [14]. Another benefit is that solid waste can be pyrolyzed to create products with high energy contents and ratios [15]. To create a good and effective operating system or mechanism, pyrolysis techniques for treating solid waste, especially HHW, must be continuously developed. The degradation of materials during pyrolysis in the HHW process needs to be thoroughly explored. Chemical kinetics of substances, such as time, temperature, and catalyst, are directly related to the degradation of chemical compounds in the pyrolysis process. They have an impact on the production yield. The use of a catalyst may hasten chemical deterioration and shorten the hydrocarbon chain [16, 17]. Additionally, using catalysts can boost product selectivity according to desired outcomes [18] and can raise the yield of pyrolysis product yields. Pyrolysis is a reaction that breaks down materials at high temperatures with little oxygen present. There are multiple steps to the reaction, including the stages of breakdown, gas production, condensation, and termination. According to Ramadhani et al., zeolite has the characteristics of being hollow with a certain pore size and has cations that can be exchanged [19]. Therefore, zeolite can be used as a catalyst. The catalyst used in the pyrolysis process has the main requirements of having a large surface area and pore volume [20]. An example is shown by pyrolysis of polypropylene (PP) plastic waste using a physically activated zeolite catalyst [21]. Chemical activation by acidification using a hydrochloric acid activator to remove impurities and increase the acidic atmosphere of the zeolite could affect the activity of the zeolite as a catalyst [22]. Natural and synthetic zeolites are widely used because they have distinctive and unique properties, especially for adsorption and catalysts [23].

When solid waste, particularly household hazardous waste (HHW), is pyrolyzed, it yields liquids, gas products, and the remainder as solids or charcoal. Liquid smoke and charcoal are typically the sole byproducts studied by scientists who study the pyrolysis of solid materials; the gaseous byproducts are rarely investigated. There is currently a dearth of knowledge regarding the dynamics of the gaseous and liquid smoke produced by the pyrolysis of HHW. In order to determine the sequence of the reaction and to calculate the activation energy using the Arrhenius equation, the pyrolysis of HHW in this study will concentrate on watching and describing the behavior of the produced gas and liquid smoke. Specifically, HHW liquid smoke was treated with physical properties analysis and combined analysis of gas chromatography with mass spectroscopy (GC-MS).

2   Materials and Methods

2.1   Materials

The equipment includes an analytical balance (Metler Toledo), a digital bomb calorimeter (IKA C2000), a stative clamp, a condenser, a round bottom flask, glass tubes, measuring cups, glass pipes, thermometers, stoppers, scissors, and a combination of GC-MS tools. HHW that has been washed and dried in the sun serves as the study's sample. The natural zeolite catalyst was 40 mesh in size. The pyrolysis apparatus circuit is depicted in Fig. 1 and was created under vacuum. Figure 2 displays HHW samples and zeolite catalyst.
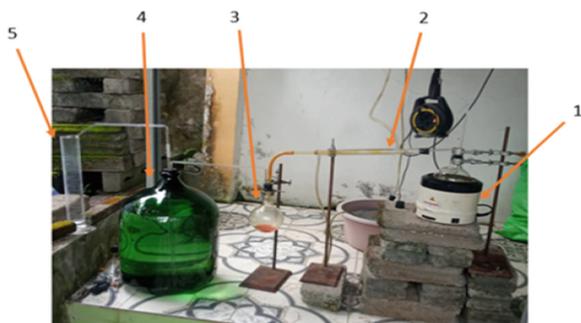
Fig. 1. The pyrolysis equipment circuit. The numbers indicate (1) electric coat, (2) condenser, (3) liquid smoke reservoir, (4) gas reservoir, and (5) measuring cup for water reservoir.
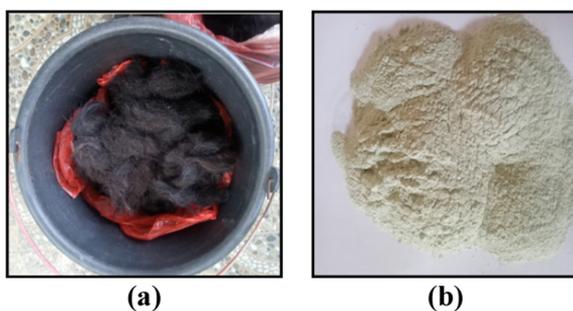
Fig. 2. (a) HHW sample and (b) zeolite catalyst

2.2 Method

Pyrolysis of as much as 200 gr of washed and sun-dried HHW was slowly put into a round flask. Then 10 g of zeolite catalyst was added. After all the samples had been filled in the flask, it was closed tightly until no leaks were found in the flask cover or leaks in the tool connections. After that, HHW pyrolysis was started. Observations on the pyrolysis of HHW were carried out on the produced gas and liquid smoke. A sizable bottle that contained 25 L of water was the recipient of the generated gaseous product. The water is forced into the gas reservoir by the gas, and the amount of water flowing into the cup is then accommodated. The amount of water that can be held is inversely related to the amount of gas produced during pyrolysis. Bottles were used to collect liquid-smoked goods, and new, empty bottles were added to the collection bottles every 15 min. Every 15 min, the temperature and length of the pyrolysis were also noted, along with the volume of produced gas and liquid smoke. To ascertain the reaction sequence and activation energy, the volume of gas and liquid smoke was measured and studied. Physical characteristics analysis and GC-MS analysis were specifically done for liquid smoke.

3   Results and Discussion

3.1   Gas Products

The physical change process began to melt during the catalytic pyrolysis of HHW at a temperature of 85 °C. Additionally, a white smoked substance that resembles fog forms in its wake. The produced gas was poured into the gas storage bottle. A measuring cup was used to contain the water that was forced to flow by the gas. The pyrolysis temperature of the HHW or the pyrolysis system with an isothermal process is nearly constant at 134 °C after 75–210 min. Within 15 min, the amount of gas produced by the catalytic pyrolysis of HHW was visible. The entire HHW pyrolysis procedure lasted for about 5 h. It is discovered after measurement that the amount of gas generated was 12.205 mL. By poking a needle-sized hole in the bottle stopper, the pyrolysis gas was released from the container and burned. The gas flame had the same size and clear yellowish hue as a candle flame. Short-chain hydrocarbon molecules or chemical compounds that fit the description of methane gas make up the gas produced by the pyrolysis of HHW. Pyrolysis produces a gas that can be burned directly [24] or converted into another fuel, like liquefied petroleum gas (LPG).

3.2   Liquid Smoke Products

Pyrolysis observations were performed at 75–210 min when the temperature began to approach a constant value at 134 °C. The duration of the HHW pyrolysis process was approximately 5 h. The total volume of liquid smoke produced was 50.5 mL (25.25%) from the 200 g sample. Other studies revealed that the pyrolysis process could be carried out with and without a catalyst [25, 26]. The advantage of catalyst pyrolysis is that it reduces the liquid fraction and increases the gas fraction. The catalyst in the pyrolysis process serves to lower the reaction temperature, speed up the reaction, and produce products with more specific carbon atoms and lighter hydrocarbons.

    The liquid smoke obtained from the pyrolysis of HHW was subjected to a flame test. The liquid smoke could burn with a clear yellowish flame in the middle and at the bottom and red ends of the flame. A current report showed that the physical properties of the fuel mixture between bioethanol and premium showed that the higher the viscosity of the mixture, the lower the heat of the fuel [27]. This decrease is caused by the difference in the calorific value of the two fuels, and the low calorific value is reddish yellow. The picture of the HHW liquid smoke flame is shown in Fig. 3.

    The longer the HHW pyrolysis time, the fewer the reactants and the higher the products are. As a result of considerably high pyrolysis temperature, chemical compounds with large molecular weights in the reactants were reduced and decomposed into smaller molecules as liquid smoke products. Meanwhile, the product of liquid smoke was increasing, and there was less charcoal left in the reactor. Theoretically, it can be explained that the use of the catalyst in the pyrolysis process speeds up the decomposition process of chemical compounds and supports the reactants to react more easily. Catalysts can also lower the activation energy so that the reaction is easier to achieve. HHW pyrolysis using the catalyst can be achieved at a temperature of 134 °C which was lower (173 °C) when compared to pyrolysis without the catalyst [28]. This agrees other opinion that the

Fig. 3. Flame test of liquid smoke from HHW in this study.

Table 1. Data of gas and liquid smoke generated from HHW pyrolysis with the addition of zeolite catalyst.

Time (minute)	Volume of gas product (mL)	Volume of liquid smoke product (mL)
15	1450	0
30	1300	0
45	1200	2
60	1100	4
75	1150	8
90	1000	7
105	920	7
120	780	5
135	750	4
150	730	3
165	500	2
180	340	2
195	300	2
210	300	2
225	210	1
240	115	1
255	0	0.5
270	0	0
285	60	0
300	0	0
Total	12,205	50.5

Table 2. Table of reaction rate equations

Order	Line Equation
Order 0	$[A] = -kt + [A_0]$
Order 1	$\ln[A] = -kt + \ln[A_0]$
Order 2	$\frac{1}{[A]} = -kt + \frac{1}{[A_0]}$
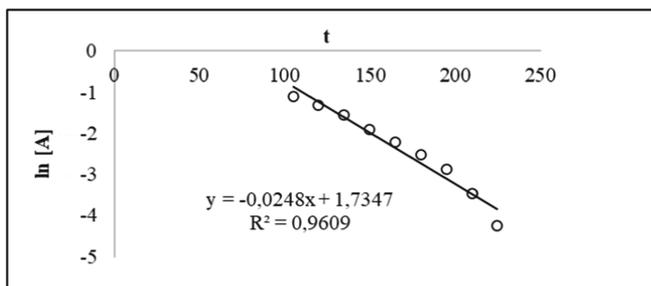
Fig. 4. Line equation curve of gas produced from HHW pyrolysis using zeolite as a catalyst.

catalyst could lower the activation energy and result in a greater reaction rate [29]. In this study, the use of a catalyst in HHW pyrolysis was proven to reduce the temperature of the reaction.

3.3 Order of Reaction for Gases and Liquid Smoke

The equations of the lines and curves are obtained by processing the data in Table 1 graphically in MS Excel. To choose the best response order, the values from the line equation were compared to the integrated rate equation formula [30, 31], as shown in Table 2.

Figures 4 and 5 are line equation curve obtained from data on gas and liquid smoke resulting from HHW pyrolysis with the addition of a zeolite catalyst.

By comparing the value of R square (R^2) obtained from the integrated rate equation, the determination of the order of the reaction can be done by observing the value of R square (R^2), which is closest to one [32]. Based on the calculation of the rate equation, the R^2 values for the gas and liquid smoke produced by pyrolysis are 0.9609 and 0.9652, respectively. The R^2 values of the two-line equations in Figs. 4 and 5 are both close to the value of one that both equations follow a first-order reaction meaning that if the gas concentration is increased, the reaction rate will increase.

3.4 Activation Energy and Arrhenius Equation

The rate constant exponential equation illustrates the connection between the reaction rate constant and temperature. As a result, the activation energy's value can be determined. The very minimum energy required for a reaction to occur is called activation
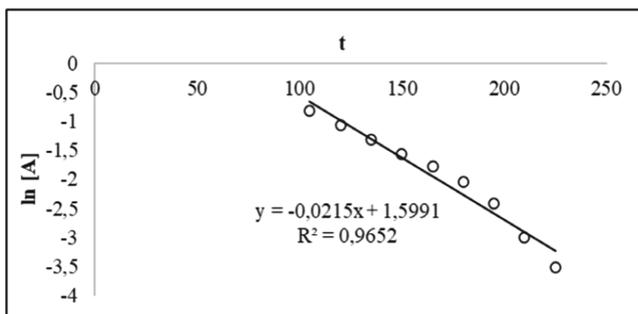
Fig. 5. Line equation curve of liquid smoke produced from HHW pyrolysis using zeolite as a catalyst.
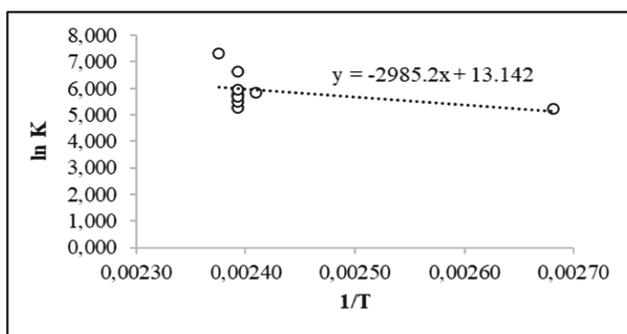
Fig. 6. Plot ln K versus 1/T and line equation of gas product from HWW pyrolysis.

energy. The following formula [33, 34] was used to calculate the rate constant for each time interval from observation data on the outcomes of the pyrolysis of gas volume and liquid smoke coming from the pyrolysis of HHW with the addition of a zeolite catalyst (Table 1).

$$\frac{dml}{dt} = K.f\left(\frac{ml - mla}{mlo - mla}\right) \tag{1}$$

And dt is the change in time, ml is the volume at the time interval, mlo is the initial volume, and mla is the final volume; where dml is the change in volume. A plot of ln K against 1/T can be produced to identify the equation of the line based on the constant value for each time interval acquired.

In order to calculate the activation energy (Ea), each line equation created from the plot of ln K vs 1/T in Figs. 6 and 7 is entered into the Arrhenius equation in a linear form [35].

$$\ln k = \left(-\frac{E_a}{R}\right)\left(\frac{1}{T}\right) + \ln A \tag{2}$$
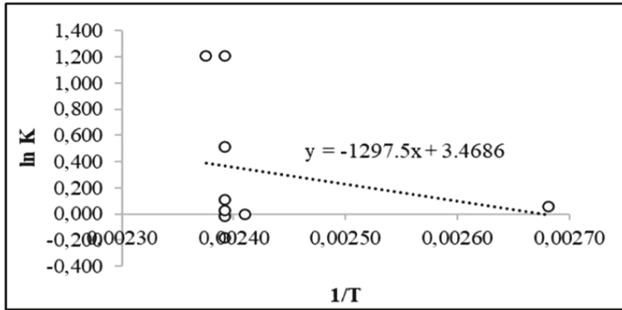
Fig. 7. Plot ln K versus 1/T and line equation of liquid smoke from HHW pyrolysis

Table 3. The value of activation energy and Arrhenius equation

Products of pyrolysis of HHW with Catalyst	Observation temperature (K)	Slope ($-E_a/R$)	E_a (kJ/mol)	Exponential rate constant equation (minute^{-1})
Gas	413	2,985.2	24,818,953	$k = Ae^{-2.985,2/T}$
Liquid smoke	413	1,297.5	10,787,415	$k = Ae^{-1.297,5/T}$

where $-E_a/R$ is the slope (m), and ln A is the point of intersection. The relationship between the rate constant and the pyrolysis temperature is obtained, as shown in Table 3.

A reaction could occur if the kinetic energy between the reactants exceeds the activation energy [36]. The use of a catalyst will provide a new pathway for chemical reaction mechanisms [37, 38], and also reduce the reaction temperature [39]. In this study, the temperature is an independent variable because, during heating, it was not possible to control the temperature in the reactor.

3.5 Physical Properties of Liquid Smoke from HHW Pyrolysis

a. Density (g/mL)

The density of liquid smoke from HHW pyrolysis was 0.879 g/mL, which is above the gasoline quality standard recommended by SNI (Indonesian National Standard) and also higher than the results of research conducted by [15] which is 0.76 g/mL. The density of gasoline fuel oil is between the density of kerosene (0.78 – 0.81 g/mL) [40]. The high density of HHW liquid smoke can be caused by the influence of organic compounds that have a chain number of more than 5 carbon atoms or more heavy molecules that are decomposed. Based on the density value of HHW liquid smoke, it is still below the density of water (1 kg/m^3). Thus, the liquid smoke from HHW pyrolysis can be recommended as a fuel to replace kerosene. The high value of the density of fuel could increase engine thirst, causing damage to the engine [41].

b. Viscosity

The viscosity of the liquid smoke from HHW was 0.9 cP which is greater than the SNI viscosity value of 0.7 cP. Viscosity is strongly influenced by density. The heavier the density of the liquid, the greater the viscosity, meaning that more particles hinder the flow of the fluid because they rub against each other [40]. The thickness of the liquid smoke from HHW pyrolysis could be caused by the greater long-chain chemical structure bonds, which turn to greater viscosity [42]. Fuel that has a high viscosity will be difficult to flow and affect the engine [43]. If the viscosity is too high (thick), the fuel will be difficult to pump into the combustion chamber and affect the quality of automation, which is difficult to occur. When viewed from the SNI of gasoline viscosity value (0.70 cP), the liquid smoke from HHW cannot be classified as gasoline but can be suggested as an alternative fuel to kerosene.

c. Boiling point

The boiling point of liquid smoke from HHW was 139.8 °C. Based on SNI 06-3506-1994, the maximum allowable final boiling point of a fuel is 205 °C [44]. The boiling point of liquid smoke from HHW effluent shows a number below the maximum limit of the relevant quality standard. Therewith, liquid smoke from HHW can be categorized in the same category as gasoline or diesel fuel.

d. Flash point

The flash point of liquid smoke from HHW pyrolysis was 28.4 °C. As a comparison, the flash point of diesel fuel is 50 °C [45]. Meanwhile, the flash point of gasoline circulating in the market is 57 °C. The flash point of HHW liquid smoke shows a number below the flash point of diesel fuel and gasoline on the market. The flash point of fuel indicates the safe limit against fire hazards during storage. The increase in the composition of short-chain hydrocarbon compounds results in a lower flash point and a higher boiling point. Conversely, if the short-chain hydrocarbon compounds produced decrease, the longer-chain hydrocarbon compounds produced will increase [46].

e. Calorific value

The calorific value of liquid smoke from HHW hydrolysis was 8.684 cal/g. The calorific value obtained cannot meet the standard calorific value of gasoline produced by Pertamina, which is at least 10.160 cal/g and a maximum of 11,000 cal/g. Fuels with low calorific values indicate that the composition of short-chain hydrocarbons is lower [46]. The calorific value of fuel is a quantity that shows the value of the heat energy produced from a combustion process per unit mass of fuel [47].

f. GC-MS analysis

The GC chromatogram analysis of the liquid smoke from HHW hydrolysis is shown in Fig. 8. The GC chromatogram shows that the liquid smoke from the HHW consists
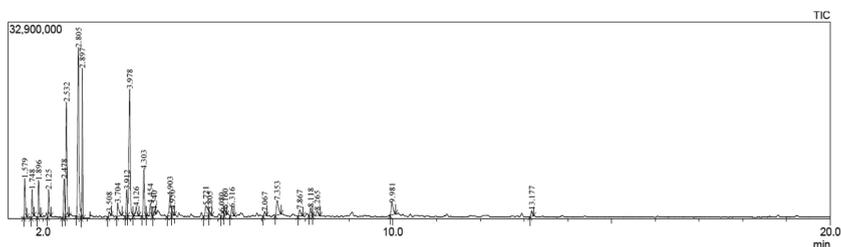
Fig. 8. GC chromatogram of the liquid smoke from HHW hydrolysis

of several compound peaks with different percentages. From MS spectra and peak area on the GC chromatograms, liquid smoke from HHW consists of 30 compounds dominated by 18 compounds derived from 3.06% acetonitrile, 2.22% propanenitrile, 2.94% 2-methyl propanenitrile, 2.37% butanenitrile, 3.57% 2-methyl butanenitrile, 11.07% 3-methyl butanenitrile, 19.27% pyrrole, 8.38% toluene, 1.87% 2-methyl pyridine, 3.0% 4,4-dimethyl-3-oxopentanenitrile, 15.65% 4-methyl pentanenitrile, 1.24% 2-methyl pyrrole, 4.78% ethyl benzene, 1.63% 1,2-dimethyl benzene, 2.98% styrene, 2.39% 2,5-dimethyl pyrrole, 2.62% phenol and 2.10% 4-methyl phenol. Liquid smoke from HHW is a very unstable mixture of chemical compounds. This can be indicated by a change in the color of the smoke from slightly clear to dark brown in a relatively short time. Therefore, for long-term storage, liquid smoke from HHW must be stabilized by a chemical stabilizer or hydrogenation immediately after collection. In addition, the produced liquid smoke requires further purification to make it suitable for use as fuel or other chemicals [48].

4 Conclusion

1. HHW is categorized as a waste that is difficult to decompose by microorganisms in water and soil and has high stability to temperature.
2. Pyrolysis of 200 g of HHW produced 12,205 mL of gas that can be burned directly with a clear yellowish flame. In addition, 50.5 mL of liquid smoke was also obtained, and after a flame test, it turned out that the liquid smoke could burn with a clear yellowish color and a red color at the ends and bottom sides.
3. The gas and liquid smoke formation during HHW pyrolysis with the addition of zeolite catalyst follow a first-order reaction with a value of $R^2 = 0.9609$ and 0.9652, respectively. This means that if the pyrolysis temperature is increased, the reaction rate of gas or liquid formation will increase.
4. The optimum temperature of HHW pyrolysis was 413 K and took place isothermally. The produced gas and liquid smoke are the main products. The yield increases along with pyrolysis temperature while the remaining charcoal in the reactor decreases.
5. The relationship between the value of the reaction rate constant for the formation of gaseous products and temperature can be expressed in the Arrhenius equation $k = Ae^{-2,985.2/T}$ with the activation energy value (E_a) of 24.818×10^3 kJ/mol, while the liquid smoke is expressed in the Arrhenius equation $k = Ae^{-1.297.5/T}$ with E_a of 10.787×10^3 kJ/mol.

6. The kinetics of the decomposition reaction of HHW is influenced by changes in mass per unit of time. The value of the exponential factor A will affect the reaction speed in the pyrolysis process, and the greater the value of A, the greater the reaction speed or vice versa.

7. The effect of using a zeolite catalyst on the pyrolysis of HHW is a reduction in the value of the activation energy. Thereby, the pyrolysis decomposition reaction will run faster, and the gas and liquid smoke will be produced more.

8. The characteristics of physical properties of liquid smoke from HHW are not yet suitable for use as fuel oil. But a method for further purification is needed so that the results can be used as fuel oil or other chemicals.

9. Liquid smoke resulting from HHW pyrolysis is a mixture of 30 chemical compounds, and some of them are unstable compounds. This was indicated by a change in the color of the liquid smoke from pale yellow to dark brown in a relatively short time. Therefore, liquid smoke from HHW must be stabilized with chemical stabilizers for long-term storage.

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