The role of alkali metal and alkaline metal earth in natural zeolite on combustion of *Albizia Falcataria* sawdust

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**Abstract**

The combustion process of *Albizia falcataria* (AF) sawdust with the addition of natural zeolite (NZ) was observed experimentally using PT 1600 LINSEIS Simultaneous thermal analyzer (STA). The results showed that alkali metal and alkaline metal earth in NZ play an essential role in the process of decomposing the Hemicellulose AF molecule. The results of the molecular analysis show that the chemical balance of the mixture determines the combustion temperature. Excess NZ becomes a thermal burden which slows down the combustion reaction because heat does not sufficiently activate alkali metal and alkaline metal earth in NZ. In a small amount, NZ is less involved in the AF decomposition process. It shows that a mixture of AF and NZ can increase combustion kinetic in the right mix. Addition of 15–20% of NZ decreases the ignition temperature within faster burning rate. Activated alkali metal and alkaline metal earth decompose hemicellulose faster so that they burn completely in minimizing pollutant and maximizing LHV. Greater NZ completes the decomposition much earlier so that at the resting time of the process NZ slightly absorbs heat sinking LHV. The drastic reduction of Ca due to NZ make the fuel is suitable for boiler because Ca is responsible for agglomeration and corrosion.

**Keywords** *Albizia falcataria* · Ignition temperature · Combustion kinetic · Zeolite catalyst

**Introduction**

*Albizia falcataria* (AF) is one of the most widely planted trees in Indonesia because it is quickly harvested at the age of 5–7 years [1, 2]. AF is processed into semi-finished material and then made into pallets, wooden crates. AF sawmills produce large amounts of sawdust waste. The waste of wood sawdust from AF processing in Indonesia will continue to grow because of the short harvest age of the plant and thus become an environmental problem. The research on application of sawdust biomass fuel and the effort to improve boiler efficiency have been done in the last decade [3–5]. The effect of the biomass fuel on the boiler corrosion has also been studied [6]. To get better understanding on the combustion characteristics of sawdust, this paper provides a scientific discussion about the use NZ to improve sawdust as fuel.

Biomass combustion reaction occurs in many steps [7], namely: heating, pyrolysis (volatiles), pre-combustion reaction, primary gas-phase combustion, secondary combustion, effluent stack gas. Some previous researchers conducted pyrolysis of biomass with additional zeolites capable of producing more gas. The researcher [8] used H-ZSM-5 for pyrolysis of pine and cypress whose results explained that the function of catalyst acid was able to increase aromatic concentration. Whereas [9, 10] made pyrolysis of the various types of wood chips by adding ZSM5 discontinuously and additional stainless steel ball bearings to make pyrolysis effective. Stainless steel ball bearings inhibit pyrolysis vapor and expand the hot surface during thermal cracking. The result shows that the percentage of H, CH4, C2H4, C3H6 gas increases with the addition of zeolite. The study was conducted [11] on the pyrolysis of beech wood (*Fagus sylvatica L.*) with five types of zeolites (SN-27, MSN-15, MSM-15, H-ZSM-5-28, and H-ZSM-5-80) to determine its effect on lignocellulose. The results showed that zeolite acid
catalyst increases the amount of Furfural and Levoglucosan. H-ZSM-5 with lower alumina content is the most effective in improving furfural because of better molecular diffusion through pores. Overall, the above research shows that acid catalyst can improve the final yield of lignocelluloses thermal degradation in pyrolysis. This suggests that additional zeolite must also improve reaction that makes combustion characteristics better. Therefore, the present study aims to uncover the role of natural zeolite (NZ) on combustion process of AF.

Analysis of biomass combustion using thermogravimetry is important to know the thermochemical conversion of combustible solid waste. The burning process of mixture of pine sawdust and coal was analyzed using a thermogravimetric analyzer [12]. The result shows that pine sawdust is decomposed at the beginning then followed by coal and pine charcoal which are burned together at the same time. The study [13] mixed cellulose, xylan and lignin with coal and then analyzed its burning process using a thermogravimetric analyzer. The result is that lignocellulose can increase the rate of devolatization, reduce flame temperature, and accelerate charcoal combustion. The thermogravimetric analysis of additional catalysts (KOH, NaOH, KCl, CuCl2/KCl, and CaCl2/CaO) in burning of Municipal Solid Waste found the flame temperature reduced. At a temperature of 200 °C Municipal Solid Waste with the addition of KOH catalyst, CuCl2/KCl, and CaCl2/CaO have been degraded 5% while others are still below it [14]. Research on thermogravimetric analysis by burning rice and wheat straw with additional catalysts (NiO, CuO, CaO, and MgO) was carried out [15]. The result is that the MgO catalyst at 200 °C has burned more than 5% of rice straw while the others are lower. It means that the catalyst can increase the rate of the volatility of biomass. The same study was done by [16] investigating a thermogravimetric analyzer. In this study, combustion of peanut shells, rice husks and wheat straw added with organic calcium compounds were analyzed. The result shows that the index of peanut skin and wheat straw burning performance increase while the rice husk decreases. It is due to the lowest C and H content of rice husks compared to the others so the percentage of catalysts must be different.

Scientific data about the use of natural catalysts is very limited, so more researches are needed. This study describes the combustion of sawdust AF with the addition of Natural Zeolite (NZ) using a thermogravimetric analyzer as a reactor. A good understanding of the decomposition of biomass during the thermochemical conversion process is very important for the efficiency of the process and its technology utilization [17].

Many NZ are available in Indonesia, making them easy to obtain and cheap. NZ mines are found in the southern part of Java, starting from Malang—East Java and Cikembar, Cipatujah, Bayah—West Java. So far NZ has been applied only for dehydration of ethanol and transesterification of crude palm oil [18, 19]. The study of improving biomass combustion using NZ is rarely found. Therefore this study provides a discussion of NZ applied to improve biomass combustion characteristics. Since NZ contains a lot of minerals which easily disintegrate and become active when they get enough thermal energy, then the discussion emphasizes the role of minerals in NZ for molecular decomposition of hemicellulose.

### Materials and Methods

#### 2.1 Materials and properties analysis

The AF used in this study was taken from sawmills in Lumajang, East Java, Indonesia. AF was dried in the sun and crushed to a size of 100 mesh. The ultimate analysis and proximate analysis were carried out using LECO CHN-2000 and S-632 equipment. The AF composition was analyzed to determine the percentage of cellulose, hemicellulose, and lignin content as shown in Table 1a. It is seen that AF consists mostly of volatile matter but also contains very little sulfur and nitrogen as well. Three main elements are forming AF, detected carbon which is almost the same as oxygen and hydrogen; there is a small part. The structure of

| Ultimate analysis (dry basis, wt.%) |
|-------------------------------------|
| Carbon 45.53                        |
| Hydrogen 6.49                       |
| Nitrogen 0.31                       |
| Sulfur 0.1                          |
| Oxygen 46.13                        |

| Proximate analysis (dry basis, wt%) |
|-------------------------------------|
| Moisture 7.2                       |
| Volatile matter 74.9               |
| Fixed Carbon 16.46                 |
| Ash 1.44                           |
| Gross Caloric Value (J/g) 18.229   |

| Compositional analysis (%)          |
|-------------------------------------|
| Cellulose 41.88                     |
| Hemicellulose 14.55                 |
| Lignin 24.78                        |

| Chemical formula                    |
|-------------------------------------|
| Al 8.16 Si 27.84 O 97.52 Na 4.80 C 1.88 K 0.28 Mg 0.09 |

| Crystal system, Density (g/cm³)    |
|-------------------------------------|
| Monoclinic 2.17                     |
the composition of the AF-forming compound such as lignocellulose generally consists of cellulose at most followed by lignin and hemicellulose.

NZ was obtained from mining in Malang, East Java, Indonesia, cleaned with distilled water and it was crushed into the same size as AF sawdust. NZ is characterized using XRD to determine the mineral content and crystal structure. The XRD analysis results are listed in Table 1b. NZ chemical formula is dominated by Oxygen and Si and Al which are the main elements that form the tetrahedral structure. Also detected are alkali metal and alkaline metal earth elements as ions counterbalancing Al-O bonds to create a tetrahedral structure.

**Thermogravimetric analysis**

Sawdust combustion from AF with a mixture of NZ was analyzed using PT 1600 LINSEIS Simultaneous thermal analyzer (STA) as a reactor shown in Fig. 1. This technique was also applied in [20, 21]. The sample is placed in the sample pan while the reference pan is usually empty, tested at three different initial weight AF; AF1 = 10 mg, AF2 = 15 mg and AF3 = 20 mg. The addition of NZ to the test sample are 15% and 25% of the initial weight AF, namely, AF1 + NZ (15% xAF1) and so on. Samples at three different weights were burned from room temperature to 900 °C with a heating rate of 80 °C/min and air flow of 5 Ls/min [22], then held for 10 min at 900 °C for a complete combustion. Heating using a heating element (heater) was controlled by the CPU. Airflow is generated using a compressor that is regulated using valves and flow indicator meters. Temperature is measured using a thermocouple attached to the sample pan and reference. Weight changes are weighed by the scale connected with the sample pan and reference. These weight changes are then recorded as relative weight loss (TG) = \( \frac{m}{m_0}(\%) \), where \( m \) is instantaneous mass of the sample and \( m_0 \) is initial mass of the sample. The rate of relative weight loss was estimate from time derivative of TG as \( \frac{d(TG)}{dt} \).

**2.3 Catalytic Combustion Analysis**

The use of NZ catalysts on combustion sawdust AF can reduce the ignition temperature so that it can achieve lower activation energy. The formula of the Conversion Degree of Combustible (CDC) is used to calculate the temperature around wood powder [23, 24] based on data from TG defined as in Eq. 1.

\[
\text{CDC} = \frac{m_o - m}{m_o - m_\infty} \times 100\% \quad \text{(1)}
\]

where \( m_\infty \) is the final mass of the sample obtained from STA. Besides the temperature around sawdust AF powder, CDC is also used to estimate the effect of NZ on combustion of AF. There are two parameters used to estimate the effect of NZ on AF. One is the temperature when CDC of the sample reaches 5% and the other one is the CDC when temperature reaches 900 °C [14].

![Schematic principle of STA](image)

**Fig. 1  Schematic principle of STA**
Chemical analysis of ash

The ash from AF combustion was analyzed using SEM (FEI INSPECT type S50). This instrument was also equipped with EDAX which is useful to analyze the content of the ash surface structure. The content of the ash is presented in Table 2.

Result and discussion

TG and DTG analysis

Biomass combustion occurs in two main stages: de-volatilization stage and oxidation stage [21]. Likewise, burning a mixture of AF and NZ also can be divided into two phases. The first is volatile combustion, and the second is fixed char combustion.

Data obtained from the reactor are time, temperature, relative weight loss (m/mo) and derivatives of relative weight loss. In burning AF, the catalyst works more effectively in combustion when lowering the flame temperature. The effect of adding catalysts at combustion AF is high when a sudden increase in CDC occurs at a lower ignition temperature. In this paper, the impact of adding NZ percentages and initial weight differences are discussed.

Figure 2 describes TG (Relative weight loss, right side) and DTG (Derivative weight loss, left side) at combustion AF, AF + 15% NZ, and AF + 25% NZ at various AF weights. The lignocellulose content in AF in Table 1a refers to [25] according to the general biomass content. Solid line represents TG and dashed line represents DTG.

Figure 2a explains about TG-DTG at the initial weight of 10 mg. Combustion AF1 without NZ have faster weight degradation. It is because NZ is a thermal burden as the heat from combustion is small so it cannot activate minerals in NZ. Consequently, ash containing SiO2 and Al2O3 is burned so that there is still residue in the pan [26]. Adding NZ to AF causes the percentage of ash to increase because Si and Al elements arrange the compounds. At the end of the process, the percentage of TG of combustion ash AF2 with the addition of NZ is higher than that without additives.

In Fig. 2b for sample weight of 15 mg, the rate of weight reduction in combustion AF2 + 15% NZ is slower than AF2 + 25% NZ. That is because hemicellulose, cellulose, and lignin decompose together due to the NZ catalysis effect. It is seen that the blue line (TG) is steeper than the red line (TG), but when the temperature approaches 600 °C, the blue line (DTG) rises almost to zero indicating that hemicellulose, cellulose, and lignin burnout. At the end of the combustion, only NZ is left so that the ash percentage is 25% more at AF2 + 25%NZ; the blue line (TG) is more than the red line (TG). The residue of using NZ is almost half of that at 10 mg sample (Fig. 2a) indicating that NZ work better.

In Fig. 2c NZ has started working and controlling combustion. At 20 mg AF combustion, NZ has enough heat to be active even though AF + 25% NZ has not been burned better than AF + 15% NZ. It can be seen that in burning lignin (fixed carbon) at temperatures above 600 °C, the DTG blue line is close to zero followed by red and black lines. It means that at the fix carbon combustion stage, NZ could break down lignin faster than without NZ. The percentage of ash which is around 20% equals to the percentage of NZ in the AF. This shows that the AF burned completely indicating that the burning of AF with NZ is almost free of pollutant emission. This result is supported by Table 2 which presents that the ash mainly contains Si and O and then Al, these are the main elements in NZ while C was completely disappear.

Table 2 Chemical analysis results of ash

| Element     | C   | O   | Na  | Mg  | Al  | Si  | P   | S   | K   | Ca  | Fe  |
|-------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| AF          | 10.5| 35.2| 2.8 | 2.0 | 1.8 | 14.5| 1.3 | 0.3 | 2.9 | 24.5| 3.8 |
| AF + 15%NZ  | 16.5| 39.3| 0.4 | 0.2 | 4.3 | 29.4| –   | –   | 3.9 | 4.6 | 1.0 |
| AF + 25%NZ  | –   | 45.2| 1.5 | 0.8 | 7.2 | 37.0| –   | –   | 4.3 | 1.8 | 1.8 |

Influence of NZ on catalytic combustion

Figure 3 shows the CDC which explains the influence of NZ on combustion AF. CDC index compares the character of combustion by measuring temperature when the weight loss reaches 5% [14]. The higher the CDC temperature means the effect of the catalyst does not affect the combustion of AF. Whereas on the contrary, it is true that the catalyst has a significant effect on combustion AF. For an initial weight of 10 mg (Fig. 3) the addition of NZ does not increase the rate of weight degradation. AF without NZ is at a lower ignition temperature, followed by additions of 15% and 25% NZ at higher temperatures. CDC AF temperature occurs at a temperature of 253 °C, while an additional 15% NZ makes the CDC AF occur at a temperature of 275 °C.

Furthermore, the addition of NZ to 25% makes CDC AF occur at 309 °C. That happened because the weight of AF burned too little and the heat produced by combustion was absorbed by NZ so that it was not enough to make minerals in NZ become active to help combustion in other words NZ only became a thermal burden. As a result, AF + NZ requires higher amount of heat to burn.

In Fig. 4 with the addition of NZ to the initial weight of 15 mg in combustion, AF shows the difference in temperature of the CDC. The addition of 15% of NZ causes CDC AF
Fig. 2  DTG –TG AF vs. Temperature. a AF1 = 10 mg; b AF2 = 15 mg; c AF3 = 20 mg

Fig. 3  CDC AF 10 mg vs. Temperature

Fig. 4  CDC AF 15 mg vs. Temperature
at the lowest temperature (251, 256, and 259 °C). The same thing happens at the initial weight of 20 mg, which is shown in Fig. 5. Addition of 15% of NZ gives the lowest CDC temperature. It shows that the amount of heat is only enough to activate NZ to 15%. Above that percentage, NZ became a thermal load.

AF + NZ sawdust combustion with an initial weight of 20 mg has the lowest CDC temperature (234, 239, and 248 °C) (Fig. 5). That is, the heat from the fuel has been able to activate NZ to 25% even though in this amount NZ is not as active as 15%.

In Figs. 4 and 5 there is a red line which is AF + 15% NZ increasing precedently at 214 °C and 208 °C. It is due to the composition of the right mixture in AF + NZ so that NZ can break AF bonds thoroughly. AF sawdust 14.55% of its content is hemicellulose which will be decomposed by heat energy together with the change in NZ structure. Hemicellulose consists of several polymers, e.g., xylan. The Explanation of polymer breakdown in this discussion use xylan for the main polymer that forms hemicellulose [27, 28]. Hemicellulose consists of branched polymers which are more easily broken down than cellulose. One of the essential branches is xylan, so if there is an initial decomposition of combustion, it can be predicted that it is xylan [29, 30]. Data obtained after processing proves that the addition of NZ can reduce the decomposition temperature of lignocellulose AF as the results of research conducted by [31].

During the combustion process, some heat energy makes NZ release Na+, Mg2+, K+, and Ca2+ (Alkali metal and Alkaline metal earth) as counterbalancing ions (Fig. 6). Alkali metal and alkaline metals earth are more energetic atoms in tearing the connecting oxygen between the monomers of hemicellulose in AF so that the hemicellulose will be broken into shorter bonds (Fig. 7).

It can be seen in the NZ structure in Fig. 6 that alkali metal and alkaline metal earth in NZ, namely Na, Mg, K, and Ca act as counterbalancing of Al–O and Si–O to form tetrahedral structures in NZ. When NZ is heated the structure changes to Bronsted acid by releasing Na+, Mg2+, K+, and Ca2+. These ions will make the partial negative oxygen connecting the hemicellulose polymer bonds experience a continuous attracting force which eventually dislodges. As a result, the polymer will turn into a monomer (Fig. 6). It results in 15 mg of AF burning faster at lower temperatures because of shorter bonds which are more flammable. The results of this study are in accordance with the study in [32, 33] which explained that alkali metal and alkaline metal earth are useful catalysts for pyrolysis, combustion, and gasification.

Larger alkali metal and alkaline metal earth molecules disrupt oxygen which binds polymer carbon so that the
polymeric bonds will be broken. For small initial fuel mass (10 mg), the addition of NZ causes AF to burn more slowly at higher temperatures because the heat energy from combustion of AF is not enough to activate zeolite to release alkali metal and alkaline metal earth ions so that it only functions as a thermal load. It is different for the initial weight of 15 and 20 mg, especially in 15% NZ which can break the hemicellulose chain faster. This mechanism is caused by the volatility of AF before it burns. At the same time, alkali metal and alkaline metal earth in NZ have got enough heat energy to be activated and released so that they attract oxygen polymer chains (red lines in Figs. 4 and 5).

For the addition of 25% NZ there is a slowdown in flame temperature because heat is absorbed more by NZ than AF so that the process of AF volatility it is slower (blue lines in Figs. 4 and 5).

Figure 7 illustrates that air flow and heat make alkali metal and alkaline metal earth, e.g., black Na which is released from the NZ. Na attracts red oxygen from the polymer bonds so that the broken bonds are shorter a and b. Na—O bonds is temporary, making Na (or other balancing ions) separated into free atoms to break other bonds (a) and (b) with the same mechanism so that they become short chain bonds that are more flammable.

Na electron in the outer shell induces O electrons because the C–O–C bond is weakened due to heat. As a result, the C–O–C bond is broken so that the polymer turns into a monomer. The electron of Na (or another balancing ion) induces and attracts O atoms that weaken O–C bond due to increased heat, accelerating the process of decomposition of Lignocellulose. Finally the combustion temperature decreases.

**Influence of NZ on LHV**

Table 3 shows the heat released (LHV) by the burning of AF at various NZ. The change of mean heat released caused addition of NZ is presented in Fig. 8. It can be seen that at small sample (10 mg) the addition of NZ increases LHV since the decomposition rate takes place along the process as shown in Fig. 2a. However at larger samples (15 mg and 20 mg) increasing NZ reduces slightly LHV. This is due to the fact that decomposition of AF is completed much earlier at lower temperature (see Fig. 2b and 2c) so that as the time proceeds the NZ does not work to decompose cellulose but

| No | AF1 = 10 mg | AF2 = 15 mg | AF3 = 20 mg |
|----|-------------|-------------|-------------|
| 1  | 0%          | 15%NZ       | 25%NZ       |
| 2  | 56.13       | 21.14       | 33.75       |
| 3  | 72.85       | 51.72       | 69.3        |
| 4  | 0%          | 15%NZ       | 25%NZ       |
| 5  | 6365.16     | 6873.44     | 7690.46     |
| 6  | 6820.55     | 6845.2      | 5855.69     |
| 7  | 0%          | 15%NZ       | 25%NZ       |
| 8  | 54.52       | 57.51       | 34.84       |
| 9  | 5430.45     | 5454.2      | 5376.67     |

Table 3 Heat released by AF at various NZ
3.4 Influence of NZ on ash chemical composition

As presented in Table 2 the chemical compositions in ash (Wt%) of AF change over addition of NZ. All minerals which are not component of NZ reduce. The Ca, which is responsible for agglomeration and corrosion [6] reduces drastically. The Si which is increased largely together with O due to the NZ addition which is composed mainly by silica and alumina. This result suggests that addition of NZ makes AF suitable for boiler fuel.

Conclusions

The study of AF combustion with the addition of NZ has been shown to reduce flame temperature. Proper addition of NZ can accelerate the devolatization of AF which eventually speeds up the combustion process. The role of alkali metal and alkaline metal earth in NZ can break the lignocellulose polymer chain so that it decomposes quickly into a monomer. The monomer then decomposes into several flammable gases. However, the ratio of AF and NZ affects the combustion process. The addition of NZ to the small number of AF burning makes NZ to become a thermal load. When the weight of AF is high, the addition of the right percentage of AF accelerates the process of breaking C–O–C bonds. Addition of 15–20% of NZ decreases the ignition temperature within faster burning rate. Activated alkali metal and alkaline metal earth decompose hemicellulose faster so that they burn completely in minimizing pollutant and maximizing LHV. At greater number of NZ, the decomposition is completed much earlier so that as the time proceeds NZ does not work to decompose cellulose but slightly absorbs heat sinking LHV. The Ca that is responsible for agglomeration and corrosion decreases to a minimum number due to NZ. This makes the AF added with NZ catalyst become suitable for boiler.

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Data availability

The data used to support the findings of this study are available from the corresponding author upon request.

Compliance with ethical standards

Conflict of interest

The authors declare that they have no conflicts of interest.

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