Mathematical model of enzymatic reactions of coconut coir substrate treated by alkaline and ionic liquid into the reducing sugars using power series

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Abstract. The research purposed at formulating the kinetics model of reducing sugars liberated from coconut coir dust treated with alkaline, ionic liquid, alkaline followed by ionic liquid employing a power series. The method was successful to predict the sugar concentration released from the substrate in a biocatalytic reaction employing cellulose, and a mixture of cellulase+xylanase at any time. The yield of sugar released enzymatically was modeled as an equation in power series in which the results were comparable to those of the experimental data. The biggest constant rate obtained was of 0.0200000 h⁻¹ for an application of a mixture of enzymes on the conversion of NaOH-treated substrate into the reducing sugars. Generally, the trend of the modelled graph was close to the experiment but the error was quietly wide in the initial times since the conversion rate was very high. The lines were an agreement when the time was above 8h for all pre-treatments (native-, 1%NaOH-, IL-, and 1%NaOH+IL substrates). It was indicative the power series can be used to solve a differential equation generated from a rate equation.

1. Introduction
Currently, the scientists have been interested in the lignocellulosic materials because they can be converted into sugars and biofuels and are available abundantly on earth especially, in the tropical countries [1][2]. The study showed that prior to conversion into more valuable materials, lignocellulose should be treated that purposed to decline the lignin content and to modify the cellulosic structure [3]. The conversion evaluation of lignocellulose treated by chemical or physical pre-treatment has been reported by many investigators [4][5][6].

The kinetic study is an important aspect of the chemical reaction in which it can be predicted an amount of the product in the extended range of time. The poplar wood has been liquefied into the reducing sugar by using the wet oxidation pre-treatment and the yield of product was formulated in the form of the mathematical model. The equation obtained was successful in predicting the reducing sugar yielded [7]. The transformation of sugarcane bagasse into levulinic acid enhanced by acid was reported by authors [8]. The kinetic formulas of enzymatic hydrolysis were written in some equations depending on the temperatures and sulfuric acid concentrations employed.

The kitchen waste was converted to the bioethanol by using acid and hot water treatments whereby the kinetics of the biochemical reaction was modeled in the simple form [9]. Authors [10] reviewed the kinetic model of enzymatic hydrolysis of many substrates. The results found the simultaneous hydrolysis and yeasting in continuous mode gave the biggest yield. The mathematical model of anaerobic digestion in the fermenter in producing biogas was studied previously [11].
discovered as described that through a kinetic mathematical model, the biochemical process inside the reactor would be understood deeply and the prediction of product yield was well known.

This study reported the simple kinetic model of bioconversion of coconut coir dust treated by ionic liquid 1-ethyl-3-methylimidazolium dimethyl phosphate ([mmim][dmp]) and alkaline using the power series. The IL employed in this study was synthesized in the author's Lab and the result obtained through the kinetic model was compared to that of the experimental data. The kinetic model was formulated after the reducing sugars were obtained through enzymatic hydrolysis with a catalyst loading was constant.

2. Materials and Methods

2.1. Chemicals

The coconut coir was obtained from a copra farmer in Minahasa Regency, North Sulawesi Indonesia. The biomass was milled until the size of 100-120 mesh employed an apparatus the Retsch GmbH Rheinische Strade 362/2781, Haan Germany. The ionic liquid [MMIM][DMP], cellulase, and xylanase were purchased from Sigma-Aldrich, St. Louis, MO, USA. The procedures for all steps started with pretreatments and solids characterization were published previously [3][4][6].

2.2. Enzymatic reaction

The enzymatic reaction to convert substrate into sugars was conducted by adapting a previous report (Zhang and Bao, 2017). The sodium acetate set at pH 3, 30 mL, and 0.1M was employed as a suspension and was mixed with a 1 gram of (120 mesh) inside Ellen-Meyer flask. The temperature of the reactor was kept at 60 °C by being stirred for 48h. The sugars started liberating from the substrate when 0.2ml cellulase or cellulase+xylanase was added to the reactor and if needed the citric acid was also added to maintain pH at 3. The sample as much as 0.4ml was taken periodically from the mixture and dissolved with 1.8 ml of pure water as well as 3 ml DNS. The concentration of sugars obtained was measured by employing a DNS method [12].

2.3. The enzymatic reaction model

The mathematical model, which was proposed, followed and modified from an established equation as proposed previously by authors [7][13]. The first, lignocellulose converted into the reducing sugars could be modeled as

\[ \text{coconut coir} \rightarrow \text{sugars} \] (1)

The rate formulation was defined as follows:

\[ -\frac{dm}{dt} = km_S^n \] (2)

Where, \( r \) is reaction rate (g/s); \( k \) is the reaction constant (unit depends on the reaction order), \( m_S \) is the substrate mass (g) and \( n \) is the reaction order. In this study, since the catalyst loading is constant, the influence of cellulase was absorbed in \( k \) parameter and the temperature reaction was kept in 60°C and the variable \( r \) was the decrease rate of substrate mass with respect to time or \( \frac{dm_S}{dt} \).

To solve Eq. (2) was assumed that the order of enzymatic hydrolysis was an order one, \( n=1 \) as proposed by investigators [13][14]. The mass of the initial substrate and reducing sugar were assigned by \( m_0 \) and \( m_{RS} \). The substrate mass could be written as \( m_S = m_0 - m_{RS} \) so the differential equation of Eq. (2) was modified as:

\[ \frac{dm_{RS}}{dt} + km_{RS} = km_0 \] (3)

The solution of Eq. (3) was solved in two ways by the analytical method and mathematical series. The solutions of Eq. (3) were two parts, a homogeneous solution in which the right side was zero and a particular solution that was easy to obtain and will be elaborated widely in results and discussion below. By employing a variable separation, the mass of reducing sugar \( (m_{RS}) \) can be written as

\[ m_{RS} = m_0 \left( 1 - e^{-kt} \right) \] (4)
The solution of Eq. (3) could be expanded in form of mathematical series by assuming the \( k \) is much less than unity as follows:

\[
m_{RS} = \sum_{n=0}^{N} a_n t^n = a_0 + a_1 t + a_2 t^2 + a_3 t^3 + a_4 t^4 + \cdots \tag{5}
\]

The advantage of using the power series method is that we do not need to solve the differential equation analytically which is more difficult by employing the conventional way. The using of power series technique only needs the ability to equate the parameters which attach to the similar order and then quantities are made as function \( k \) and \( a_n \).

The first work was to find the solution of the homogeneous part by equalizing zero on the right side in Eq. (3). The Eq. (5) was derived with respect to time obtained Eq. (6) as follows:

\[
\frac{dm_{RS}}{dt} = n \sum_{n=1}^{N} a_n t^{n-1} = a_1 + 2a_2 t + 3a_3 t^2 + 4a_4 t^3 + \cdots \tag{6}
\]

After Eq. (6) is substituted into Eq. (3), the new expression will be obtained the following:

\[
a_1 + 2a_2 t + 3a_3 t^2 + 4a_4 t^3 + \cdots = -k(a_0 + a_1 t + a_2 t^2 + a_3 t^3 + a_4 t^4 + \cdots) \tag{7}
\]

Now, parameters \( a_1, a_2, a_3, a_4, \ldots, a_n \) are attributed with quantities \( k \) and \( a_n \) by equalizing the terms both sides which have the same power and the results are shown below.

\[
a_1 = -\frac{ka_0}{k^2}, \quad a_2 = -\frac{a_0}{k^2}, \quad a_3 = -\frac{1x2x3}{k^4}a_0, \quad a_4 = \frac{1x2x3}{k^4}a_0, \quad a_5 = -\frac{1x2x3x4x5}{k^6}a_0, \quad a_6 = \frac{1x2x3x4x5x6}{k^6}a_0, \quad \ldots, \quad a_m, a_n
\]

The eq. (8) shows that the parameter is split into two parts, even and odd series as presented in Eq. (9) and (10).

\[
a_n = \sum_{n=0}^{N} \frac{k^{2n}a_0}{(2n)!} \tag{9}
\]

\[
a_m = -\sum_{m=0}^{N} \frac{k^{2m+1}a_0}{(2m+1)!} \tag{10}
\]

The particular part is found through supposing that \( m_{RS} = \text{constant} \) whose value is \( a_n \) after being substituted for Eq. (3). The general solution obtained is an addition to the homogeneous- and particular parts are written as:
The Eq. (11) can be expanded in the form of

\[ m_{RS} = m_o + \sum_{n=0}^{N} \frac{(kt)^{2n}a_o}{(2n)!} + \sum_{m=0}^{N} \frac{(kt)^{2m+1}a_o}{(2m+1)!} \]  \tag{11}

The experiment was observed that at \( t=0 \), \( m_{RS}=0 \), the \( a_o \) found is a negative of the initial mass of the substrate, so the Eq. (12) could be written as

\[ m_{RS} = m_o \left\{ 1 - \left[ 1 - (kt) + \frac{1}{2!(kt)^2} - \frac{1}{3!(kt)^3} + \frac{1}{4!(kt)^4} - \frac{1}{5!(kt)^5} + \cdots \right] \right\} \]  \tag{12}

\[ \text{Yield} = \left\{ \left( (kt) - \frac{1}{2!(kt)^2} + \frac{1}{3!(kt)^3} - \frac{1}{4!(kt)^4} + \frac{1}{5!(kt)^5} + \cdots \right) \right\} \]  \tag{13}

The Eq. (13) derived from the power series is actually similar to the Eq. (4) that is obtained by solving the differential equation of Eq. (3) by an analytical method [15][16]. The fittings the data from the experiment and model were carried out to obtain the rate constant \( k \) using the solver software that is available in an excel program and would be elaborated the following.

Table 1. The comparative yields of sugar released from treated substrates (in g sugar/g substrate) with experiment and kinetic model data vs time course for employing the cellulase.

| t(h) | No Pretreatment | 1%NaOH- | 4%NaOH- | IL- | 1%NaOH+IL- |
|------|----------------|---------|---------|-----|-------------|
|      | Exp. Yield     | Model Yield | Exp. Yield | Model Yield | Exp. Yield | Model Yield | Exp. Yield | Model Yield | Exp. Yield | Model Yield |
| 0    | 0.0000         | 0.0000   | 0.0000   | 0.0000   | 0.0200     | 0.0000     | 0.0000     | 0.0000     | 0.0000     | 0.0000 |
| 1    | 0.0500         | 0.0030   | 0.1100   | 0.0064   | 0.0900     | 0.0056     | 0.0700     | 0.0047     | 0.1099     | 0.0074 |
| 2    | 0.0600         | 0.0060   | 0.1100   | 0.0128   | 0.0700     | 0.0112     | 0.0700     | 0.0093     | 0.1394     | 0.0148 |
| 4    | 0.0500         | 0.0119   | 0.1200   | 0.0254   | 0.0900     | 0.0224     | 0.0900     | 0.0185     | 0.1361     | 0.0293 |
| 6    | 0.0600         | 0.0178   | 0.1400   | 0.0379   | 0.1200     | 0.0334     | 0.1000     | 0.0276     | 0.1485     | 0.0436 |
| 8    | 0.0600         | 0.0237   | 0.1500   | 0.0501   | 0.1300     | 0.0442     | 0.1100     | 0.0367     | 0.1498     | 0.0577 |
| 10   | 0.0700         | 0.0295   | 0.1500   | 0.0623   | 0.1300     | 0.0550     | 0.1100     | 0.0456     | 0.1675     | 0.0717 |
| 12   | 0.0700         | 0.0354   | 0.1500   | 0.0743   | 0.1400     | 0.0656     | 0.1200     | 0.0545     | 0.1661     | 0.0854 |
| 18   | 0.0800         | 0.0526   | 0.1600   | 0.1093   | 0.1400     | 0.0968     | 0.1200     | 0.0806     | 0.1866     | 0.1253 |
| 24   | 0.0800         | 0.0695   | 0.1600   | 0.1430   | 0.1500     | 0.1269     | 0.1200     | 0.1060     | 0.2065     | 0.1634 |
| 30   | 0.0900         | 0.0860   | 0.1800   | 0.1755   | 0.1600     | 0.1560     | 0.1400     | 0.1307     | 0.2041     | 0.1999 |
| 36   | 0.0900         | 0.1023   | 0.1900   | 0.2067   | 0.1700     | 0.1842     | 0.1500     | 0.1547     | 0.2162     | 0.2348 |
| 42   | 0.1000         | 0.1184   | 0.1900   | 0.2367   | 0.1700     | 0.2114     | 0.1400     | 0.1781     | 0.2068     | 0.2682 |
| 48   | 0.1000         | 0.1341   | 0.1800   | 0.2656   | 0.1600     | 0.2377     | 0.1400     | 0.2008     | 0.2001     | 0.3002 |

3. Results and Discussion
In the present work elaborated the yield of the reaction kinetic model of the substrate treated by an ionic liquid, alkaline and, alkaline followed by ionic liquid converting into the reducing sugars after conducting a fitting process and compared to the experimental data which are presented in Table 1 and
2. Fig. 1 shows the yield model of reducing sugar liberated from treated substrates (coconut coir) using a single enzyme, cellulase. If compared to the experimental yield that the error of the initial reaction was quite big since the sugar rate released from the substrate for pre-treatment was relatively high [17][18]. The assumption that the reaction for enzymatic hydrolysis is the first order is necessary to be evaluated for this case. As time increases, however, the experimental- and kinetic model yields tend to high accuracy that they are coming to an agreement.

The experimental data of sugar yields (in g sugar/g substrate) released from solids at 1h for original-, 1%NaOH-, 4%NaOH, IL, and 1%NaOH+IL substrates are 0.0500, 0.1100, 0.1000, 0.0700, and 0.10991, while the model yields are 0.003, 0.0064, 0.0056, 0.0047, and 0.0074, respectively. When time course is approaching 12h, the experimental and model yields decrease their differences. The experimental and model yields liberated from native coconut coir and substrates treated by 1%NaOH-, 4%NaOH, IL and 1%NaOH+IL at 12h are 0.0700; 0.0354, 0.1500; 0.0743, 0.1400; 0.0656, 0.1200; 0.0545, and 0.16613; 0.0854, respectively. The final time course the two yields resulted to close figures as follows: 0.1000; 0.1341, 0.1800; 0.2656, 0.1600; 0.2377, 0.1400; 0.2008, and 0.2001; 0.3002, respectively.

The data shows that the combination method, NaOH+IL pre-treatment gives the biggest yields of the sugar obtained from both an experimental and the model hydrolysis that was comparable to previous reports by the other authors.

![Figure 1](image_url)

**Figure 1.** The kinetic model of reducing sugars yields released from substrates with respect to the time course employing a single cellulose

The second place of sugar yield found is the NaOH pre-treatment and then followed by IL technique and the least is the original substrate. It was found that NaOH pre-treatment could increase significantly the sugar released from substrate since lignin and hemicellulose dissolved into alkaline and the porosity of the substrate increased [19]. When alkaline concentration inclined to 4%, the sugars liberated decreased slightly. The increase of alkaline concentration started liquefying cellulose and hemicellulose as the source of sugar. It meant that the more alkaline added can be decreasing the yield of sugar obtained since cellulose is decomposed into the water-soluble substances.

Table 2 shows the yields of sugar (experiment and model) liberated from coconut coir treated by 1%NaOH-, 4%NaOH, IL, and 1%NaOH+IL versus time course using a mixture of cellulase+xylanase.
Meanwhile, the sugar yield of the kinetic model which is fit from experimental data is presented in Fig. 2. Generally, the behaviour of the kinetic model of enzymatic hydrolysis using mixture enzymes as shown in the figure is similar to that of employing single cellulase as described previously. The real difference is that the yield of sugar freed enzymatically from substrate treated all methods and employing mixture enzyme is higher than that of applying a single enzyme (cellulase). The significant increase occurred since the synergy of cellulase trims the 1,4-β-connections into glucose while xylanase broke down the hemicellulose rings becoming the xylose monomers [20]. The sugar yield of a model for an original solid using mixture of enzymes at 48h is of 0.2066 that is higher than that of cellulase recorded at 0.1800 g sugar/g substrate. The sugar from the model and hydrolyzed of 1%NaOH-treated solid gave of 0.3061 g sugar/g substrate that increased from 0.2656 using cellulase. While NaOH+IL pretreatment resulted in the biggest yield modelled observed at 0.3760 employing a mixture of enzymes if compared to that of single cellulase obtained at 0.3002 g sugar.g substrate.

Table 2. The comparative yields of sugar released from treated substrates (in g sugar/g substrate) with experiment and kinetic model data vs time course for employing the mixture cellulase and xylanase.

| t(h) | No Pretreatment | 1%NaOH- | 4%NaOH- | IL- | 1%NaOH+IL- |
|------|----------------|---------|---------|-----|------------|
|      | Exp. Yield | Model Yield | Exp. Yield | Model Yield | Exp. Yield | Model Yield | Exp. Yield | Model Yield | Exp. Yield | Model Yield |
| 0    | 0.0000     | 0.0000   | 0.0000   | 0.0000   | 0.0000     | 0.0000   | 0.0000     | 0.0000     |
| 1    | 0.0593     | 0.0048   | 0.1212   | 0.0076   | 0.0900     | 0.0064   | 0.2340     | 0.0146     | 0.1338     | 0.0098     |
| 2    | 0.0766     | 0.0096   | 0.1557   | 0.0151   | 0.1136     | 0.0128   | 0.2669     | 0.0289     | 0.1742     | 0.0195     |
| 4    | 0.0843     | 0.0191   | 0.1633   | 0.0300   | 0.1214     | 0.0254   | 0.2991     | 0.0570     | 0.1954     | 0.0385     |
| 6    | 0.0851     | 0.0285   | 0.1730   | 0.0446   | 0.1379     | 0.0378   | 0.2943     | 0.0843     | 0.2216     | 0.0572     |
| 8    | 0.0897     | 0.0378   | 0.1783   | 0.0591   | 0.1446     | 0.0501   | 0.3153     | 0.1107     | 0.2259     | 0.0756     |
| 10   | 0.1054     | 0.0471   | 0.1707   | 0.0733   | 0.1494     | 0.0622   | 0.2912     | 0.1365     | 0.2434     | 0.0936     |
| 12   | 0.1257     | 0.0562   | 0.1770   | 0.0873   | 0.1509     | 0.0742   | 0.2943     | 0.1614     | 0.2507     | 0.1112     |
| 18   | 0.1473     | 0.0831   | 0.1942   | 0.1280   | 0.1635     | 0.1092   | 0.3100     | 0.2321     | 0.2527     | 0.1621     |
| 24   | 0.1459     | 0.1093   | 0.1983   | 0.1670   | 0.1722     | 0.1429   | 0.3328     | 0.2968     | 0.2541     | 0.2100     |
| 30   | 0.1419     | 0.1347   | 0.2180   | 0.2042   | 0.1780     | 0.1753   | 0.3394     | 0.3561     | 0.2527     | 0.2552     |
| 36   | 0.1423     | 0.1594   | 0.2173   | 0.2397   | 0.1964     | 0.2065   | 0.3530     | 0.4104     | 0.2525     | 0.2979     |
| 42   | 0.1444     | 0.1833   | 0.2080   | 0.2736   | 0.1851     | 0.2365   | 0.3490     | 0.4601     | 0.2441     | 0.3381     |
| 48   | 0.1360     | 0.2066   | 0.1917   | 0.3061   | 0.1737     | 0.2653   | 0.3168     | 0.5057     | 0.2322     | 0.3760     |

Table 3 presents the constant rate of reaction kinetic model of sugars released from substrates that were pretreated by 1%NaOH-, 4%NaOH, IL, and 1%NaOH+IL and compared to non-pretreatment written in the first row. The constant rates of sugar from native coconut coir employing cellulase and mixture of enzymes are of 0.0029999 and 0.0048220 h⁻¹. When solid was treated with 1%NaOH, the constant rate increased to 0.0064311 (cellulase) and 0.0076116 h⁻¹ (cellulase+xylanase). If alkaline concentration inclined to 4%, the constant rate went down to 0.006000 h⁻¹ for two enzymatic. The decrease was similar to the experimental data obtained which was caused by the decomposing of cellulose into water-soluble substances. The combination method (NaOH+IL), the constant rate of an enzymatic reaction increased to 0.0074400 (cellulase) and 0.02 h⁻¹ (cellulase+xylanase) that were the highest rate obtained. The significant increase was primarily caused by the dissolution of lignin and hemicellulose because of alkaline treatment and then the change in substrate crystallinity after the IL application [21]. The substrate used in this study was coconut coir dust that had a lignin composition...
of around 41% [22]. The combination method, alkaline followed by ionic liquid was the best option for high lignin biomass before converting it into sugars, or bioethanol.

Figure 2. The kinetic model of reducing sugars yields released from substrates with respect to the time course employing a mixture of cellulase and xylanase.

Table 3. The constant rate the chemical reaction of sugar released from substrates using cellulase and mixture cellulase+xylanase

| No | Pretreatment     | The constant rate (1/h) cellulase | Cellulase+xylanase |
|----|------------------|-----------------------------------|--------------------|
| 1  | No-              | 0.00299999                        | 0.0048220          |
| 2  | 1%NaOH-          | 0.0064311                         | 0.0076116          |
| 3  | 4%NaOH-          | 0.0060000                         | 0.0060000          |
| 4  | IL-              | 0.0046700                         | 0.0146717          |
| 5  | 1%NaOH+IL-       | 0.0074400                         | 0.0200000          |

The regressions show that the parameters of determination for employing cellulase were calculated and found from 0.9874 (no-pretreatment), 0.8364 (1%NaOH-), 0.7966 (4%NaOH-), 0.8066 (IL-), and 0.8127 (1%NaOH+IL-). While those employing mixture of enzymes declined to 0.6111, 0.4161, 0.5572, 0.3499, and 0.34419, respectively. The low R² values are indicative that the employing of a mixture of enzymes is harder to predict than that of single cellulase.

4. Conclusions

The power series were successful to predict the amounts of sugar liberated from lignocellulose treated by an ionic liquid, alkaline, alkaline followed by ionic liquid and compared to the experimental data. It was discovered that the power series equation derived from the general reaction rate was similar to
that of the conventional method. The constant rates of an enzymatic reaction of treated substrates employing the mixture of enzymes were generally higher than those of applying the single cellulase. The determination parameters employing the mixture of enzymes were lower than that of cellulase. It meant that the sugar yielded enzymatically using cellulase+xylanase was more difficult to be predicted.

Acknowledgments
The authors would like to give thanks very much for the Higher Education Department of Indonesia Government for financial assistance and the Biochemical Engineering Lab of Institute Technology Sepuluh Nopember (ITS) Surabaya that gave authors wide access in conducting many experiments.

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