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RESEARCH PAPER

Ammonia Based Pretreatment Optimization of Cornstover Biomass Using Response Surface Methodology and Artificial Neural Network

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Abstract

Effective pretreatment of lignocellulosic biomass could be used to produce fermentable sugar for renewable energy production, which reduces problems related to nonrenewable fuel. Therefore, the purpose of this study was to produce monosaccharide sugar for renewable energy from agricultural waste via ammonia pretreatment optimization using response surface methodology (RSM) and artificial neural network (ANN). Cornstover was collected and mechanically pre-treated. RSM and ANN were applied for experimental design and optimum parameters estimation. Cornstover was converted into simple sugars with a combination of ammonia treatment subsequently enzymatic hydrolysis.

The maximum yield of glucose (87.46%), xylose (77.5%), and total sugar (442.0g/Kg) were all accomplished at 20 min of residence time, 4.0 g/g of ammonia loading, 132.5 ^{0}C of temperature, and 0.5 g/g of water loading experimentally. While 86.998% of glucose, 76.789% of xylose, and 439.323(g/Kg) of total sugar were achieved by prediction of the ANN model. It was shown that cornstover has a massive potential sugar for the production of renewable fuel. Ammonia loading had a highly significant effect on the yield of all sugars compared to other parameters. Interactively, ammonia loading and residence time had a significant impact on the yield of glucose, while water loading and residence time, had a significant effect on the yield of xylose. The accuracy and prediction of an artificial neural network are better than that of the response surface methodology.

Keywords:
Artificial Neural Network,
Biomass,
Central Composite Design,
Pretreatment,
Sugar

Introduction

Increment in the exhaustion of fossil fuel and fossil fuel drawbacks leads to hunting for elective vitality source from a renewable source [1]. If we are expected to achieve the changes required to address the effects of global warming and transportation problem, the use of a renewable energy source is becoming more and more important [2]. Investigation on the utilization of lignocellulosic biomass as elective vitality feedstock to fossil fuel has picked up significant consideration majorly due to their availability and critical parts in CO_2 reduction [3]. Lignocellulosic biomass is the foremost abundant renewable energy source worldwide and it is expected to substitute the biofuel generation within the future [4]. Low cost and local biomass such as corn Stover, wheat straw, and rice straw byproducts could be utilized for the production of bioethanol. One noteworthy biomass source is corn Stover, which is particularly plenteous around the world [5].

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The transformation of lignocellulosic biomass into ethanol requires three handling steps: pretreatment, hydrolysis, and maturation of the sugars into ethanol. Since cellulose, hemicellulose and lignin are the main components of lignocellulosic biomass, a practical kind of pretreatment to be carried out for the expulsion of hemicelluloses and lignin which are reinforced by covalent cross-linkages and noncovalent forces [6].

Different pretreatment strategies have been used during the last decades. These strategies incorporate (i) physical (ii) chemical (iii) organic pretreatments utilizing microorganisms; (iv) physical-chemical and so on [7]. However, the generation of harmful chemicals, retentive processing time, and serious corroding of processing equipment commonly influence the usage of the above stated for pretreatment of corn stover biomass [8]. Therefore, selecting appropriate pretreatment techniques and optimizing the pretreatment conditions for effective conversion of biomass to suitable products are critical steps[2].

Liquid ammonia pretreatment is currently a popular method because of several desirable features, including the utilization of mostly non-polluting, non-corrosive, non-poisonous, mild conditions, and high effectiveness [9], expel lignin, depolymerizing hemicellulose, and debasing the crystalline range of cellulose [10, 11]. In addition, the residue of ammonia can serve as a nitrogen source for microorganisms amid maturation.

The different investigation has been utilized using physical pretreatment as well as chemical pretreatment methods on the optimization of pretreatment conditions for cornstover biomass conversion to fermentable sugar. However, pretreatment of this biomass was not investigated using ammonia-based pretreatment optimization of cornstover using response surface methodology (RSM) and artificial neural network (ANN) methods for efficient degradation of this biomass. To overcome this issue, optimization and prediction of RSM and ANN are widely used in optimizing various process variables for pretreatment conditions [12]. RSM was applied to optimize the multivariate system to determine individual as well as combination influences of process parameters. However, these models are used only for a restricted range of parameters and thus, impose a restriction on the use of RSM models for non-linear behavior [13] and this limitation has been fulfilled by ANN model [14-16]. MATLAB® 2014a was used to build up the ANN model with feed-forward Multilayer backpropagation (FMBP) to predict the response.

Therefore, the goal of this study was ammonia-based pretreatment parameters optimization and prediction of cornstover biomass using RSM and ANN methods for lignocellulose degradation to simple sugars.

Materials and Methodology

Materials and chemicals

Cornstover was collected from local farmer Jimma, Ethiopia. It was washed, dried, milled, and screened to select the fraction of particles with a size lower than 0.6 mm and stored for a subsequent experiment.

All chemicals were purchased from Birbirsa Goro chemical purchaser (Addis Ababa, Ethiopia). The cellulase (Novozyme 4513) and b-glucosidase (Novozyme 4510) were purchased from the Holeta agricultural research center in Ethiopia.

Ammonia pretreatment

The liquid ammonia pretreatment of cornstover was conducted in a 1000 mL using a high-pressure reactor (CBC-2L 30/300, automatic control, Chemistry department Lab.) with designed ammonia loading to dry biomass ratio. Cornstover was pre-homogenized with distilled water for optimum water loading. The homogenized sample was placed into the high-pressure reactor. Then; ammonia loading according to experimental design was injected into the reactor containing biomass. The reactor was heated rapidly to set temperature, and the treated samples were dried at 40 0C for one day and stored.

Liquid ammonia pretreatment parameters were optimized to maximize degradation of cornstover biomass using an RSM on central composite design and ANN model. By varying temperature (100−160 0C), the mass ratio of water to cornstover biomass (0.5−2.0 g/g, the mass ratio of ammonia to cornstover biomass (0.6−4.0 g/g), and residence time (10−30 min). The selected independent variables and their ranges were selected according to previous studies [14].

Response surface methodology experimental design

Center composite designs Expert 11.0.1version was applied for experimental design and optimize the pretreatment parameters of cornstover biomass. The independent variables were temperature (C^0), water loading (g/g), ammonia loading (g/g), and residence time (min). The scopes and levels of pretreatment parameters as indicated in Table 1. The glucose, xylose, and total sugar yields were used as dependent parameters for analyses.

Table 1. Central composite design parameters and levels

Parameters	Unit	Minimum	Maximum	Levels		
A	min	10	30	10(-1)	20(0)	30(+1)
B	g/g	0.5	4	0.5(-1)	2.25 (0)	4(+1)
C	g/g	0.6	2	0.6(-1)	1.3 (0)	2(+1)
D	C	100	160	100(-1)	130(0)	160(+1)

A: Time, B: Ammonia loading, C: Water loading, D: Temperature, g/g: mass %

The second-order polynomial was employed to generate the model between responses and pretreatment conditions, and it was expressed as Eq. 1.

$$Y = \beta 0 \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i X_i + \sum_{i=1}^{k-1} \sum_{j=i+1}^{k} \beta_{ij} X_i X_j + \epsilon \quad (1)$$

where, Y, xi and xj, β0, βi, βi, βii, k and ϵ are anticipated response, input variable, constant term, linear coefficient, quadratic coefficient, interaction term, number of variables, and random error respectively.

Artificial neural network (ANN) modeling and prediction

MATLAB® 2014a was used for the formulation of an artificial neural network model using a feed-forward multilayer network contains three primary layers known as input (used hyperbolic tangent sigmoid transfer function), hidden and output layers (used pure-linear transfer function) to anticipate the yield [17].

$$F(x) = \mathrm{tansig}(x) = (1 - e^x)/(1 + e^x) \quad (2)$$

The developed artificial neural network model constitutes inputs (residence time, ammonia loading, water loading, and temperature) and outputs (glucose, xylose, and total sugar yield). The Marquardt–Levenberg back-propagation (MLBP) algorithm was selected for training. The input and output data detected from the actual values are categorized into three different parts; 70% (20 samples) for training, 15% (5 samples) for testing, and 15% (5 samples) for validation.

To find out the best training efficiency and reduce the effect of larger values in input and output data, input, and output was normalized between -1 and 1 [12]. The normalized data was forwarded to the ANN in feed-forward multilayer backpropagation. The mean square error values between the output neurons and the observational outputs were determined and backward propagated via the network. Then, the individual weights of the neuron, corrected by the algorithm. After the ANN tool memorizes the data from the training, cross-validation was applied to prevent the training's fit. By repetition in testing several NN, the best number of neurons in the hidden layer was determined when the mean square error (MSE) value of the output reached its minimum value [18].

Analytical methods

Moisture was measured using an analyzer (Sartorius, Model MA25; Jimma Ethiopia). Glucan, xylan, lignin arabinan, and ash contents of cornstover biomass were determined by the analytical procedure of Ethiopian Paper and Pulping Industry Laboratory (EPPIL) using two-step acid hydrolysis.

Enzymatic hydrolysis

Enzymatic hydrolysis was developed using LAP0011 [17] and substrate gotten from ammonia pretreatment was hydrolyzed using 1.5% glucan stacking in vials without washing. Every hydrolysis was accomplished in 100 mL by adding volume up to 15 mL. A 0.08 mol/L citrate buffer was utilized to alter the hydrolysis holding pH solution at 4.78. To avoid microbial defilement, tetracycline; 40 mg/L, and cycloheximide; 30mg/L were included in hydrolysis, individually.

The cellulose of 20 FPU/g glucans, b-glucosidase of 25 CBU/g glucans, and xylanase of 800 IU/g glucans were utilized, for all hydrolysis tests, separately. Shaking incubator at 48 0C and 16.5 rad/s, was utilized in all hydrolysis tests [20]. The tests were solidified at a negative temperature of 20 0C for HPLC investigation. All tests were carried out in triplicated.

HPLC analysis

The hydrolyzates from compositional investigation monosaccharide sugars and enzymatic digestibility were decided by the HPLC framework (Agilent 1300 Arrangement). The Agilent HPLC framework was prepared with a refractive index detector and a CG column (Aminex HPX-75H). The versatile stage was 0.005 mol/L H_2SO_4 solution working at a stream rate of 0.6 mL/min, with temperature operated at 60 0C. All tests were repeated, and the mean value was demonstrated. The yield of monosaccharide sugar was determined according to [14], Eqs. 3 and 4.

$$\mathrm{Gyield} = \frac{\mathrm{mass_{gluose}}}{\mathrm{f_1} * \mathrm{mass_{glucan}}} * 100\% \tag{3}$$

$$\mathrm{Xyield} = \frac{\mathrm{mass_{xylose}}}{\mathrm{f_2} * \mathrm{mass_{xylan}}} * 100\% \tag{4}$$

where mass $_{glucose}$ and mass$_{xylose}$ are, the mass of glucose and xylose discharged from glucan and xylan by enzymatic hydrolysis, mass$_{glucan}$ and mass $_{xylan}$ is, the mass of glucan and xylan in crude fabric, respectively, and f_1 and f_2 are the transition factor for glucan and xylan to glucose and xylose respectively (f_1=1.11, f_2 = 1.14). The mass of monosaccharide sugars (glucose and xylose) expelled per kg of dry cornstover biomass was used to determine the yield of total sugar as indicated in Table 2.

Table 2. Factors, response (experimental and predicted) value of the central composite design

Factors					Responses%						
Run	**Time**	**Amonia loading**	**Water loading**	**Temp**	**Solid**	**Glucose**		**Xylose**		**Total sugar**	
S/N	Min	g/g	g/g	^{0}C		Exp.	Pred.	Exp.	Pred	Exp.	Pred.
1	20	4	1.25	165	84.76	54.14	55.63	64.02	65.33	406.00	414.90
2	20	0.5	1.25	100	89.56	43.50	44.31	55.00	55.34	310.00	322.47
3	20	0.5	0.5	132.5	92.5	61.10	63.35	56.43	58.18	320.00	329.22
4	10	4	1.25	132.5	79.2	63.50	64.14	67.81	67.48	417.00	422.73
5	20	0.5	2	132.5	97.14	66.09	68.64	52.30	54.00	301.56	299.83
6	20	4	1.25	100	91.5	70.60	69.72	72.10	71.44	425.01	430.78
7	30	2.25	2	132.5	86.1	78.90	81.67	58.02	57.10	350.80	354.10
8	30	2.25	1.25	165	85.78	68.98	65.93	60.32	59.66	346.45	360.85
9	20	2.25	2	165	84.58	65.32	65.56	59.42	58.66	350.70	346.05
10	10	2.25	1.25	100	90.5	65.10	66.08	64.21	63.77	375.12	376.51
11	30	2.25	1.25	100	90.68	62.60	64.95	70.50	69.98	370.10	376.73
12	20	2.25	0.5	100	89.42	73.50	70.08	68.05	66.11	405.90	391.32
13	20	2.25	1.25	132.5	92.67	75.43	72.60	65.30	63.98	372.70	368.68
14	30	4	1.25	132.5	89.31	77.89	77.80	75.70	76.25	433.10	422.95
15	30	2.25	0.5	132.5	95.61	82.45	84.46	73.00	72.54	396.70	383.49
16	10	2.25	2	132.5	94.1	81.50	80.09	71.70	69.20	360.70	353.88
17	20	0.5	1.25	165	96.1	53.90	52.43	57.89	56.84	313.57	306.59
18	10	2.25	1.25	165	83.6	58.89	59.13	61.30	62.51	367.80	360.64
19	20	2.25	0.5	165	84.21	75.76	78.16	65.45	63.50	389.58	375.44
20	20	4	0.5	132.5	91.73	87.40	84.48	77.50	78.41	442.00	437.53
21	30	0.5	1.25	132.5	90.5	54.80	52.67	52.30	53.39	303.12	314.64
22	10	2.25	0.5	132.5	87.32	83.80	80.37	59.04	57.08	388.78	383.27
23	10	0.5	1.25	132.5	88.34	60.80	60.66	58.67	58.79	312.34	314.42
24	20	2.25	2	100	78.98	80.54	79.61	66.60	67.63	370.00	361.93
25	20	4	2	132.5	77.74	77.50	76.13	73.10	72.30	410.10	408.14
26	10	4	0.5	100	82.6	68.98	71.94	62.00	62.85	440.60	445.37
27	10	4	0.5	165	83.56	64.78	64.94	56.89	57.48	420.12	429.49
28	20	2.25	1.25	132.5	87.8	70.21	72.60	60.90	63.98	355.80	368.68
29	30	0.5	2	100	80.4	56.78	54.92	49.04	47.51	316.50	307.88
30	20	2.25	1.25	132.5	79.56	70.90	72.60	60.67	63.98	365.05	368.68

For determination of biomass compositions, all experiments were carried out in the triplicated run and the average mean was indicated in Table 3.

Table 3. Cornstover biomass composition analysis on a dry basis

Components	**Glucan**	**Xylan**	**Arabinan**	**Lignin**	**Ash**	**Total carbohydrate**
Cornstover (%)	31.01±0.1	17.1 ± 0.0	4.505 ± 0.1	13.1 ± 0.3	8.4.43 ± 0.1	52.615± 0.20

Comparison of ANN and RSM performance

The coefficient of determination; R^2, Root mean square error; RMSE, mean average error; MAE, standard error of prediction; SEP, and absolute average deviation; AAD was determined to check the accuracy and predictive ability of ANN and RSM using Eqs. 5 to 10:

$$R^2 = 1 - \sum_{i=1}^{x} \left[\frac{(pr - ex)^2}{(pr - m)^2} \right] \quad (5)$$

$$\mathrm{MSE} = 1 - \frac{1}{x}\sum_{i=1}^{x}[(pr - ex)^2] \tag{6}$$

$$\mathrm{RMSE} = \sum_{i=1}^{x}[\mathrm{MSE}]^{1/2} \tag{7}$$

$$\mathrm{SRP} = \left[\frac{\mathrm{RMSE}}{m}\right] \tag{8}$$

$$\mathrm{AAD} = \frac{100}{x}\sum_{i=1}^{r}\left[\frac{e_x - p}{e_x}\right] \tag{9}$$

$$\mathrm{MAE} = \sum_{i=1}^{x}\left[\frac{ex - p}{ex}\right] \tag{10}$$

where x is the number of runs, p_r is predicted values from the model, e_x is experimental values and m is mean experimental values

Results and Discussion

Cornstover biomass composition analysis

The components of cornstover used in this study are indicated in Table 3. This biomass contains glucan of 31.01 ± 0.07%, xylan of 17.1 ± 0.02%, arabinan of 4.505 ± 0.11%, lignin of 13.1 ± 0.29% and ash of 8.4.43 ± 0.10% on dry basis (Table 3). Weiss et, al. [21] reported the previous result on the compositions of cornstover was in line with these results., Developing areas, seasonal, evaluation methods, and so on, can affect the chemical composition of feedstock [14]. The relative variation in the cell wall content of various cornstover did not make a difference significantly. Relative to the total sugar, glucan, and xylan are accounts for 91.4% of raw material (Table 3). This result indicates ammonia-based pretreatment is an alternative method for lignocellulose biomass degradation.

Response surface methodology statistical data analysis

A very good correspondence between the observational and anticipated values for the sugar yields was obtained from the check bit plot between the expected and the observational values as shown in Fig. 1a, 1b, and 1c. The scatter plots were distributed relatively near to the diagonal, and the correlations between anticipated and experimental values of sugar yields were satisfied with above ninety percent.

The test framework and the outcome of solid and sugar yields are displayed in Table 2. As indicated in Table 2, the solid yields of liquid ammonia-treated substrates extended from 77.5% to 97.14%. Relative to the number of runs, most solid yields of the experiments were over 86%, which demonstrated a slight amount loss of material at the pretreatment stage, due to human or process equipment problems. The yields of glucose, xylose, and total sugar were extended from 43.5−87.4%, 52.3− 77.5%, and 301.56−442.0 g/kg dry biomass respectively (Table 2). It appears that the pretreatment conditions of fluid alkali (ammonia) pretreatment had the most prominent impact on the yields of sugar. The most extreme yield of glucose, xylose, and total sugar were all accomplished at 20 min of residence time, 4.0 g/g of ammonia loading and 132.5 ^{0}C of temperature, 0.5 g/g of water loading (Table 2).

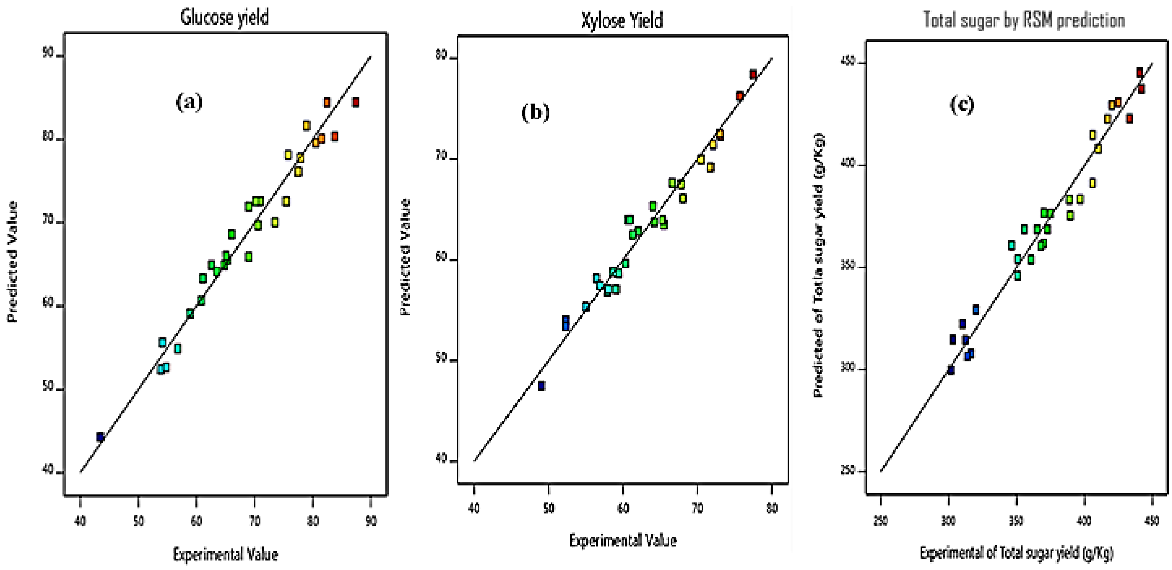

Fig. 1. Actual sugar yields versus predicted sugar yields from regression models: (a) glucose, (b) xylose, and (c) Total sugar by RSM prediction

Past works demonstrate that ammonia fiber expansion treated biomass is 80−91% of glucose and 50−78% of xylose yields separately [22]. In this manner, the quality of liquid ammonia pretreatment was corresponding to ammonia fiber expansion. The outcome of ANOVA for the models is demonstrated in appendix Table 4. To analysis the statistical significance of the proposed models, F-value and P-value were applied. It is shown that the model is statistically significant with a confidence level of above 95% if the P-value for the model is below 0.05 [21]. As demonstrated in Table 4, the P-value for all yields was below 0.0001. In this manner, all models are highly significant, meaning variable needs to be controllable.

The regression coefficient (in terms of coded), and all correlation coefficients (R^2, R^2adj. and R^2-pred.) have been used to test the goodness of the model [23]. The Pred. R^2 is in reasonable estimate with the Adj. R^2; i.e. the distinction is much less than 0.2 (Table 4). A very small value of the coefficient of variation (CV: Table 4) clearly shown a very strong degree of precision and a great deal of reliability of the experimental values [23].

The quadratic, 2FI (two-factor interaction), and linear model were suggested by central composited design for glucose, xylose, and total sugar yields respectively. The suggested model equations based on ANOVA analysis results were indicated in Eqs. 11 to 13.

Final Equation in Terms of Coded Factor

$$G_Y(\%) = +63.98 + 0.84\ A + 7.89\ B - 0.83\ C - 2.89\ D + 3.54\ AB - 6.89\ AC - 2.26\ + 1.26\ BC - 3.65\ BD - 1.59\ CD \qquad (11)$$

$$X_Y\ (\%) = \ +63.98 + 0.84\ A + 7.89\ B - 0.83\ C - 2.89\ D + 3.54\ AB - 6.89\ AC - \ 2.26\ AD + 1.26\ BC - 3.65\ BD - 1.59\ CD \qquad (12)$$

$$T_{SY}\left(\frac{g}{Kg}\right) = +368.68 + 0.11 * A + 54.16 * B - 14.70 * C - 7.94 * D \qquad (13)$$

Where G_Y, X_Y and T_{SY} are glucose yield, xylose yield and total sugar yield respectively, A is residence time, (min), B is ammonia loading (g/g), C is water loading (g/g) and T is temperature (^{0}C).

Table 4. ANOVA information for central composite design models

Glucose yield						
Source	SS	DF	MS	F-value	p-value	
Model	3072.13	14	219.44	27.14	< 0.0001	significant
Residual	121.29	15	8.09			
Lack of Fit	105.21	13	8.09	1.01	0.6033	not significant
Pure Error	16.08	2	8.04			
Total	3193.42	29				
Xylose yield						
Source	SS	DF	MS	F-value	p-value	
Model	1489.54	10	148.95	46.64	< 0.0001	significant
Residual	60.67	19	3.19			
Lack of Fit	47.06	17	2.77	0.406	0.8847	not significant
Pure Error	13.62	2	6.81			
Total	1550.21	29				
Total sugar yield						
Source	SM	DF	MS	F-value	p-value	
Model	51619.9	4	12904.9	150.21	< 0.0001	significant
Residual	2147.77	25	85.91			
Lack of Fit	2004.54	23	87.15	1.22	0.5478	not significant
Pure Error	143.23	2	71.62			
Total	53767.69	29				
Correlation coefficients						
Coefficients	Glucose	Xylose	Total sugar			
R^2-squared	0.962	0.961	0.960			
Adjusted R^2	0.836	0.940	0.954			
Predicted R^2	0.927	0.913	0.942			
Adeq Precision	19.978	28.552	38.460			
CV %	4.15	2.83	2.50			

CV is coefficient of variation, SS is Sum of Squares, MS is Mean Square and DF is Degree of freedom

Effect of pretreatment conditions on the responses

Analysis of variance (Table 4) was carried out to measure the significance of the formulated model as well as each of the coefficients (p-value < 0.05). Ammonia loading had a highly significant effect on all the sugar yields. The effect of ammonia-to-biomass ratio was studied at 0.4–4:1 g of anhydrous ammonia: g of dry biomass. Glucan and xylan transformation enhanced with enhancing ammonia stacking and achieved the higher value at ammonia to biomass ratio of 4:1 g/g. The results indicated that the ammonia loading and temperature had more critical impacts than water loading and residence time on glucan transformation, which concurs well with the conclusion of [4].

With increasing temperature, the expanding cleavage of inside bonds in biomass, and expanding dissolvability of biomass components encourage enzymatic hydrolysis. In addition, in raise temperatures, the maximum yields can be achieved using shorter residence time. The lower temperatures are not sufficient to penetrate the bonds between the biomass molecules within a short time to achieve higher sugar yields [24].

To promote the sugar yield well, certainly, the higher ammonia loading was benefitable. Since liquid ammonia pretreatment was anticipated to extend chemical availability to the polysaccharides and ammonia is capable to adhere to lignin-carbohydrate ester linkages, cause the swelling of cellulose, destruct the crystalline area, and alter the precious structure [14].

Direct water stacking was useful to higher glucose discharge of liquid ammonia-treated substrate. This is confirmed with the results found from switchgrass and corn stover treated by ammonia fiber expansion [22].

Interactively, residence time and ammonia, as well as residence time and water loading had significant effects on the yield of glucose and xylose respectively at a 95% confidence interval. The impact of variables on glucose and xylose yields taking after enzymatic hydrolysis are displayed as 3D plots (Figs. 2a, and 2b) for highly significant variables.

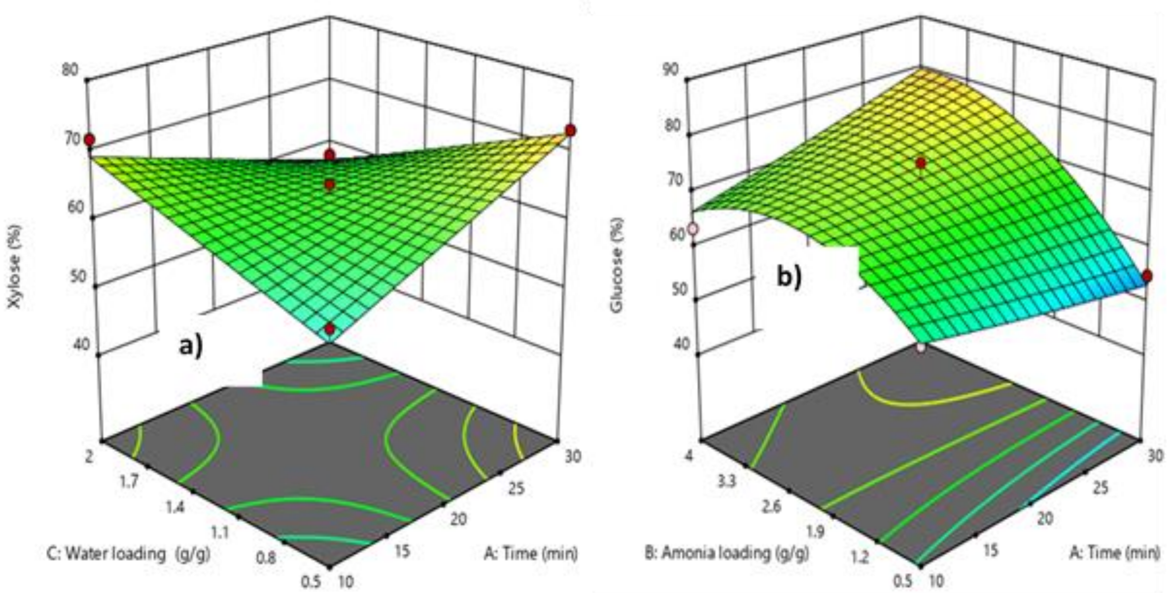

Fig. 2. Actual sugar yields versus predicted sugar yields from regression models: (a) glucose, (b) Xylose and (c) total sugar by RSM prediction

Residence time is one of the factors affecting pretreatment effectiveness. Referring to Table 1, the influence of residence time was studied at 10 to 30 min. Figs. 2a and 2b, show the effects of residence time on both xylose (2a) and glucose (2b) sugars yield. The sugar yield was maximum at 20 min of residence time. This indicated that the lower yield of sugar at higher residence time could have been due to the sugar's destruction into furfural. At more insufficient residence time, the degradation of lignocellulosic biomass to simple sugar could not effectively occur [25]. In addition, increasing the residence time slightly enhanced xylan and glucan transformation and in other cases diminished this transformation [26].

The maximize glucan transformation calculating by Eq. 12 was 89.1%, pretreated with prescribed conditions. Fig. 3 indicates the 3D plots for the (glucose and xylose) yield whereas holding the other two factors at center point esteem (zero, coded units). As viewed from Figs. 2a and 2b, all sugar yields were highly sensitive to ammonia stacking. Water stacking had the slightest impact on glucose and xylose yield, even though it had a critical effect on total sugar yield, at higher, water loading, the yield of total sugars become lower while at lower water loading, the yield becomes higher. This fluctuation

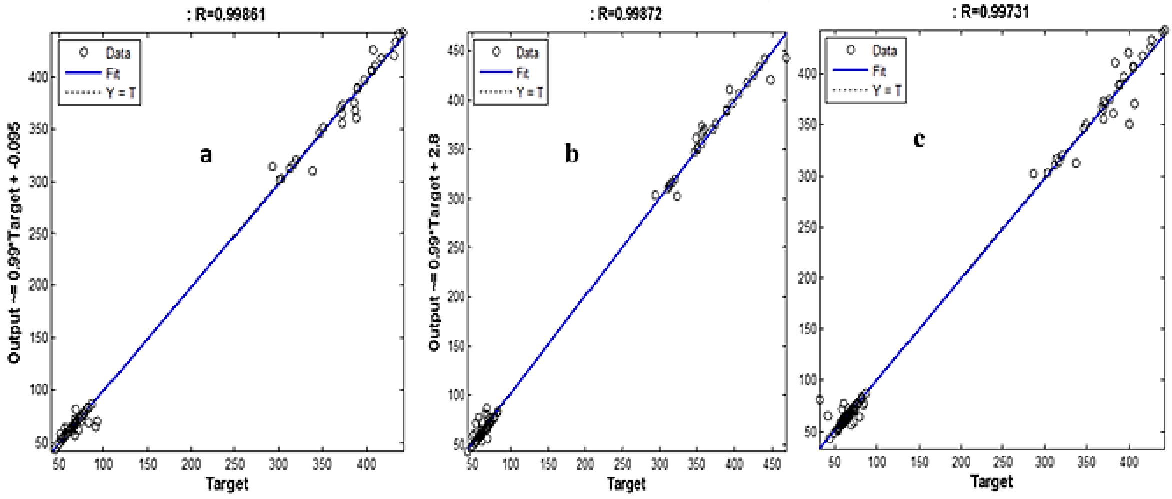

Fig. 3. Glucose (a), Xylose (b), and Total sugar yield (c) by ANN prediction

Artificial neural network (ANN) modeling and prediction

The experimental results, utilized for ANN training and individual test, are shown in Table 5. The ANN-multi-layer perceptron (MLP) has four inputs (time, ammonia loading, water loading, and temperature), a hidden layer, and three outputs (glucose, xylose, and total sugar yield). The ANN-based model was done by choosing the appropriate training algorithm, determining the optimum value of the neuron, and validating the model. 70% of samples of the data were used for training, 15% samples for testing, and 15% samples for validation. Using the available actual value, the Levenberg–Marquardt (LM) ANN fitting tool and TANSIG Transfer Function 4–10–3 (number of the input layer, neurons in the hidden layer, and output layer) model were implemented.

Statistical parameters were used to determining the higher predictive power of the two model techniques using Eqs. 5 to 10 as shown in Table 4. The relationships among the parameters are specified by the correlation coefficient (R). A unit (1) value of R implies a perfect relationship between variables; while a zero (0), value is believed to be the absence of a linear relationship between the parameters.

The actual value and anticipated values for each observation were plotted in Figs. 3a, 3b, and 3c. These graphs consist of the exact line shows y = x, meaning, the anticipated value is equivalent to the actual value with the highest level of correlations and highest coefficient of determination (R^2) compared to RSM prediction indicated in Figs. 1a, 1b, and 1c.

After assigning training information, transfer function, the number of the hidden layer, the number of the neuron, the performance of the ANN tool needed to be evaluated [25]. The best solution was chosen based on the highest coefficient of correlation and least MSE for training, testing, and validation. Therefore, ten neurons were selected depending on, best ANN performance evaluation. Figs. 4a to 4d indicate the regression plots of the training, validation, test, and all R-value with the LM algorithm. The correlation coefficients (R) between the actual and the expected values; 1 for training, 0.9993 for testing, 0.9978 for validation, and 0.9994 for overall correlation. Therefore, the ANN anticipation for training, validation, and testing is highly substantial and meritorious in terms of correlation and MSE.

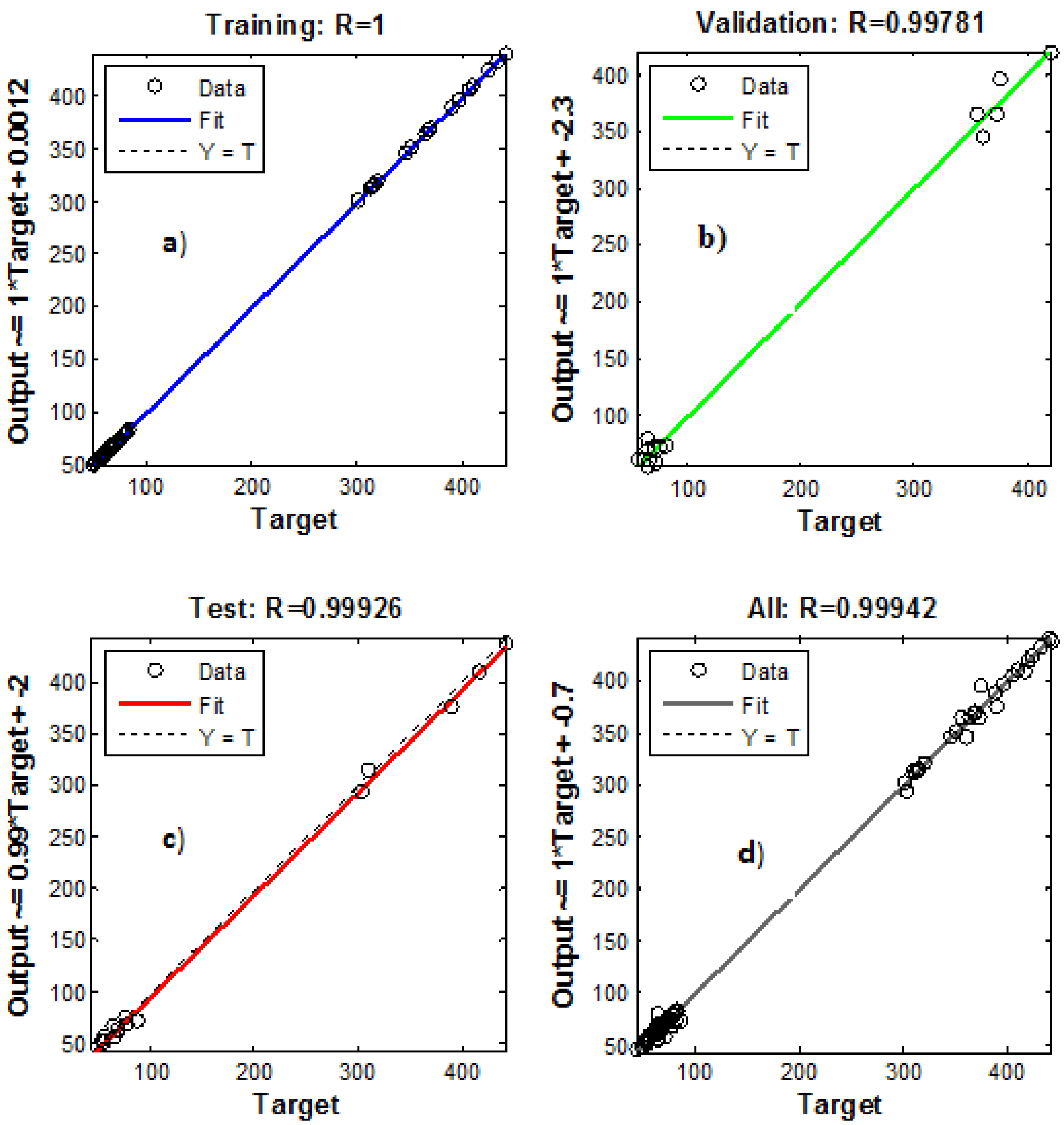

Fig. 4. Correlation coefficients for mean sugar yield

Comparison of RSM and ANN Performance

To identify the best model that accurately predicts the effect of optimum parameters on the yield, the statistical parameter has been calculated using Eqs. 10 to 13. The statistical analysis results and comparison between RSM and ANN models are listed in AppendixTable 6. The result has shown that ANN has a lower RMSE and higher correlation values compared to the RSM model. Therefore, the ANN model is a superior modeling tool than the RSM demonstration due to its lower RMSE, high prediction of the correlation value.

Table 5. Result of RSM and ANN prediction on sugar yields.

Run.	Glucose yield (%)		Xylose yield (%)		Total sugar yield (%)	
	RSM	ANN	RSM	ANN	RSM	ANN
1	55.63	54.1384	65.33	64.018	414.9	405.9979
2	44.31	45.79212	55.34	51.42702	322.47	314.4898
3	63.35	61.10169	58.18	56.43171	329.22	320.0074
4	64.14	66.83197	67.48	64.21016	422.73	409.6739
5	68.64	66.08825	54	52.30104	299.83	301.561
6	69.72	70.59853	71.44	72.0932	430.78	424.9858
7	81.67	78.89818	57.1	58.0182	354.1	350.7988
8	65.93	68.98285	59.66	60.32072	360.85	346.4544
9	65.56	65.31761	58.66	59.41596	346.05	350.6958
10	66.08	79.62241	63.77	69.632	376.51	396.1174
11	64.95	62.59318	69.98	70.49514	376.73	370.0907
12	70.08	73.50235	66.11	68.0461	391.32	405.8856
13	72.6	70.90034	63.98	60.66659	368.68	365.0397
14	77.8	77.8896	76.25	75.6958	422.95	433.0937
15	84.46	86.45689	75. 54	76.00776	437.49	439.7053
16	80.09	73.20735	69.2	58.22501	353.88	345.4346
17	52.43	53.90294	56.84	57.88776	306.59	313.5717
18	59.13	58.89263	62.51	61.29849	360.64	367.7932
19	78.16	75.01486	63.5	56.6561	375.44	375.2802
20	84.48	86.9983	75.41	76.78915	437.53	439.3232
21	52.67	56.38946	53.39	53.70105	314.64	293.4242
22	80.37	83.80626	57.08	59.03256	383.27	388.7599
23	60.66	60.80118	58.79	58.66797	314.42	312.3385
24	79.61	80.53932	67.63	66.5937	361.93	369.9795
25	76.13	77.49898	72.3	73.09892	408.14	410.0975
26	71.94	68.98422	62.85	61.99243	445.37	440.5547
27	64.94	54.36141	57.48	60.44439	429.49	419.5385
28	72.6	70.90034	63.98	60.66659	368.68	365.0397
29	54.92	56.77855	47.51	49.04	307.88	316.5027
30	72.6	70.90034	63.98	60.66659	368.68	365.0397

Table 6: Relative statistical data information of RSM and ANN model

	Glucose		Xylose		Total sugar	
Variables	**RSM**	**ANN**	**RSM**	**ANN**	**RSM**	**ANN**
R2	0.962	0.9986	0.961	0.9987	0.960	0.9973
RMSE	0.627368	0.568154	0.6412	0.5123	0.5831	0.51034
SEP	0.007201	0.006576	0.0085	0.0051	0.0082	0.00612
MAE	0.521	0.352	0.6214	0.4234	0.5889	0.4012
AAD	0.547091	0.37780	0.5632	0.4012	0.51041	0.3505

Conclusions

Ammonia-based cornstover biomass pretreatment was applied using response surface methodology (RSM) and artificial neural network (ANN) models. The main compositions of cornstover biomass were glucan; 31.01 ± 0.07%, xylan; 17.1 ± 0.02%, lignin; 13.1 ± 0.29%.

The optimum conditions for ammonia-based pretreatment of cornstover biomass were; at 20 min of residence time, 4.0 g/g of ammonia to biomass ratio, 132.5 ^{0}C of temperature, and 0.5 g/g of water to biomass ratio. Under these conditions, 86.998 % of glucose, 76.789 % of xylose, and total sugar 439.323 g/Kg yields were achieved by predicting artificial neural networks compared to experimental and response surface methodology results.

The outcome from enzymatic hydrolysis of ammonia pretreated corn stover showed that liquid ammonia is a viable pretreatment method.

The effect of parameters was studied and the result has shown that ammonia loading had a highly significant effect on the yield of all sugars. Whereas water loading had the slightest impact on the yield of glucose and xylose and in contrast, water loading had a critical effect on the total sugar yield.

In addition, the results indicated that the ammonia loading and temperature had more critical impacts than water loading and residence time on glucan transformation. This indicated that expanding temperature, the extending cleavage of interior bonds in biomass, and the dissolvability of biomass components empower enzymatic hydrolysis.

The higher ammonia loading is benefitable to promote the sugar yield well since ammonia is capable to adhere lignin-carbohydrate ester linkages, cause the swelling of cellulose, and altering the biomass structure.

Residence time is one of the variables affecting pretreatment conditions. Increasing the residence time slightly enhanced sugar transformation however, behind optimum value diminished this transformation, meaning behind this value, the sugar starts to form substance-like furfural.

ANN model predicted the maximum yields of all responses compared to RSM. Therefore, ANN model is a superior tool than that of the RSM demonstration, due to its higher prediction of the yield

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Conflict of interest:

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Abbreviations

^{0}C	Degree Celsius
2 FI	Two-factor interaction
ANN	Artificial neural network
C.V	Coefficient of variation
CCD	Central composite design
Eq.	Equation
F	Fisher test
g	Gram
g/g	Gram/Gram
kg	kilogram
mg	Mill gram

min	Minute
mL	Mill liter
P-value	Probability value
R^2	Coefficient of determination
R^2_{adj}	Adjusted coefficient of determination
RMSE	Root Mean Square Error
RSM	Response surface methodology
SSE	Sum of Squared Errors
Std. Dev.	Standard deviation
wt	Weight

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