SYNTHESIS AND OPTIMIZATION OF ACTIVATED CARBON FROM WASTE TEA USING RESPONSE SURFACE METHODOLOGY TECHNIQUE FOR THE ADSORPTION OF METRONIDAZOLE

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ABSTRACT

Waste tea activated carbon (WTAC) was produced at optimum condition through chemical activation of potassium hydroxide (KOH) for the adsorption of Metronidazole (MNZ) at various conditions. Response surface methodology statistical technique was used to optimized the preparation conditions using central composite design (CCD). The preparation conditions are activating temperature, activating time and impregnation ratio (IR) with percentage yield and percentage removal as targeted response. The optimal conditions obtained for good percentage yield and the removal of MNZ removal were 735 °C activation temperature, 45 min activation time, and impregnation ratio of 1.5. From the ressult of analysis of variance (ANOVA), the significance of various factors and their influence on the response were identified. The regression coefficient (R^2) of the model developed and result of validation experiments conducted at optimum conditions shows that the predicted values are in good agreement with the experimental result. Response surface plots were used to determine interaction of the variables on the percentage yield and MNZ removal. The sample, (waste tea) was characterized before and after activation using FTIR analysis to determine the functional groups present.

Keywords: Pharmaceuticals, Antibiotics, Metronidazole, Waste tea, Activated carbon, Response surface methodology

1. Introduction

Polluted water, most especially one with toxic substances such as heavy metals, pharmaceuticals and organic pollutant (chemicals, dyes, hydrocarbons, phenols, herbicides, pesticides, insecticides and detergents) are a major problem to man and his environment (Erdem *et al.*, 2004). Among the different toxic substances released into water bodies; Pharmaceuticals, been one of the most dangerous are considered to be a class of emerging contaminants (Manjunat *et al.*, 2017) because they are still active even at low concentration and are not susceptible to biological degradation (Guo *et al.*, 2017). They are synthetic or natural medicines, formulated to give pharmacological effects (WHO, 2011). Its photo active nature (ability to absorb light) ultra-violet (UV) and visible radiation are possible due to the presence of hetero-atoms, aromatic rings and other functional groups (Prados *et al.*, 2011) which enables scientists to detect their presence in water bodies. Pharmaceutical include; antiepileptic, antidepressants, hormonal drugs, antibiotics, anti-inflammatory and analgesics (EPA, 2010).

Antibiotics are majorly employed in human and veterinary medicine for the treatment and prevention against wide range of bacterial, parasitic and fungal infections (Ur-Rehman *et al.*, 2015; El-Sayed *et al.*, 2014). They include sulfamethoxole, metronidazole, tetracycline, chloramphenicol, quinolenes, norfloxacin, and penicillin (Ravdin, 1995; Ahmed and Theydan, 2013a; Ahmed, 2017; Pan and Chu, 2016). However the application of this organic compound cannot be over emphasized: with the threat it poses to aquatic and terrestrial organisms (Halling *et al.*, 1998; Chang *et al.*, 2010). Metronidazole (MNZ), also known as flaggyl, is a synthetic derivative of Azomycin; a nitroimidazole antibiotic which consists of dimetridazole (DMZ), ronidazole (RNZ), and tinidazole (TNZ) (Fakhri *et al.*, 2017). It possess antibacterial and anti-inflammatory properties which are the reasons they are prescribed for treating infections caused by anaerobic bacteria, protozoans, and bacteroides such as trichomenas vaginalis, gardia lamblia and gingivitis (Bendesky *et al.*, 2002; Lanzky and Haning, 1997). It also serves as an additive in poultry and fish feed to eliminate parasite (Cohen *et al.*, 2010; Balark and Mostafapour, 2016).

Therefore, this research work is aimed at the synthesis and optimization of waste tea activated carbon, its optimum preparation conditions that will give good yield and high percentage MNZ removal from aqueous solutions.

2. Materials and methods

2.1. Raw Material

Waste tea was collected from local tea shops around Federal University of Technology Minna in Bosso Local Government Area of Niger State, Nigeria. It was repeatedly boiled and washed with distilled water to remove soluble and coloured components until the water was virtually colourless. The effort was done to avoid possible interference of the soluble portion or coloured component of the waste in the absorbance readings on the UV spectrophotometer. It was rinsed with distilled water and dried in an oven at 80 °C for 150 min. The sample was sieved with a 125 μm mesh, packed in an air tight container for further use.

2.2. Preparation of Activated Carbon

Waste tea was activated with KOH as activating agents under the consideration of the following factors: temperature of activation (315–735 ^{0}C) and time of activation (20–70 min) and impregnation ratio (0.66– 2.34 g). 4 g of dried precursor of particle sizes 125 μm was measured and mixed with KOH activating agent to precursor ratio (IR) in a beaker. The impregnated samples were left for 24 h, then oven dried and sent to the muffle furnace for carbonization. The carbonized samples was cooled and then washed with 0.2 M HCl followed by washing with distilled water until a pH of 7 is attained. At the end of the experimental, the prepared adsorbent was dried in an oven at 105 ^{o}C for 90 min and then stored in airtight container for further use.

$$\text{IR} = \frac{weight\ of\ KOH}{Weihgt\ of\ waste\ tea} \quad (1)$$

2.3. Design of Experiments using Response Surface Methodology (RSM)

Central Composite Design (CCD) under RSM was applied in this work to study the interaction of the three variables (activation temperature, activation time and impregnation ratio) involved in the preparation of WTAC. This method helps to prune unnecessary experiments and checkmate if there is synergy amongst the factors or not (Auta and Hameed, 2011a). CCD is characterized by three operations namely: 2n axial runs, 2^n factorial runs and n_c center runs (Cronje *et al.*, 2011; Nasehir *et al.*, 2010). It is translated to 6 axial points, 8 factorial points and 6 replicates at the center which gives a total of 20 experiments.

Total number of experiments = $2^n + 2n + n_c$ (2)

where n is the number of factors. This design consist of a 2^n factorial code of ±1 was used to represent the eight factorial points, 2n axial points used to represent the six axial points located at (±, 0, 0), (0, ±, 0), (0, 0, ±), and n_c center points (0, 0, 0) which represent the six replicates located at the center (Sahu *et al.*, 2009). The center points are used to determine the experimental error and reproducibility of the data (Hoseinzadeh *et al.*, 2013; Khodadoust *et al.*, 2007).

Table 1. Independent variables and their coded levels for central composite design

Variable (factors)	code	unit	coded variable levels -1	 0	 +1
Activation temperature	x_1	^{0}C	400	525	650
Activation time	x_2	Min	30	45	60
Impregnation Ratio	x_3	-	1	1.5	2

Table 1 represents the coded points and their corresponding values. The value of rotatability α which depends on the number of points in the design of the factorial (Ahmad and Alrozi, 2010), was obtained from the equation (3).

$$\alpha = N_p^{1/4} \qquad (3)$$

Where $N_p = 2^k$ is the number of points in the cube portion of the design, k is the number of factors.

The optimal conditions for the responses yield and percentage removal of MNZ were determined using the optimal model predictor equation given as:

$$Y = b_0 + \sum_{i=1}^{n} b_{ii}x_i + \left(\sum_{i=1}^{n} b_{ii}x_i\right)^2 + \sum_{i=1}^{n-1}\sum_{j=i+1}^{n} b_{ij}x_i x_j \qquad (4)$$

Where Y is the predicted response, b_o is the constant coefficients, b_{ii} is the quadratic coefficients, b_{ij} is the interaction coefficients and x_i, x_j are the coded values of the variables considered. The regression analysis to fit the equations for both responses developed and for the evaluation of the statistical significance of the equation obtained with the aid of the experimental data were analyzed using Design Expert software (statistical) version 7.0.0 (STAT-EASE Inc., Minneapolis, USA). The characteristics of the reliability of the analysis carried out, was measured by the variability in the observed responses values expressed by co-efficient of determination R^2, the probability (P-value), fisher value (F-value) and the residual (Hassani *et al.*, 2014; de-Lima *et al.*, 2011; Roy *et al.*, 2014).

2.4. Analysis of the Simulated Effluent before Treatment

All chemicals used were analytical grade. The concentration of 100 mg/L was prepared by dissolving calculated amount of 200 mg of MNZ salt in 1000 mL of distilled water. 0.1 g activated carbon was added to a set of conical flask containing 100 mL of initial adsorbate concentration at a natural pH. The flasks was then placed on the water bath isothermal shaker set for temperature 30 °C, and shaker speed of 145 rpm (Auta and Hameed, 2011b). The flasks were afterwards removed from the shaker and the solution was filtered through Whatman No.41 filter paper. The collected supernatant was analyzed to determine the final concentration of the MNZ using spectrophotometer (UV) at a maximum wavelength of 340 nm. The percentage MNZ removal was calculated using equation (6).

2.5. Characterization of the Tea Waste

FTIR spectra were recorded on a Perkin-Elmer infrared spectrophotometer. This was carried out using KBr pellets with resolution of 4 cm^{-1} in the range of 400–4000 cm^{-1}. The sample and analytical grade KBr (2 mg of sample to 200 mg of KBr), were dried at 100 over-night prior to the FTIR analysis. These were carried out to determine the adsorbent surface chemistry before and after activation.

3.0 RESULTS AND DISCUSSION

3.1 Fourier transform infrared spectroscopy (FT-IR) analysis

Tables 2. Shows the corresponding band width of raw and activated tea waste respectively. The spectra have been evaluated under the light of literature (Auta and Hameed, 2011a; Auta and Hameed, 2011b; Ahmaruzzaman and Gayatri, 2010; Kafa *et al.*, 2016; Yagmur *et al.*, 2008; Amirhossein *et al.*, 2016; Antonio *et al.*, 2013). FTIR technique is mainly used to identify functional groups such as carboxyl, hydroxyl, and silicon. The wave number for the raw and activated tea waste sample was 500 - 4000 cm^{-1}. A number of peaks were displayed which reflect the complex nature of tea waste, while some peaks were observed to have shifted or disappeared and new ones detected. On comparing the FTIR spectra of both samples, the result showed that KOH activation modified the tea waste.

Table 2: FT-IR analysis of raw and activated tea waste

Band width (cm^{-1})	**Peak Assignments precursor**	**Band width (cm^{-1})**	**Peak Assignments WTAC before adsorption**
3857-3865	N-H	3865	N-H
3441-3448	O-H	3448	O-H
2816	symmetric C-O	3024	C-H
2325-2362	very sharp peak	2325-2362	very sharp peak
2122-2160	C ≡ C	2129-2160	C ≡ C
1651-1744	C = O	1651-1744	C = O
1543	C = C	1543	C = C
1034	S = O	1103	C-O
609	Si-O-Si	1034	S = O

3.2 Response Obtained from the Experiment

Table 3 shows the relationship of the parameters for synthesis of activated carbon yield and percentage MNZ removal. The percentage removal of MNZ was in the range of 57.47– 98.99 % while percentage yield of WTAC was in the range of 25.33– 37.55 %. The Activated carbon preparation variable include: temperature (315-735 °C), time (20-70 min) and impregnation ratio (0.6-2.34 g). Quadratic model selected on the software is based on the highest order polynomials (Hameed *et al.*, 2008). The six replicate runs were conducted to determine the experimental error and the reproducibility of the data. The high percentage removal of MNZ of 98.99 % maybe attributed to the increase in specific surface area, and the optimum synthesis parameter (activated temperature, activated time and impregnation ratio). The two responses, percentage yield and percentage MNZ removal for the WTAC were determined in the following ways.

$$\text{Yield (\%)} = \frac{weight\ of\ dried\ activated\ carbon\ produced}{weight\ of\ dried\ precursor\ used} \times 100 \qquad (5)$$

MNZ removal (%) = $\frac{initial\ MNZ\ Conc.\ -\ final\ MNZ\ Conc.}{initial\ MNZ\ Cocn,}$
x 100 (6)

Table 3: Experimental design matrix using RSM for preparation of WTAC

Run	Activated carbon preparation variable			Activated carbon yield	MNZ removal
	temp., x_1(°C)	Time, x_2(min)	IR, x_3	Y_1 (%)	Y_2 (%)
1	400	30	1	26.90	57.47
2	650	30	1	29.82	88.99
3	400	60	1	26.88	74.64
4	650	60	1	25.57	98.62
5	400	30	2	36.40	79.34
6	650	30	2	28.86	94.58
7	400	60	2	33.87	69.86
8	650	60	2	25.33	89.69
9	315	45	1.5	37.55	67.87
10	735	45	1.5	29.86	98.99
11	525	20	1.5	31.67	79.88
12	525	70	1.5	26.64	95.53
13	525	45	0.66	25.67	71.49
14	525	45	2.34	29.55	78.04
15	525	45	1.5	33.28	88.98
16	525	45	1.5	31.88	87.22
17	525	45	1.5	30.71	86.85
18	525	45	1.5	31.25	83.43
19	525	45	1.5	30.45	88.25
20	525	45	1.5	31.34	89.56

3.3. Development of regression model equation

Correlation between the response surface and factors were developed using central composite design (CCD). The models were selected based on the highest order polynomials (Chaudhary and Balomajumder, 2014; Tan *et al*., 2008). Correlation coefficient and standard deviation were used to evaluate the fitness of the model developed. The closer the R^2 value to unity, the smaller the standard deviation, the better the model in predicting the response (Alam *et al.*, 2009; Obayomi et al., 2017).

Table 4: Analysis of variance (ANOVA) of yield on WTAC

Variable	**Values**
Standard deviation	1.08
Coefficient of variation (%)	3.58
R^2- Squared	0.9484
R^2- Adjusted	0.9019
R^2 -Pred	0.7391
Adeq. Precision	15.923
Mean	30.17

Table 5: Analysis of variance of MNZ adsorption on WTAC

Variable	**Values**
Standard deviation	2.98
Coefficient of variation (%)	3.57
R^2- Squared	0.9617
R^2- Adjusted	0.9272
R^2 -Pred	0.7601
Adeq. Precision	20.232

Mean	83.46

Table 4 shows that, the quadratic model for percentage yield has standard deviation of 1.08 and relatively high R^2 value of 0.9484 which is closes to the adjusted R^2 of 0.9019 and is in reasonable agreement with R^2–Predicted of 0.7391. "Adeq Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. The ratio of 15.923 indicates an adequate signal. This model can be used to navigate the design space. The result implies that the quadratic model can be employed to describe the relationship between response of the percentage yield and the interacting variables. The R^2 values of 0.9484 implies that 94.84 % yield is attributed to the factors been considered. Table 5 also shows that the quadratic model for percentage MNZ removal has a standard deviation of 2.98 and relatively high R^2 value of 0.9617 which is closes to the adjusted R^2 of 0.9272 and in reasonable agreement with the R^2–Predicted of 0.7601. "Adeq Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. The ratio of 20.232 indicates an adequate signal. This model can be used to navigate the design space. The result implies that quadratic model for response on percentage MNZ removal can be employed to describe the relationship between percentage removal and the interacting variables. The R^2 value of 0.9617 implies that 96.17 % MNZ removal efficiency.

Table 6: ANOVA on response surface quadratic model for yield by WTAC

source	sum of squares	df	mean square	F- value	p- value
Model	214.19	9	23.80	20.42	< 0.0001
A-Temperature	54.99	1	54.99	47.17	< 0.0001
B-Time	25.85	1	25.85	22.18	0.0008
C-IR	34.85	1	34.85	29.89	0.0003
AB	3.42	1	3.42	2.93	0.1176
AC	39.12	1	39.12	33.56	0.0002
BC	0.40	1	0.40	0.34	0.5708
A^2	5.20	1	5.20	4.46	0.0608
B^2	14.64	1	14.64	12.56	0.0053

C^2	34.81	1	34.81	29.86	0.0003
Residual	11.66	10	1.17	-	-

Table 7: ANOVA for response surface quadratic model for removal by WTAC

source	sum of squares	df	mean square	F- value	p- value
Model	2221.17	9	246.80	27.87	< 0.0001
A-Temperature	14975.40	1	1495.40	168.86	< 0.0001
B-Time	109.95	1	109.95	12.42	0.0055
C-I. Ratio	44.91	1	44.96	5.07	0.0480
AB	1.09	1	1.09	0.12	0.7332
AC	52.17	1	52.17	5.89	0.0356
BC	211.87	1	211.87	23.92	0.0006
A^2	28.04	1	28.04	3.17	0.1055
B^2	0.20	1	0.20	0.022	0.8848
C^2	286.46	1	286.46	32.35	0.0005
Residual	88.56	10	8.86	-	-

Table 6 gives the response surface quadratic model for the percentage yield. The Model F-value of 20.42 and “prob >F” less than 0.0500 implies that the model is significant. Values greater than 0.1000 implies that the model terms are not significant. There is only a 0.01 % chance that a “Model F-Value” this large could occur due to noise. The "Lack of Fit F-value" of 1.27 implies the Lack of Fit is not significant relative to the pure error. There is a 39.85 % chance that a "Lack of Fit F-value" this large could occur due to noise. In this case A, B, C, AC, B^2, C^2 are significant model terms while, AB, BC and A^2 are the insignificant model terms.

Table 7 gives the Model F-value of 27.87 which implies the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise. Values of "Prob > F" less than 0.0500 indicate model terms are significant. Values greater than 0.1000 indicate the model terms are not significant. The "Lack of Fit F-value" of 2.69

implies the Lack of Fit is not significant relative to the pure error. There is a 15.04 % chance that a "Lack of Fit F-value" this large could occur due to noise. Non-significant lack of fit is good -- we want the model to fit. In this case A, B, C, AC, BC, C^2 are significant model terms while AB, A^2 and B^2 are the insignificant terms. Therefore, the response surface model equation in their actual value is:

$$\% \text{ Yield} = 31.52 - 2.01A - 1.38B + 1.60C - 0.65AB - 2.21AC - 0.22BC + 0.60A^2 - 1.01B^2 - 1.55C^2 \quad (7)$$

$$\% \text{ Removal} = 87.35 + 10.46A + 2.84B + 1.81C + 0.37AB - 0.55AC - 5.15BC - 1.39A^2 + 0.12B^2 - 4.46C^2 \quad (8)$$

Removing the insignificant model terms, the quadratic models of percentage yield and removal becomes equations (9) and (10):

$$\% \text{ Yield} = 31.52 - 2.01A - 1.38B + 1.60C - 2.21AC - 1.01B^2 - 1.55C^2 \quad (9)$$

$$\% \text{ Removal} = 87.35 + 10.46A + 2.84B + 1.81C - 0.55AC - 5.15BC - 4.46C^2 \quad (10)$$

From eqution (9) and (10), table 8 is prepared. It comprises of predicted values of percentage yield and percentage MNZ removal as well as the residual of the responses. Table 8 shows the lowest- highest error (residual) for percentage MNZ removal as (0.13 – 9.59 %), and that of percentage Yield as (0.21 – 4.03 %). This also signifies the quality of the model in terms of predicting the responses.

Table 8: Actual- predicted value on the percentage yield and removal

	Activated Carbon Yield (%)			MNZ (%)		
Run	Actual	Predicted Y_1	Residual	Actual	Predicted Y_2	Residual
1	26.90	28.53	1.63	57.47	63.08	5.61
2	29.82	28.93	0.89	88.99	83.10	5.89
3	26.88	25.77	1.11	74.64	79.06	4.42
4	25.57	26.17	0.67	98.62	99.08	0.46
5	36.40	36.15	0.25	79.34	76.10	3.24
6	28.86	27.71	1.15	94.58	97. 92	3.24
7	33.87	33.39	0.48	69.86	71.48	1.62
8	25.33	24.95	0.38	89.69	93.30	3.67

9	35.55	33.52	4.03	67.87	76.89	9.02
10	29.86	29.50	0.36	98.99	97.81	1.18
11	31.67	31.88	0.21	79.88	84.51	4.63
12	26.64	29.12	2.48	95.53	90.19	5.34
13	25.67	28.36	2.69	71.49	81.08	9.59
14	29.55	31.60	2.05	78.04	84.70	6.66
15	33.28	31.51	1.77	88.98	87.35	1.62
16	31.88	31.51	0.37	87.22	87.35	0.13
17	30.71	31.51	0.80	86.85	87.35	0.50
18	31.25	31.51	0.26	83.43	87.35	3.92
19	30.45	31.51	1.06	88.25	87.35	0.90
20	31.34	31.51	0.51	89.56	87.35	2.21

3.4 Combined Effect of Factors

3.4.1 Combined effect of activation temperature and time at constant impregnation ratio

Figure 1, is a three-dimensional graph which shows an interaction between the time and temperature on percentage yield. An increase in any of the factors does not increase the percentage yield of WTAC.

Figure 2, shows that the combined effect is almost majorly as a result of the effect of activation temperature with time having small effect. It was also observed that percentage removal is significantly increased with increase in activation temperature. Similar three-dimensional plot were observed by Auta and Hameed, (2014) on the combined effect of temperature and time on adsorption capacity of Reactive blue 19 (RB 19).

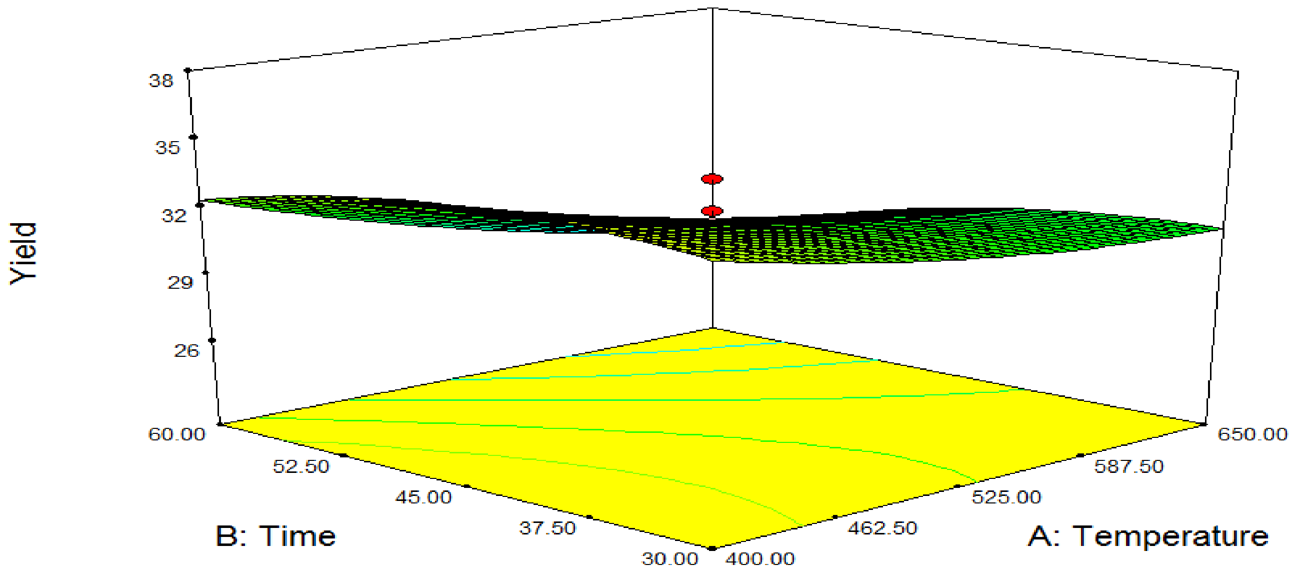

Figure 1: A three- dimensional response for combined effect of activation temperature and time on percentage yield of WTAC (X_1=A: Temperature, X_2= B: Time, Actual Factor C: Impregnation Ratio= 1.50, Yield: 25.33-37.55).

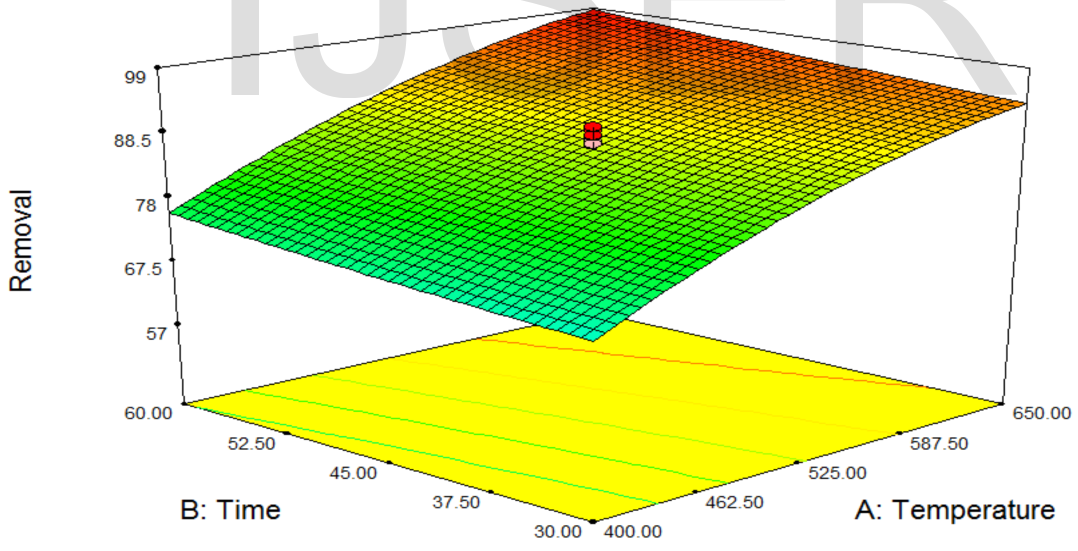

Figure 2: A three- dimensional response for combined effect of activation temperature and time on percentage removal of MNZ (X_1=A: temperature, X_2= B: time, Actual Factor C: impregnation ratio = 1.5, Removal: 98.99-57.87).

3.4.2 Combined effect of activation temperature and impregnation ratio at constant time

Figure 3, shows that combined effect is completely as a result of the single effect of impregnation ratio. Increase in activation temperature has no significant change on percentage yield. Similar result was reported by Mohammad *et al.* (2014) on the combined effect of temperature and concentration on adsorption capacity. Figure 4 shows that the combined effect is completely as a result of the double effect of activation temperature and impregnation ratio. It was also observed that percentage removal is significantly increased with increase in activation temperature. Similar three-dimensional plot were observed by Auta and Hameed, (2014) on the combined effect of temperature and impregnation ratio on adsorption capacity of Reactive blue 19 (RB 19).

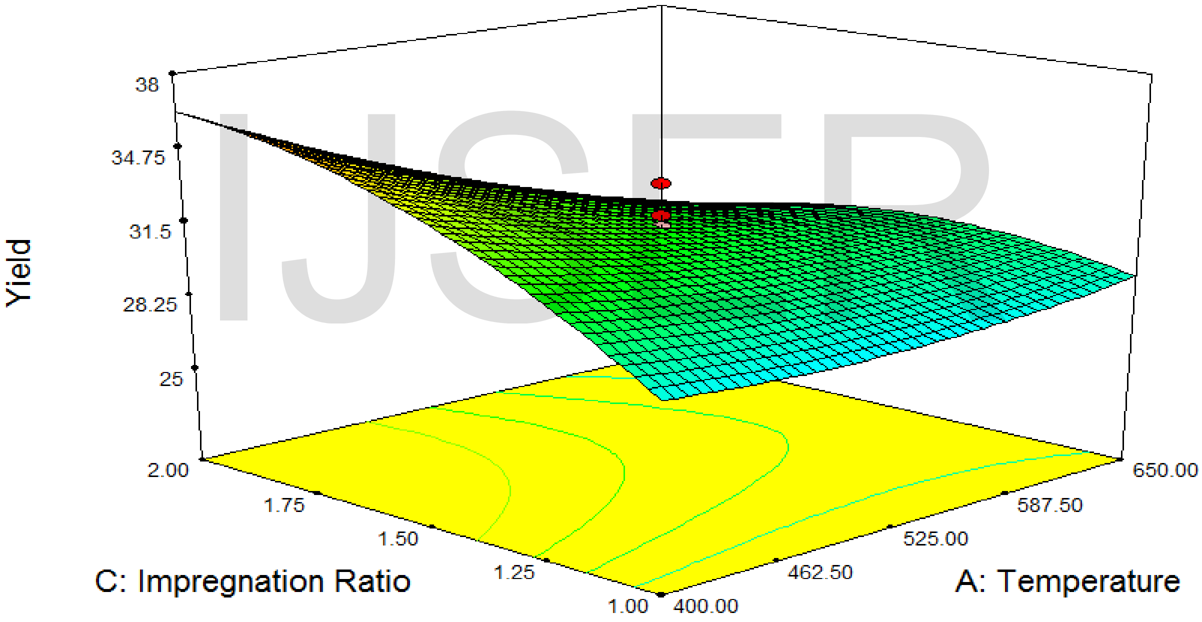

Figure 3: A three- dimensional response for combined effect of activation temperature and impregnation ratio on percentage yield of WTAC (X_1=A: temperature, X_2= C: impregnation ratio, Actual Factor C: time= 45, Yield: 25.33-37.55).

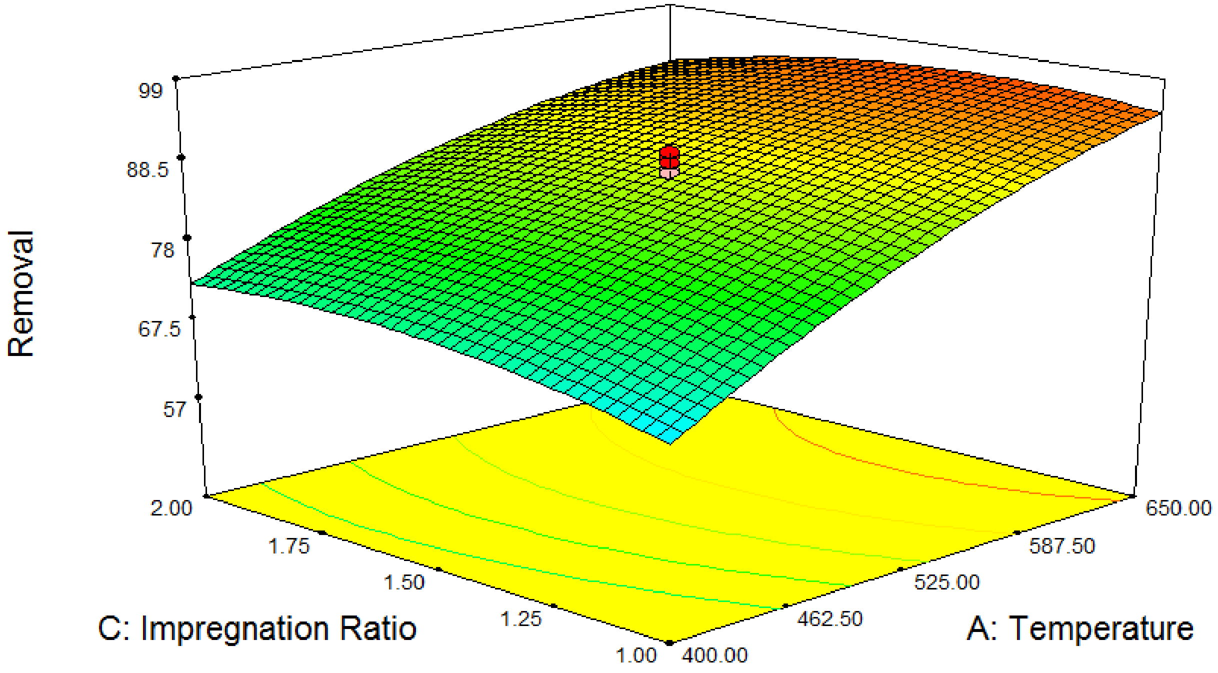

Figure 4: A three- dimensional response for combined effect of activation temperature and impregnation ratio on percentage removal of MNZ (X_1=A: temperature, X_2= C: impregnation ratio, Actual Factor B: time = 1.5, Removal: 98.99-57.87).

3.4.3 Combined effect of activation time and impregnation ratio at constant temperature

Figure 5, it was observed that the combined effect is majorly as a result of the effect of impregnation ratio while activation time has a slight effect. Percentage yield is significantly increased with increase in impregnation ratio. Percentage yield was also increase as time increases. Figure 6 shows the combined effect of activation time and impregnation ratio on percentage removal of MNZ. It was observed that the combined effect is completely as a result of the double effect of impregnation ratio with activation time having slight effect. Percentage removal of MNZ is significantly increased with increase in impregnation ratio and activation time.

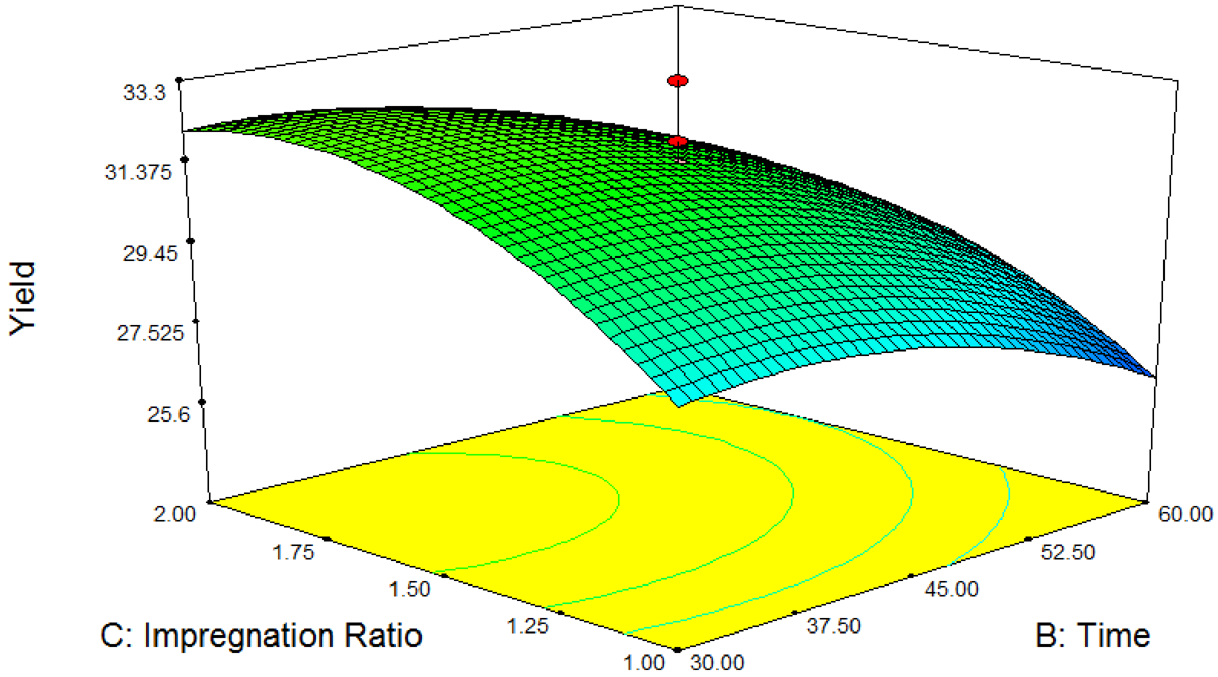

Figure 5: A three- dimensional response for combined effect of activation time and impregnation ratio on percentage yield of WTAC (X_1=B: time, X_2= C: impregnation ratio, Actual Factor A: temperature= 525, Yield: 25.33-37.55).

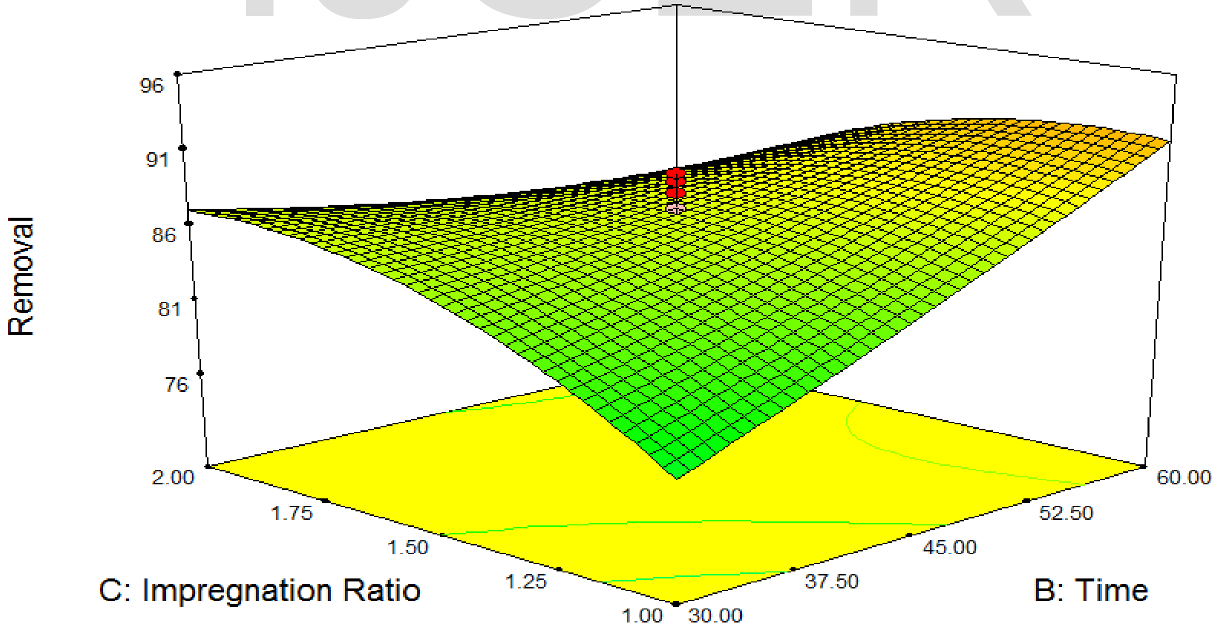

Figure 6: A three- dimensional response for combined effect of activation time and impregnation ratio on percentage removal of MNZ (X_1=B: time, X_2= C: impregnation ratio, actual factor A: temperature = 525, removal: 98.99-57.87).

The plot in figure 7 is the relationship of predicted values against actual values for percentage yield. It shows very minimal divergence of points from the diagonal indicating; that these response surface model equation for the yield can be used to adequately represent the interactions on the three factors. The plot in figure 8 shows the relationship of predicted against actual values for percentage MNZ removal. It shows minimal divergence of points from the diagonal indicating; that these response surface model equation for the MNZ removal can be used to adequately represent the interactions on the three factors. Figure 7 and 8 is a straight line graph of predicted and actual plot on percentage yield and percentage MNZ removal of WTAC respectively. From the graph, a close correlation is observed.

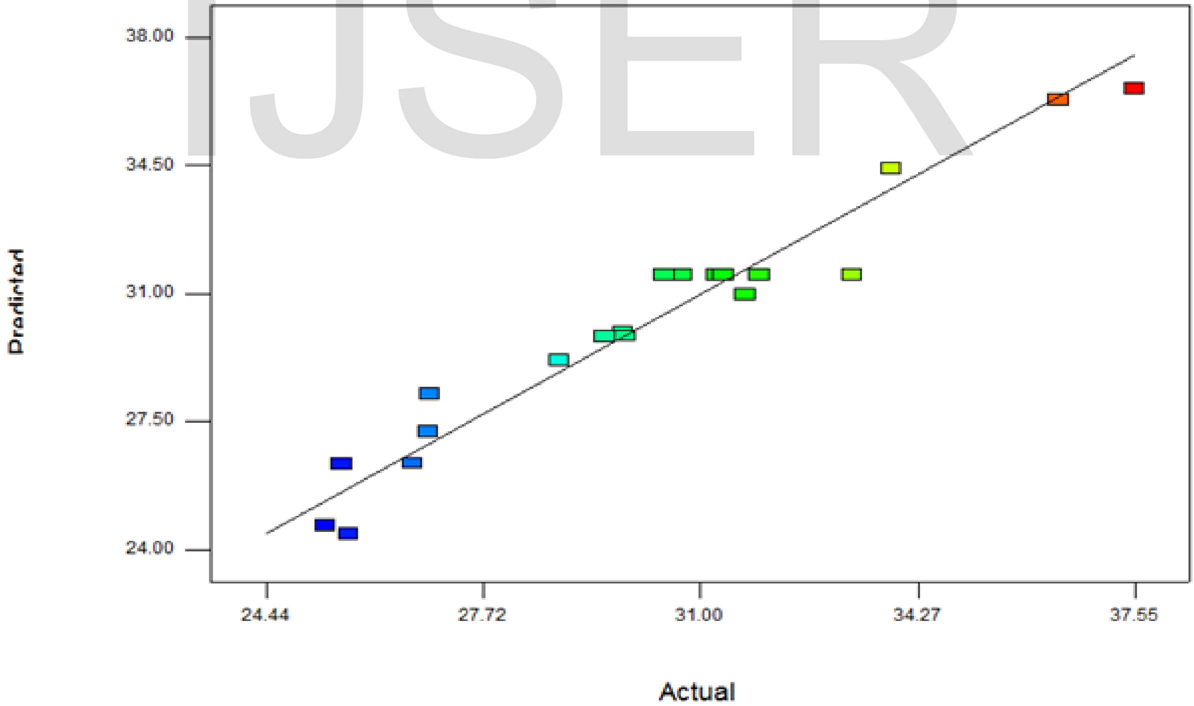

Figure 7: Predicted – Actual value plot on percentage yield of WTAC

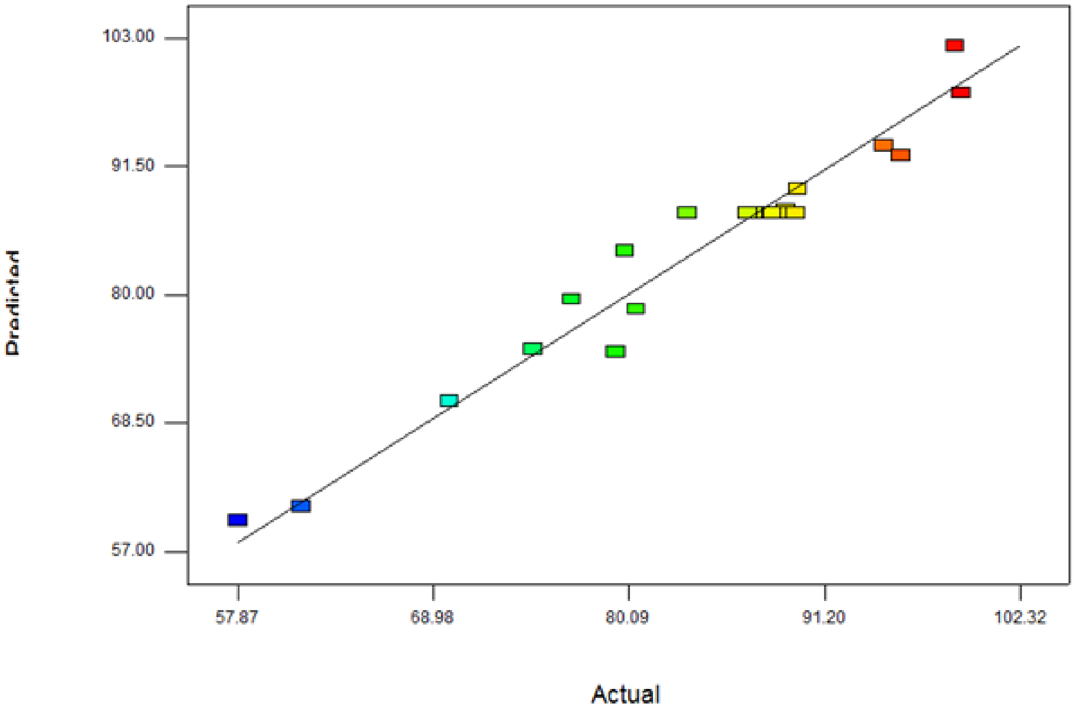

Figure 8: Predicted – Actual value plot on percentage adsorption for MNZ.

4. Conclusion

Central composite design was used to optimize the process of waste tea activated carbon for the removal of MNZ. Analysis of variance (ANOVA) shows a good agreement between theoretical analysis and experimental data. The validity of the model was verified by an experiment at the optimum conditions. The yield of 29.86 % which gave percentage MNZ removal of 98.99 % where obtained at the operating conditions of 735 °C activation temperature, 45 min activation time, and impregnation ratio of 1.5. Since the predicted and the actual experimental value obtained for the maximum adsorption of MNZ are within 95 % confidence intervals, the final model is considered valid and has satisfactory predictive ability. Considering the results, it can be concluded that RSM is an efficient statistical method for optimization of experimental conditions. The FTIR analysis showed the presence of several functional groups: N-H, O-H, C-H, C-O, C = O, C ≡ C, C= C, S = O, and Si-O-Si. WTAC can be used for the removal of MNZ from aqueous solution due to its high adsorptive capacity, availability, low cost.

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