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## MODELLING CARBON MONOXIDE CAPTURE THROUGH POST-COMBUSTION USING MONOETHANOLAMINE

Chukwu M.M.<sup>\*1</sup>, Igbokwe P.K.<sup>2</sup> & Nwabanne J.T.<sup>3</sup>

<sup>\*1,2&3</sup>Department of Chemical Engineering, NnamdiAzikiwe University, Awka, Nigeria.

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### ABSTRACT

There is need to solve the prominent issues of carbon monoxide hazards by promptly capturing it at the source. This paper presents the modelling and optimization analysis of the capture experimental results generated with independent use of Monoethanolamine solution as sorbents for Post-combustion capture of CO from exhaust of Yamaha EF1000. The Yamaha EF1000 was appropriately connected to a laboratory scale glass absorber column for counter current flow of the flue gas with the sorbent. Modelling and optimization of the process were carried out using Design Expert version 10, with concentration of sorbents, process time and sorbent flow rate as the process variables. From the results obtained, quadratic models were obtained as the best model for the capture for the sorbent, with CO of the exit flue gas as response. Optimum response value of 4.139% at the respective factors of 20.044g/L, 5.666mins and 223.848cm<sup>3</sup>/min for sorbent concentration, process time and sorbent flow rates, were obtained. The strong correlation of the statistical modelling and response surface methodology with the experimental outcome made it evident that the variability observed in the experimental data can be relied upon in the design of prototype equipment which will serve for the pilot performance of such capture process.

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### INTRODUCTION

There is no question that carbon monoxide is a pollutant with potential to harm all living things. But does CO also affect earth's climate? This will also be unravelled in the research. Unlike carbon dioxide, a compound that contains the same atoms as carbon monoxide is not known as a direct contributor to climate change. It does, however, play a role in this area and such roles can be investigated with the results of the sequestration.

Global warming is the increase in the average temperature of the earth. This effect is caused by anthropogenic greenhouse gases released to the atmosphere. The control of these greenhouse gases is arguably the most challenging environmental policy issue facing most countries. An approach that is gaining widespread interest is to control carbon oxides emissions by capturing them from fossil fuels, to continue to be used without contributing significantly to greenhouse warming.

Many researchers are aiming to develop new solvent technologies to improve the efficiency of the carbon oxides removal. Process model, simulation and evaluation are essential items to maximize the absorption process performance. Several researchers have modelled and studied the MEA absorption process (Abu-Zahra *et al.*, 2007), and most of their conclusions focused on reducing the thermal energy requirement to reduce the overall process expenses. This high-energy requirement makes the capture process energy intensive and costly considering the cost of procuring even the MEA. Also as part of an effort to study climate change, effects of carbon monoxide as a pollutant to atmosphere, have been given attention.

### MATERIALS AND METHODS

The various concentrations (in g/L) of the MEA solution were prepared from the stock solution by dilution, to obtain various concentrations of the sequestrant, for the carbon capture experiments from flue gas of Yamaha EF1000.

#### 2.1 Materials

##### a. Reagents Used

- Monoethanolamine stock solution
- Distilled and De-ionized water
- 0.1M NaOH Solution
- Phenolphthalein indicator

##### b. Apparatus/Equipment Used

- Laboratory scale absorber column set up
- Dosing pump (Model: JM 15.77/4.2)

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- Yamaha (Model: EF1000)
- Gas analyzer (Model: Ambro 2000)
- Analytical balance (Model: Adventurer Pro A35, Make: OHAUS)

### 2.2 Methods

#### a) CO capture with MEA Solution

Seventeen (17) sets of MEA solution respectively, were prepared to concentrations of 1.25, 11.88 and 22.50g/L, depending on the run concentration as suggested by the 3-level experimental design. The prepared sequestrants were made available for the sequestration experiments. The experimental glass absorber was mounted on a table, and the gas and liquid inlets and outlets were connected appropriately. The dosing pump (for the sequestrant transport) was then connected accordingly and the equipment connected to a power source and set to the required % stroke value (depending on the flow rate of the experimental run to be carried out), for the required optimal flow. Yamaha EF1000 was put on, and after about two minutes the exhaust gas was analyzed for the CO composition, then the flue gas exhaust was connected through to the absorber column gas inlet pipe. After this, the dosing pump was switched on for the sequestrant circulation through the column, and in a counter current flow with that of the entry flue gas. This CO capture process was monitored for a period of 5 - 10 minutes (depending on the experimental run performed), after which the gas analyzer Ambro 2000 was used to check for the CO composition of the exiting gas (lean gas) from the column.

#### b) Determination of CO composition in flue gas

The Gas Analyzer (Model: Ambro 2000) was put on and allowed to boot for about five minutes, after which the equipment was set on the program for the parameter to be tested. The equipment gas sensor was brought to the gas exit point of the absorber set up, in order to detect the CO composition of the flue gas. Also the CO composition of the flue gas coming out directly from Yamaha EF1000 exhaust was determined and recorded.

#### c) Optimization

Design Expert software was used in the Regression Statistical Analysis of the results for the MEA absorption processes. The experimental data were gathered in the required format for the statistical study, and the collated data were used in the analysis to generate the necessary statistical parameters useful in the statistical model development and optimization.

The process variables (X) used for the computer analysis are as follows:

$X_1$  = Concentration of MEA solution (g/L)

$X_2$  = Sequestration time (mins)

$X_3$  = Flow rate of sequestrant ( $\text{cm}^3/\text{min}$ )

While the response variable (Y) for the computer analysis is:

Y = CO composition of exit flue gas (%)

For the response variable; the optimum process conditions are the  $X_1$ ,  $X_2$ ,  $X_3$  values that yield the optimum (minimum) Y value.

#### d) Experimental Design

*Table 1: Experimental design code and boundary/limit for the factors (Independent variables)*

S/N	Variable	Code (X)	Boundary	
			Lower	Upper
1	Concentration of MEA solution (g/L)	$X_1$	1.25	22.5
2	Sequestration time (mins)	$X_2$	5	10
3	Flow rate of sequestrant ( $\text{cm}^3/\text{min}$ )	$X_3$	200	250

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## RESULTS AND DISCUSSION

### 3.1 Results for CO Capture using Monoethanolamine Solution

Table 2: Result for CO capture using MEA solution as absorber liquids (sequestrants)

S/N	Run	Block	X <sub>1</sub> (g/L)	X <sub>2</sub> (mins)	X <sub>3</sub> (cm <sup>3</sup> /min)	Y (%)
1	16	Block 1	1.25	5.00	225.00	5.67
2	12	Block 1	22.50	5.00	225.00	4.16
3	2	Block 1	1.25	10.00	225.00	6.12
4	6	Block 1	22.50	10.00	225.00	5.07
5	4	Block 1	1.25	7.50	200.00	5.98
6	1	Block 1	22.50	7.50	200.00	4.90
7	17	Block 1	1.25	7.50	250.00	6.52
8	10	Block 1	22.50	7.50	250.00	5.04
9	5	Block 1	11.875	5.00	200.00	5.73
10	15	Block 1	11.875	10.00	200.00	5.83
11	14	Block 1	11.875	5.00	250.00	5.59
12	9	Block 1	11.875	10.00	250.00	6.75
13	7	Block 1	11.875	7.50	225.00	4.48
14	3	Block 1	11.875	7.50	225.00	4.51
15	8	Block 1	11.875	7.50	225.00	4.58
16	13	Block 1	11.875	7.50	225.00	4.59
17	11	Block 1	11.875	7.50	225.00	4.48

In the result Table 2 for the experimental runs, for the MEA solutions, absorber liquid/sequestrant concentrations within the range of 1.25g/L and 22.50g/L were used.

Again for the preliminary runs set up, after some initial trials have been carried out, the optimal sequestrant flow rate were found to be within the range of 200cm<sup>3</sup>/min to 250cm<sup>3</sup>/min, for the successful operation of the laboratory scale glass absorber without flooding. And the pump settings at which the aforementioned optimal liquid flow were established were 30%, 40% and 50% strokes, which were subsequently adopted in the design of experiments.

Box bekhen design for three factors and three levels was used in the design of experiments for the laboratory experimental runs, producing seventeen (17) runs. The factors (independent variables; X<sub>1</sub>, X<sub>2</sub> and X<sub>3</sub>) were taken to be the concentration of sequestrant (g/L), sequestration time (mins) and flow rate of sequestrant (cm<sup>3</sup>/min) respectively, while the response (Y) was taken to be the CO compositions of the exiting flue gas from the absorber column. For the factors (with box bekhen design), experimental design data points of 1.25, 11.88 and 22.50g/L were used for the concentration of the sequestrant, data points of 5, 7.5 and 10mins for the sequestration time, and for the flow rate of sequestrant, 200cm<sup>3</sup>/min, 225cm<sup>3</sup>/min and 250cm<sup>3</sup>/min were used.

The CO compositions of the flue gas from the absorber column when the MEA solution was used as sequestrant were found to be within the values of 4.16% and 6.75%. However, the CO composition of the flue gas exiting directly from the Yamaha EF1000 was measured by the Ambro 2000 Gas Analyzer, and found to have a value of 8.2%. According to Gale (2002), domestic power generating sets could form part of the well-known potential carbon oxides emission sources from which it is a possibility to capture and store carbon. Gielenand Moriguchi

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(2003) reiterated that absorption (or scrubbing) remains one of the effective approaches to carbon capture from flue gases of both large and medium scale industrial plants.

Mimura et al. (1995) showed immense concern through their research on energy saving technology for flue gas carbon oxides recovery and steam system in power plant. Their effort was geared towards minimizing the energy requirement for the capture process which happens to be a major advantage of the quest for this work. Moser et al. (2009) worked on enabling post combustion capture optimization with the pilot plant project at Niederaussem. Even though they used conventional liquid for capture, their optimization approach involved setting out process factors and performing the capture at the various defined factor combinations, which is an analogue of what has been done in this work.

Rao and Rubin (2002) after working on a technical, economic and environmental assessment of amine-based carbon capture technology for power plant greenhouse gas control, agree to the fact that the exercise with the chemical based liquid even though technically effective and environmental assessment commendable, the economics of the process remains a factor of concern.

From the results obtained from the experiments, the effect of each of the factors were better understood when statistical analysis were done based on the results with which statistical models were developed and response surface methodology carried out for the purpose of optimizing the capture process conditions under investigation in this work. These and their respective discussions are outlined in the subsequent section 3.2 that follows.

### 3.2 Results for the Statistical Analysis, Modelling and Optimization

*Table 3: ANOVA for Reduced Response Surface Quadratic model*

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F	
A-Conc. of MEA solution	16.96	1	16.96	26.63	0.0006	
B-Sequestration time	97.3	1	97.3	152.8	< 0.0001	
C-Flowrate of sequestrant	10.55	1	10.55	16.57	0.0028	
AB	4.51	1	4.51	7.08	0.026	
AC	3.63	1	3.63	5.7	0.0407	
A <sup>2</sup>	3.79	1	3.79	5.96	0.0373	
C <sup>2</sup>	37.12	1	37.12	58.28	< 0.0001	
Residual	5.73	9	0.64			
Lack of Fit	2.83	5	0.57	0.78	0.6111	not significant
Pure Error	2.9	4	0.72			
Cor Total	181.04	16				

Std. Dev.	0.8	R-Squared	0.9683
Mean	23.46	Adj R-Squared	0.9437
C.V. %	3.4	Pred R-Squared	0.8877
PRESS	20.33	Adeq Precision	19.245
-2 Log Likelihood	29.76	BIC	52.43
		AICc	63.76

The Model F-value of 39.33 implies the model is significant. There is only 0.01% chance that an F-value this large could occur due to noise. Values of "Prob> F" less than 0.0500 indicate model terms are significant. In this

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case A, B, C, AB, AC, A<sup>2</sup>, C<sup>2</sup> are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve your model.

The "Lack of Fit F-value" of 0.78 implies the Lack of Fit is not significant relative to the pure error. There is a 61.11% chance that a "Lack of Fit F-value" this large could occur due to noise. Non-significant lack of fit is good because it means the model would fit.

The F-value of the independent variables (sequestrant concentration, sequestration time, flow rate of sequestrant) was estimated as 26.63, 152.80, and 16.57 respectively, showing that the single effects of the independent variables are significantly high on the response (CO composition of the exit flue gas stream). The CV called coefficient of variation which is defined as the ratio of the standard deviation of estimate to the mean value of the observed response is independent of the unit. It is also a measure of reproducibility and repeatability of the models (Chen *et al.*, 2010; Chen *et al.*, 2011). The calculations indicated the CV value of 3.4% which illustrated that the model can be considered reasonably reproducible (because its CV was not greater than 10%) (Chen *et al.*, 2011). The signal to noise ratio which is given as the value of the adequacy precision is 19.245. This indicates that an adequate relationship of signal to noise ratio exists and model can be used to navigate the design space.

The selected model in terms of the coded and actual values is given in the equation below:

$$\text{CO composition of exit flue gas} = 4.528 + 0.3275A - 0.64B + 0.1825C + 0.115AB + 0.265AC - 0.100BC + 0.546A^2 + 0.181B^2 + 0.901C^2$$

The equation in terms of coded factors can be used to make predictions about the response for given levels of each factor. By default, the high levels of the factors are coded as +1 and the low levels of the factors are coded as -1. The coded equation is useful for identifying the relative impact of the factors by comparing the factor coefficients.

In terms of Actual values, the model terms are given by:

$$\text{CO composition of exit flue gas} = +87.27409 - 2.184812 * A - 0.04608 * B - 0.66875 * C + 0.004329 * A * B + 0.00424 * A * C - 0.000376 * B * C + 0.08736 * A^2 + 0.001603 * B^2 + 0.001442 * C^2$$

The equation in terms of actual factors can be used to make predictions about the response for given levels of each factor. Here, the levels have been specified in the original units for each factor. This equation should not be used to determine the relative impact of each factor because the coefficients are scaled to accommodate the units of each factor and the intercept is not at the centre of the design space.

The response values obtained by inserting the independent values are the predicted values of the model. These values are compared to the actual and experimental values. The result of this comparison is shown in figure 1 below.

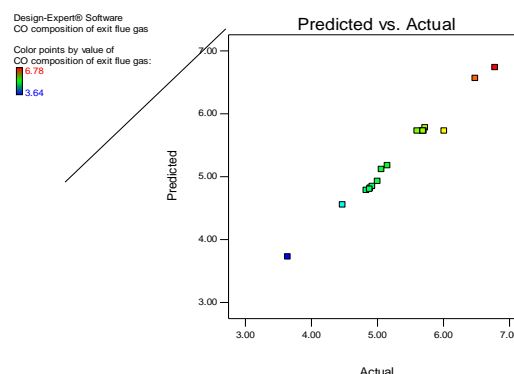


Figure 1: Linear correlation between vs. actual values for effect of CO reduction using MEA Solution 'CO Composition of Exit Flue Gas' as Response

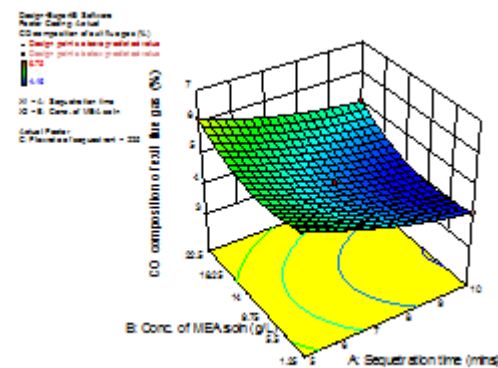


Figure 2: Surface Response Plot (MEA Solution) – Using Concentration of MEA Solution (A, g/L) and Sequestration time (B, mins) as Factors, with ‘CO Composition of Exit Flue Gas’ as Response

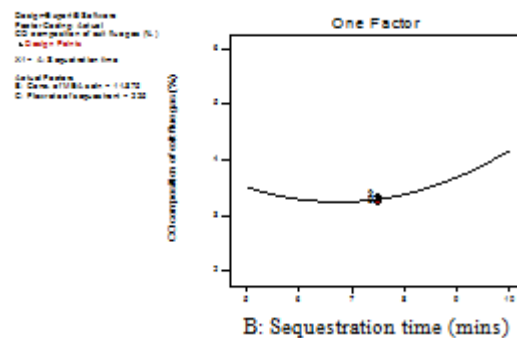


Figure 3: One Factor plot of CO comp. against sequestration time

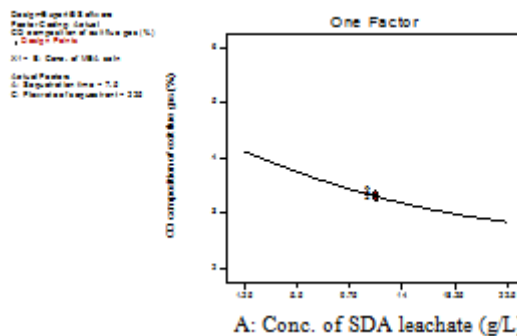


Figure 4: One Factor plot of CO comp. against concentration of MEA solution

As can be seen in figure 1, the actual values were distributed relatively near to the predicted value line, showing that there is a good correlation between the actual and the predicted values. This observation shows that the central composite design (CCD) is well fitted into the model and thus can be used to perform the optimisation operation for the process.

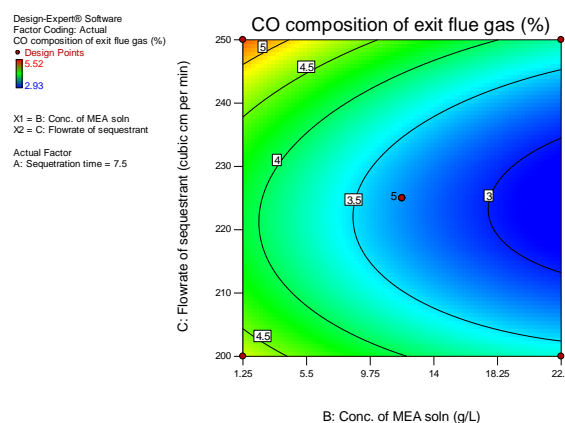
In order to visualize the relationship between the experimental variables and the response, and to study individual and interaction effects of the three factors consisting of the MEA conc., sequestration time, and flowrate of sequestrant. Response surfaces and interaction plots were generated from the quadratic model, as shown in figures 2 - 4. These figures illustrate the response of different experimental variables and can be used to identify the major interactions between the variables.

Figures 2 to 4 again show the 3D plot and its corresponding interactions for the effects of MEA concentration on composition of CO in the exit flue gas stream. The graphs show that the least recorded value for CO in the exit

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streams occur between 16.3g/L and 11.8g/L MEA concentration, which is in accordance with the model. As the MEA concentration is increased from 9.1g/L to 15.6g/L, the amount of CO in the exit stream steadily decreases from 22.40% to 20.73% as seen in figure 4. This is similar to the report of several researchers (Moser *et al.*, 2009) who agree that increase in MEA solution concentration results in decrease in CO composition of the exit flue gas stream. It is evident that the MEA concentration has a significant effect on the response. Increasing the MEA concentration beyond 19.1g/L at all levels of sequestration time results in the increase of CO composition of the exit flue gas stream. As expected, the CO levels decreased linearly with increase in sequestration time (figures 3). This effect is independent of the MEA solution concentration as seen on the 3D plot of figure 2.

The effect of sequestrant flowrate and concentration of MEA on the CO composition of exit flue gas is shown on the 2D contour plot of figure 5.



**Figure 5: 2D contour plot for the effect of sequestrant flowrate and conc. of MEA solution on CO composition of exit flue gas.**

This process was carried out at sequestration time of 7.5 minutes. The least CO composition of the exit flue gas stream is found to be 3% at flow rate of 220 cubic centimeters per minute and 21g/L MEA concentration. Between 200 - 222cm<sup>3</sup>/min flowrate, at all values of MEA concentration, the CO composition decreased steadily as seen on isolines of the 2D contour plot. At higher values of flowrate (225cm<sup>3</sup>/min - 250cm<sup>3</sup>/min) the CO composition increased steadily from 3.0 - 4.5% at 7.5 g/L MEA concentration.

Tables 4 and 5 below outline the constraints and solutions for the optimisation results of the sequestration with MEA solution.

**Table 4: Optimisation constraints values for factors and responses**

Name	Goal	Lower Limit	Upper Limit	Lower Weight	Upper Weight	Importance
A: Conc. of MEA solution	is in range	1.25	22.5	1	1	3
B: Sequestration time	is in range	5	10	1	1	3
C: Flowrate of sequestrant	is in range	200	250	1	1	3
CO composition of exit flue gas	minimize	4.16	6.75	1	1	3

*Table 5: Optimisation solutions values for factors and responses*

Number	Conc. of MEA	Sequestration time	Flowrate of sequestrant	CO composition of exit flue gas	Desirability
1	20.044	5.666	223.721	4.139	1 Selected
2	21.101	6.521	223.848	4.027	1
3	21.615	6.319	222.546	4.023	1
4	18.958	6.500	221.667	4.122	1
5	20.17	6.638	216.988	4.148	1
6	21.615	6.319	227.454	4.016	1
7	22.146	5.417	229.167	4.095	1
8	21.709	5.267	223.415	4.152	1
9	20.028	6.834	218.681	4.120	1
10	21.069	7.262	222.064	4.078	1

Table 5 shows the obtained and selected optimum values for the capture with MEA solution have CO composition of exit flue gas value of 4.139% as optimum (i.e minimum), at sequestrant concentration of 20.044g/L, sequestration time of 5.666mins and sequestrant flow rate of 223.848cm<sup>3</sup>/min respectively.

### CONCLUSION

There was commensurate decrease in the CO composition of the flue gas during the carbon capture with the MEA solution just as any other conventionally used chemicals. From the experimental results, it was clear the sequestrant concentration, process time and sequestrant flow rate, all significantly affected the reduction in the CO composition of the flue gas, at different levels depending on the variations.

Quadratic model best described the carbon capture process, with the MEA solution. The strong correlation of the statistical modelling and response surface methodology with the experimental outcome made it evident that the variability observed in the experimental data can be relied upon in the design of prototype equipment which will serve for the pilot performance of such capture process. To a large extent, there is confidence in the use of MEA solution as a good sequestrant for the capture of carbon from flue gas, having obtained good optimal result with its use, like other chemicals conventionally used as aforementioned.

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