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Statistical Design and Kinetic Studies of CO_2Capture Process from Stream of Gas Mixture at Elevated Temperature using Composites of Nanoparticles

[1]Ojong, Ojong Elias; [2]Benibo, Preniyobo Diepriye; [3]Wosu, Chimene Omeke
[1]Department of Chemical and Petrochemical Engineering, Faculty of Engineering and Technology, University of Calabar, Calabar, Cross River State, Nigeria.
[2]Department of Chemical/Petrochemical Engineering, Faculty of Engineering, Rivers State University, Nkpolu Oroworukwo, Port Harcourt, Rivers State, Nigeria
[3]Department of Chemical Engineering, Faculty of Engineering, Federal University Otuoke, Bayelsa State, Nigeria

Abstract

Chitosan and clay materials of 50:50 ratio, produced and characterized from periwinkle shells and clay soil, was applied to capture CO_2 from gas mixtureat elevated temperatures (50°C to 500°C) in a fixed bed column of dimension length 1.5m, and internal diameter 0.02m. The composition of gas mixture was methane (0.003), ethane (0.002), hydrogen (0.05), CO_2 (0.15), water vapour (0.02), and nitrogen (0.76) of condition of pressure 49kPa, temperature 500°C, and flow rate 75L/min from exhaust column into the fixed bed column for adsorption process. Statistical analysis, mainly analysis of variance (ANOVA) on the factors affecting adsorption processto determine maximum adsorption of CO_2.The kinetic and isotherm models were adopted and simulations performed from the experimental data to determine suitable adsorption parameters for the process. The coefficient of determination of 0.992 showed that the quadratic model is best suitable for the combination of factors and fitted well, while the ANOVA result gave significant difference among the bed height, temperature and time and their combinations. The result revealed that the adsorption process best followed Elovich kinetic and fitted well in the Freundlich multilayer adsorption isotherm with parameters 0.0187mg/g.hr, 175.932, and 1.5 for the kinetic constant, adsorption constant, and intensity respectively. The regression results of the factors analysis showed that the model R^2 of 0.992 is more reliable and fitted well than the adjusted R^2 of 0.985 and the predicted R^2 of 0.965.

Keywords: Statistical design, kinetic studies, CO2 capture, Chitoasan/clay materials, adsorption

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I. Introduction

The burning of fossil fuels for electricity generation in domestic and industrial settings releases pollutants like CO_2, NOx, SOx, dust, and wastewater into the atmosphere. This carbon dioxide emission is a primary contributor to global warming and climate change due to the resulting greenhouse gas buildup (Spliethoff, 2010; Sarkar, 2009; Yang *et al*., 2008).

To reduce CO_2 emissions, researchers have developed various technologies, including: Improving energy efficiency, Utilizing renewable energy sources (solar, wind, hydropower, biomass, nuclear), Increasing green hydrogen production, Carbon capture and storage (CCS) technology, which involves: Capturing CO2 from industrial sources, Transporting it to underground storage sites, Long-term isolation or conversion into useful products.

These efforts aim to mitigate greenhouse gas emissions and combat climate change (Nord &Bolland, 2020; Madejski, 2018; Madejski*et al*., 2016; ETP, 2020; Stewart *et al*., 2005).

CO_2 capture can be achieved through various methods, including: Pre-combustion capture, Post-combustion capture, Oxy-combustion capture. Among these, adsorption is a widely used technology, alongside absorption, membrane separation, and cryogenics. Effective adsorbents with high efficiency are crucial for optimal performance. Examples of adsorbents include: zeolite, metal oxides, mesoporous silica, monoethanolamine and activated carbon. These materials play a key role in capturing CO_2 emissions (Shijaz*et al*., 2017; Mukherjee *et al*., 2015; Wanga*et al*., 2017; Yong *et al*., 2002; Saha*et al*., 2007; Yates *et al*., 2000).

The limitations of existing CO_2 capture technologies, such as high costs and inadequate performance, have driven the development of new approaches utilizing novel, locally sourced materials. One promising solution is chitosan/clay nanoparticles, which offers several advantages including: Affordability, Environmental sustainability, Biodegradability, Biocompatibility, toxicity, High effectiveness in removing CO_2 from flue gas at high temperatures. These locally sourced materials provide a potential breakthrough in CO_2 capture technology (Ojong *et al*., 2025).

This study focused on statistical design and kinetic studies of CO_2 capture process from stream of gas mixtures at elevated temperature using nanoparticle composites as adsorbents in an adsorption column. The adsorption kinetics and isotherm models were developed and simulated to predict the suitable adsorption parameters required for optimum dioxide capture during the process.

II. Materials and Method

The materials utilized in this research were obtained in valid literatures and experimental data obtained from carbon (iv) oxide adsorption from flue gas mixture at elevated temperature using composites of nanoparticles by Ojong *et al*., 2025, the research methodology quantitative and analytical. The procedures adopted includes;

i. Develop the adsorption kinetics and isotherm models for the adsorption of carbon dioxide from flue gas.

ii. Simulate and validate the models and simulation results with the experimental data obtained from Ojong *et al*., 2025.

2.1 Adsorption Kinetics Models

The pseudo-first-order kinetic models are applied in this work to explain the relationship between the rate of the sorption sites of the adsorbents occupied and the number of the unoccupied sites. It states that from the Lagergrens equation (Zhou *et al*., 2011) as the rate of adsorption of CO_2 in equilibrium concentration and concentration at time, t,

$$\frac{dq}{dt} = k_1(q_e - q) \qquad (1)$$

Integrating equation (1) with boundary condition of 0 and q_t when t is 0 to t, gives,

$$\int_0^{q_t} \frac{dq}{q_e - q} = k_1 \int_o^t dt$$

$$-\ln[q_e - q]_o^{q_t} = k_1 t$$

$$\ln q_e + \ln(q_e - q_t) = -k_1 t$$

$$\ln(q_e - q_t) = -k_1 t + \ln q_e \qquad (2)$$

Plot of $\ln(q_e - q_t)$ vs t gives slope of $-k_1$ and intercept of $\ln q_e$.

where, q_e is amounts of CO_2 adsorbed by the adsorbent at equilibrium (mg/g)

$q_t$is concentration of CO_2 adsorbed by the nanoparticle composite at particular time, t (mg/g)

$k_1$is Rate constant of adsorption (hr^{-1}).

The Pseudo-second-order kinetics model is used to describe the adsorption of CO_2 from flue gas is given according to Ho, (2004) as,

$$\frac{dq}{dt} = k_1(q_e - q)^2 \qquad (3)$$

Solving equation (3) by integrating with boundary conditions and further simplification yields

$$\int_{q=0}^{q_t} \frac{dq}{(q_e - q)^2} = k_2 \int_o^t dt$$

$$\frac{1}{q_e - q_t} \int_o^{q_t} = k_2 t$$

$$\frac{1}{q_e - q_t} - \frac{1}{q_e} = k_2 t$$

$$\frac{q_e - (q_e - q_t)}{(q_e - q_t)q_e} = k_2 t$$

$$q_t = k_2 t\,(q_e^2 - q_e q_t)$$

$$q_t = k_2 t\, q_e^2 - k_2 t q_e q_t \qquad (4)$$

Divide equation (4) throughout by $q_e^2 q_t k_2$ yields.

$$\frac{t}{q_t} = \frac{t}{q_e} + \frac{1}{q_e^2 k_2} \qquad (5)$$

Model (5) is simplified and linearized pseudo-second order kinetics model for adsorption of CO_2 from flue gas using nanoparticle composite produced from chitosan/clay materials.

where, k_2 = Pseudo-second-order rate constant (g/mg.hr), t = measured time of adsorption (hr)

The plot of t/q_t vs t gives simple linear plot with slope as $1/q_e$ and intercept in $1/q_e^2 k_2$ for the calculation of $q_e$and k_2.

The Elovich Kinetic model has been of used for the chemisorption of gases onto heterogeneous surfaces and solid system and has now found useful for the adsorption of pollutants from gas mixture (Yaun *et al*., 2015). It describes the second-order-kinetics assuming that the solid surface has heterogeneous energy and no mechanism for the adsorption (Mezenner & Bensmai, 2009). According to Qiu *et al.*, (2009), Elovich Kinetic model is defined as,

$$\frac{dq}{dt} = a\exp(-\varphi q) \qquad (6)$$

where, a is adsorption constant, and φ is initial rate of adsorption (mg/g hr)

Using boundary conditions of: q =0 at t = 0 and q = q_t at t = t, equation (6) is integrated to give, $\int_{q=0}^{q_t} \frac{dq}{\exp(-\varphi q)} = a\int_o^t dt$

$$\int_o^{q_t} {}^{dq}/_{e^{-\varphi q}} = at$$

$$1/_{\varphi}\, e^{\varphi q} \int_o^{q_t} = at$$

$$\frac{1}{\varphi}\{e^{q_t-1}\} = at$$

$$e^{q_t-1} = \varphi at$$

$$q_t = \frac{1}{\varphi}\{\ln(\varphi a) + \mathrm{Int}\}$$

$$q_t = \frac{1}{\varphi}\,\mathrm{Int} + \ln(\varphi a) \qquad (7)$$

Model (7) is the linear form of model (6) and plot of q_t vs Int gives a slope of $\frac{1}{\varphi}$ and intercept of $\ln(\varphi a)$ where, φ and a, are determinant Energy (b) for chemisorption in (g/mg).

Equation (7) is transformed linearly (Yakout & Elsherif, 2010) to give,

$$q_t = \frac{\ln(\varphi a)}{b} + \frac{1}{b}\ln t \qquad (8)$$

And plot of q_t vs lnt gives b and φ from slopes and intercept respectively.

Weber & Morris, (1963) developed the intra-particle diffusion kinetics model which has been effective in many cases of adsorption of CO_2 and expressed according to Pasavant *et al.*, (2006) as,

$$q_t = k_d\sqrt{t} \qquad (9)$$

where, q_t is Amount of heavy metal absorbed at time, t (mg/g), k_d is Weber and Morris intra-particle diffusion rate constant (mg/g.hr$^{0.5}$), and t is time of adsorption (hr).

Plot of q_t vs $t^{1/2}$ gives a slope equivalent to k_d and the linearity of the plot is an indication that the adsorption will follow intra-particle diffusion and the linear plot must pass through the origin else, the adsorption does not follow intra-particle diffusion (Pasavant *et al.*, 2006; Ujile, 2013).

2.2 Adsorption Isotherm Models

The adsorption isotherm for the adsorption of CO_2 from flue gas mixture at elevated temperature in a fixed-bed column was investigated using the Langmuir, Freundlich, Temkin and Dubinin-Radush-Kevich Isotherm models.

Langmuir Isotherm

The Langmuir model describes monolayer sorption on district localized adsorption sites. It indicates no transmigration of the adsorbate in the plane of the surfaces and assumes uniform energies of monolayer sorption onto the sorbent surface (Balouch *et al.*, 2013). Accordingly, the Langmuir Isotherm model is expressed (Balouch *et al.*, 2013) as,

$$\frac{C_e}{C_{ads}} = \frac{1}{Q_b} + \frac{C_e}{Q} \qquad (10)$$

where, C_{ads} is the amount of CO_2 and other ions adsorbed at equilibrium, Q_B is sorption constant (mg/l) at a given temperature, related to the energy of sorption, Q is maximum sorption capacity (mg/g).

(Dada *et al.*, 2012), expressed isotherm model of Langmuir as,

$$q_e = \frac{Q_m k_L C_e}{1 + k_L q_e} \qquad (11)$$

where, C_e is equilibrium constant (mg/l), Q_m is maximum monolayer coverage capacity (mg/g), and K_L is Langmuir Isotherm constant (l/mg).

A dimensionless constant or separation factor (R_L) is represented as (Foo & Hameed, 2010)

$$R_L = \frac{1}{1 + k_L C_o} \qquad (12)$$

where, $R_L$is favourable nature of the absorption process described where $R_L > 1$ is unfavourable, $R_L = 0$ is linear, $0 < R_L < 1$ is favourable, $R_L = 0$ is irreversible (Yaun *et al.*, 2015; Foo & Hameed, 2010).

A plot of $1/_{q_e}$ vs $1/_{C_e}$ yielded a straight-line graph with the value of Q_m and k_L calculate from slope and intercept respectively.

$$1/_{q_e} = \frac{1}{Q_m k_L}\left(1/_{C_e}\right) + \frac{1}{Q_m} \qquad (13)$$

Model (13) is the linearized version of model (12)

Freundlich Isotherm

The Freundlich Isotherm is applied to describe adsorption processes that occurs on heterogeneous surfaces and active sites with different energies based on multilayer adsorption and equilibrium (Balouch *et al*., 2013; Boujelbem *et al*., 2013).

The isotherm model is expressed as,

$$q_e = k_f C_e^{1/n} n > 1 \tag{14}$$

Linearizing the model (Dada *et al*., 2012; Huston & Yang, 1997) give,

$$\ln q_e = {}^{1}/_{n} \ln c_e + \ln k_f \tag{15}$$

where, k_f is Freundlich Isotherm constant, n is adsorption intensity, $q_e$is equilibrium concentration of adsorbate (mg/l), $C_e$is the amount of CO_2 adsorbed at equilibrium. Plotting $\ln q_e$versus $\ln c_e$ yielded a straight line and n and k_f are calculated from the slope and intercept, respectively.

Temkin Adsorption Isotherm

The Temkin adsorption Isotherm contains a factor that considers the interaction between adsorbates (Dada *et al*., 2012). The model assumes that the heat of adsorption of all molecules in the layer will decrease linearly rather than logarithmically with coverage at average concentrations (Data *et al*., 2012; Foo and Hameed, 2010;Tempkin&Pyzher, 1940). The heat of adsorption is characterized by a uniform distribution of binding Energies up to some maximum binding energy (Foo and Hameed; 2010). The Tempkin Isotherm Model is expressed as,

$$q_e = \frac{R_T}{b_T} \ln(A_T C_e) \tag{16}$$

$$q_e = \frac{R_T}{b_T} \ln A_T + \frac{R_T}{b_T} \ln c_e \tag{17}$$

$$q_e = B \ln A_T + B \ln c_e \tag{18}$$

where, $B = \frac{RT}{b_T}$; A_T is Temkin Isotherm equilibrium binding constant (l/g), b_T is Tempkin Isotherm constant, R is universal gas constant (8.314 J/mol K), T is Temperature at 298K and B is Constant related to the heat of sorption (J/mol).

A_T and b_T are determined from the intercept and slope, respectively of the plot of $q_e$versus$\ln c_e$.

Dubinin-Radushkevich Adsorption Isotherm

The isotherm described the adsorption mechanism with a Gaussian energy distribution of the heterogeneous surface (Dada *et al*., 2012; Dabrowski, 2001). This however does not assume homogeneous surface or constant sorption potential and helps to determine the apparent energy of adsorption (Al-Auber, 2011). The model is expressed as,

$$\ln q_e = \ln q_s - \beta \varepsilon^2 \tag{19}$$

where, q_e is the number of ions adsorbed at equilibrium (mg/g), β is Dubinin-Radushkevich isotherm constant (mol^2/kJ^2), and ε is Polanyi potential which is equal to,

$$\varepsilon = RT \ln\left(1 + {}^{1}/_{C_e}\right) \tag{20}$$

The model is used to distinguish the physical and chemical absorption with its mean free energy, ε (kJ/mol), required to adsorbed CO_2 from its location in the sorption site to infinity.

where: $\boldsymbol{\varepsilon} = \frac{\mathbf{1}}{\sqrt{\mathbf{2\beta}}}$ (21)

III. Results and Discussion

The kinetic parameters and Isotherm constants for CO_2 adsorption from flue gas using Chitosan/clay nanoparticle composite were determined to show which one of them performed in the process.

3.1 Kinetic Parameters Determination

The kinetic study for the adsorption process was investigated with pseudo-1st and 2nd orders, Elovish kinetics and intra-particle kinetics. See Table 1 display the kinetic parameters for the adsorption process.

Table 1 displayed the kinetic parameters' values for the CAC process and showed that Elovich kinetic best describes the adsorption process as it gave the highest coefficient of determination of 0.991 compared to the other kinetics. This is because the data obtained from the experiment are well fitted to the Elovich kinetic, though all the kinetics are very useful to study the adsorption process.

Table 1: Kinetic and Isotherm Parameters for the CO_2 Adsorption Process at Elevated Temperature

S/N	Kinetic Parameter	K	q_e	Φ	Coefficient of Determination
1	1st order	2.926/hr	36.86mg/g	-	0.871
2	2nd order	0.0009g/mg.hr	25mg/g	-	0.922
3	Elovich	0.0187mg/g.hr	-	4.873e19J	0.991
4	Inter particle	74.831mg/g.hr$^{0.5}$	-	-	0.979

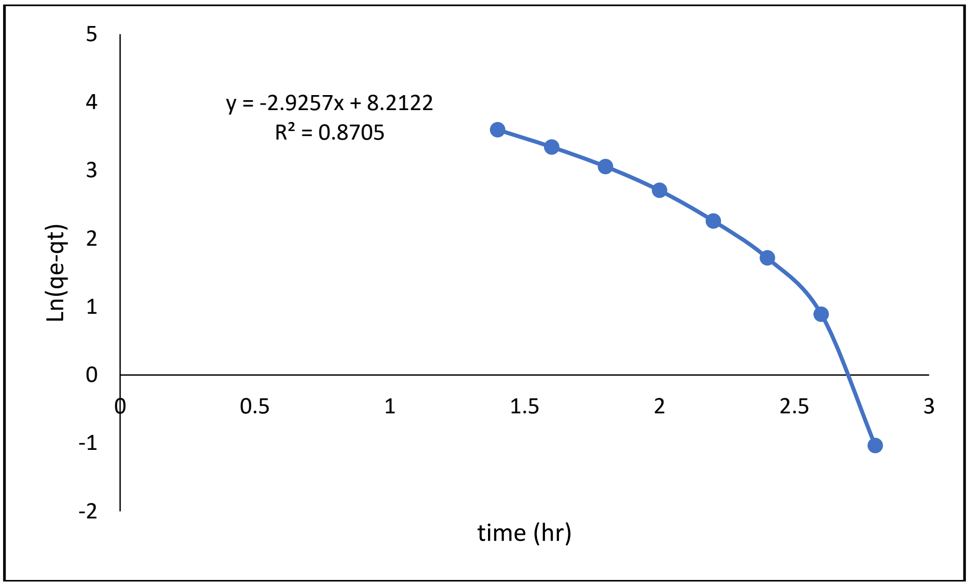

Figure 1: Pseudo-First Order Kinetics of the Adsorption CO_2 from Flue Gas using Chitosan/Clay Composites

The first order kinetic parameter k_1 is 2.9257hr^{-1} and the equilibrium concentration of the CO_2 adsorbed is q_e 36.87mg/g. The coefficient of determination of the adsorption of CO_2 is 0.871.

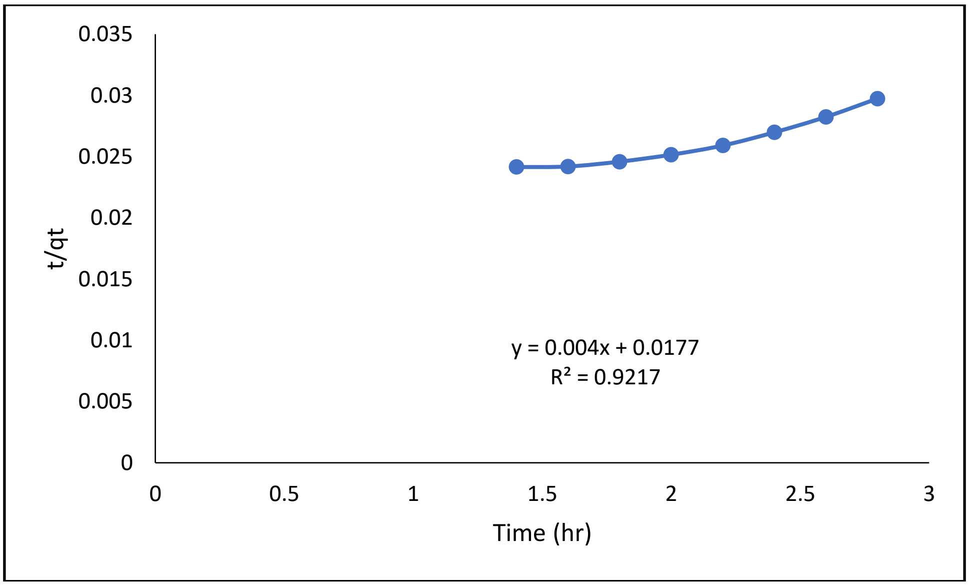

Figure 2: Pseudo-Second Order Kinetics of the Adsorption CO_2 from Flue Gas using Chitosan/Clay Composites

Figure 2showed a pseudo-second order kinetic profile for the adsorption of $CO_2$from flue gas where, the experimental data obtained was able to fit in the pseudo-second order model developed in chapter 3 of equation (3.80). The data fitted well with coefficient of determination value of 0.922, resulting in the second order kinetic constant,k_2,of 0.0009g/mg.hr and the equilibrium concentration value of 25mg/g. It is a first order process and this means that the data obtained from the adsorption process is good and acceptable for the process. This compared to the first order kinetic adsorption process, the pseudo-second order kinetic model provided better fit than the pseudo-first order kinetic and could be adopted for the adsorption process. However, though all the kinetic models provided reliable study of the experimental data as the data gave coefficient of determination values of 0.9 and above.

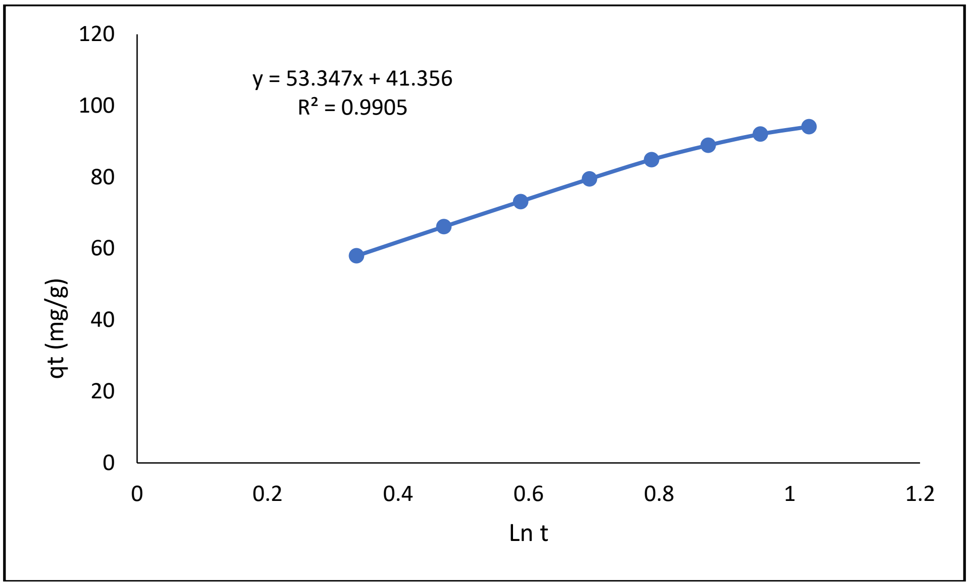

Figure 3: Elovich Kinetics of the Adsorption CO_2 from Flue Gas using Chitosan/Clay Composites

The experimental data fitted well to the Elovich kinetics model for the adsorption of CO_2 from the flue gas using the nanoparticle composite. This fitted well with the Elovich kinetic constant or initial rate of adsorption value of 0.0187mg/g.hr, with adsorption constant of also given as 0.0187g/mg and the determinant energy given as $4.873*10^{19}$J. The coefficient of determination which signifies the degree of fitness of the experimental data of the adsorption of CO_2 into the surface of the nanoparticle composites prepared from chitosan/clay materials is 0.991, an indication that the Elovich kinetic fitted the adsorption process very well compared to pseudo-second order and pseudo-first order kinetics respectively. Therefore, the Elovich kinetic constants are best for this adsorption process and the order of kinetic which the adsorption of CO_2 onto the surface of the nanocomposite is Elovich > pseudo-second order > pseudo-first order. The R^2 value obtained from the Elovich kinetics showed that the adsorption process is a first order one, data acceptable and reliable.

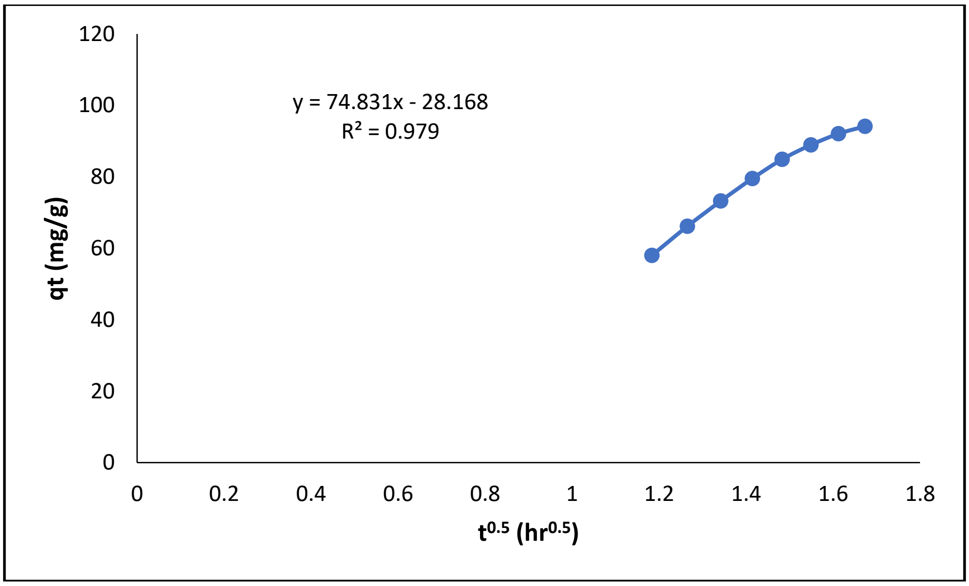

Figure 4: Intra-Particle Diffusion Kinetics of the Adsorption CO_2 from Flue Gas using Chitosan/Clay Composites

Intra-particle diffusion kinetic model was used to study the kinetics of the adsorption of CO_2 onto the surface of the chitosan/clay nanoparticles composite as shown in Figure 4. This kinetic gave the kinetic adsorption constant of k_d as 74.831mg/g.hr$^{0.5}$ and coefficient of determination value as 0.979. The kinetics is a good match for the study of the adsorption process and gave a reliable, acceptable and of first order process, though when compared to Elovich, it is not better. However, the order of acceptability of the type of kinetics needed to study the adsorption process is Elovich> intra-particle diffusion >> pseudo-second order > pseudo-first order. From the various kinetics studied for the adsorption of CO_2 from flue gas onto the surface of chitosan/clay nanoparticles, it can be agreed and recommended that Elovich kinetics is the best to study the adsorption process since it is highly reliable, fitted well and acceptable due to R^2 value is 0.991 > 0.979 > 0.922 > 0.871 for intra-particle diffusion, pseudo-second order and pseudo-first order.

3.2 Isotherms Parameters Determination

The isotherms considered in this study are the Langmuir, Freundlich and Temkin. One of the isotherms which best fitted well to the experimental data is recommended based on coefficient of determination. Table 2 display isotherms parameters and coefficient of determination to ascertain which of the isotherm

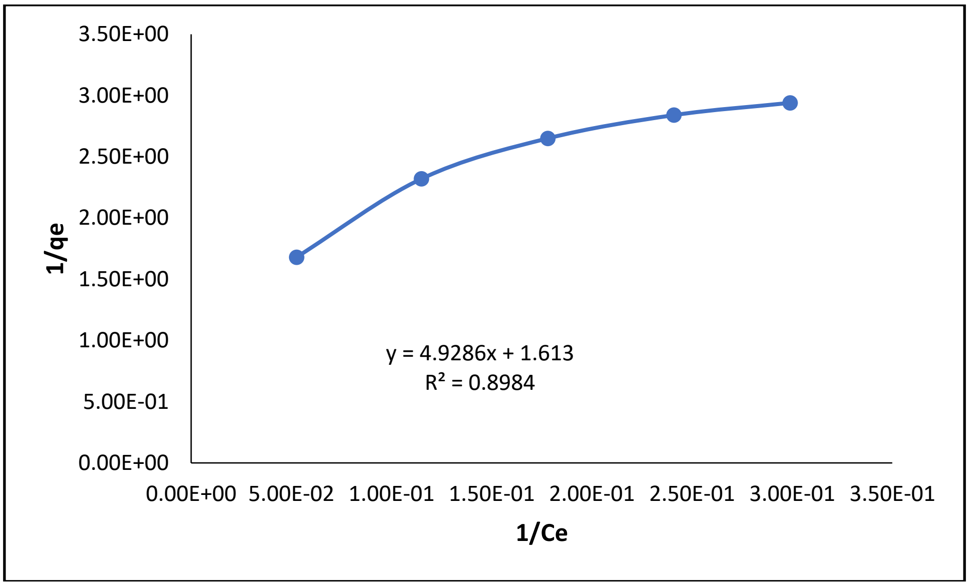

Figure 5: Langmuir Plot to Study the Fitness of the Isotherm into the Adsorption Process.

The Langmuir isotherm study for the adsorption of CO_2 from flue gas using chitosan/clay adsorbent is studied and fitted in a straight line with equation for the line shown in Figure 5, which showed that this isotherm can be used to study the adsorption process. The coefficient of determination value for this adsorption isotherm is 0.898, which implies that the Langmuir isotherm is fitted well with maximum monolayer coverage capacity value of 0.620mg/g and Langmuir isotherm constant of 0.126l/mg. The adsorption process is a first order one, reliable and acceptable data, thus the experiment conducted fitted well onto the Langmuir isotherm.

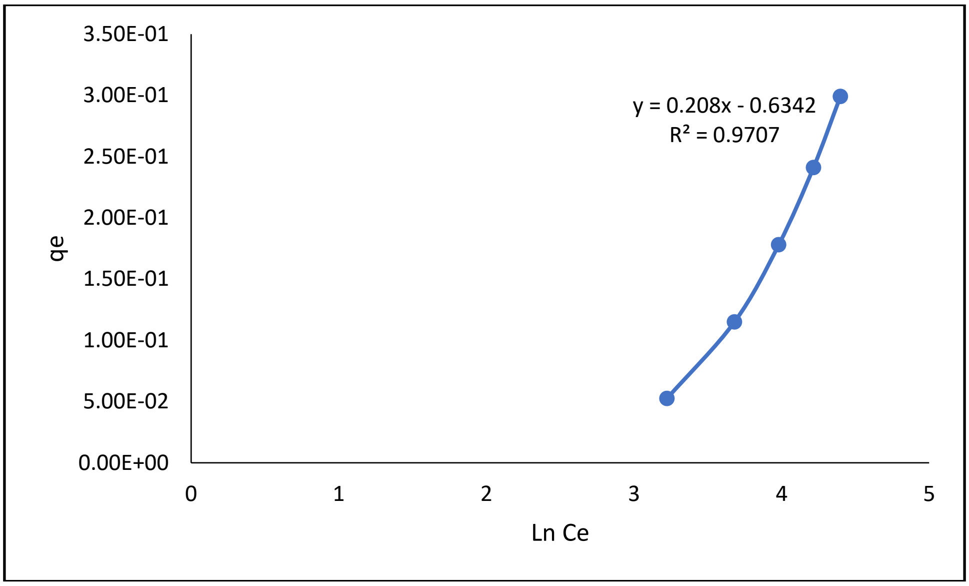

Figure 6: Tempkin Isotherm for the Adsorption of CO_2 and determination of Constants

The Tempkin isotherm is used to fit the experimental data of the adsorption of CO_2 from flue gas using chitosan/clay nanoparticle (adsorbent) as shown in Figure 6. the R^2 value for the adsorption on the isotherm is 0.971, better compared to that of Langmuir isotherm. This means that the adsorption data fitter better into the Tempkin isotherm than the Langmuir isotherm with constant related to heat of adsorption value of 0.208J/mol, Tempkin isotherm constant value of 11911.4, and Tempkin isotherm constant of 0.047l/g. This means that the heat of absorption of all molecules in the layer will decrease linearly rather than logarithmically with coverage at average concentrations (Data *et al*., 2012; Foo and Hameed, 2010; Tempkin & Pyzher, 1940) since the heat of adsorption is characterized by a uniform distribution of binding Energies upto some maximum binding energy as agreed with literatures and the theory behind the adsorption of pollutants into Tempkin isotherm.

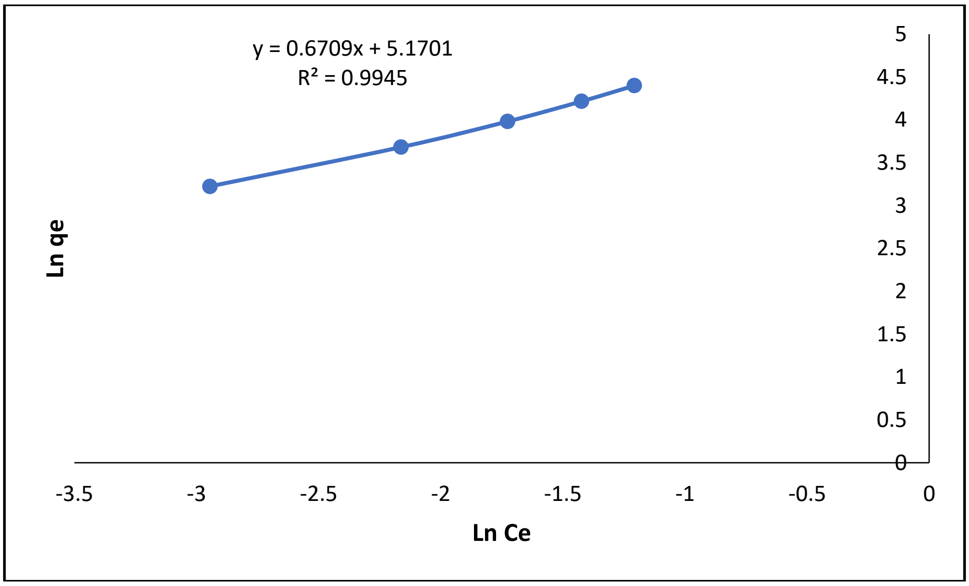

Figure 7: Freundlich Isotherm for Adsorption of CO_2 from Flue Gas

The Freundlich isotherm model shown in model (3.90) was used to fit the experimental data of the adsorption of CO_2 from flue gas using chitosan/clay nanoparticle (adsorbent) as shown in Figure 7. The R^2 value for the adsorption on the Freundlich isotherm is 0.995, better compared to that of Langmuir isotherm. This means that the adsorption data fitter better into the Freundlich isotherm than the Langmuir isotherm with adsorption intensity value of 1.5 and Freundlich constant value of 175.932. This means that the adsorption of CO_2 on to the heterogeneous surfaces of the chitosan/clay nanoparticles and active sites with different energies based on multilayer adsorption and equilibrium (Balouch *et al*., 2003; Boujelbem *et al*., 2013), since the adsorption intensity $n = 1.5 > 1$ as agreed with literatures and the theory behind the adsorption of pollutants onto the multilayer surfaces of the adsorbent and fitted very well with Freundlich isotherm. Therefore, based on this type of adsorption of CO_2 from flue gas on to the multilayer of the chitosan/clay nanoparticles at elevated temperature, the process followed Freundlich isotherm best compared to Tempkin and Langmuir isotherms respectively as the experimental data fitted best using the Freundlich isotherm model compared to the Tempkin and Langmuir isotherm models. Hence, the degree of sorption of the pollutant on to the surface of the nanoparticle is in the order of Freundlich multilayer adsorption >Tempkin interaction of adsorbates> Langmuir monolayer adsorption since the R^2 value are 0.995 > 0.971 > 0.898 respectively.

IV. Conclusion

The research considered the statistical design and kinetic studies of CO_2 capture process from stream of gas mixtures at elevated temperature using composite of nanoparticles and the results obtained showed that;

i. The adsorption process followed Elovich kinetic with 0.991 goodness fit as against intra-particle diffusion, pseudo-second order and pseudo-first order kinetics with goodness fit values as 0.979, 0.922, and 0.871 respectively. Freundlich multilayer adsorption fitted well to the adsorption process with coefficient of determination value of 0.995 compared to Tempkin interaction off adsorbate 0.971, and Langmuir monolayer value of 0.898.

ii. It was noticed that time and temperature play a major role in high adsorption of CO_2 at elevated temperature of 85% and 80% absorbed compared to time and bed height factors of 85% and 79%, more reliable when compared to temperature and bed height of 79% and 79% CO_2 adsorbed respectively. At minimum adsorption process, time and temperature factors are still considers the least when compared to temperature and bed height, followed by time and bed height factors. Hence, temperature is the most dependent and highly needed factor for the adsorption efficiency.

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