SCIENTIFIC PAPER

CO_2 ABSORPTION IN NANOFLUID WITH MAGNETIC FIELD

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ABSTRACT

Acidic gases like CO_2, SO_2, NO_2, H_2S etc., are to be removed as these are polluting the atmosphere in one way or another by inducing temperature rise which further results in undesirable climatic change. Among all these gases CO_2 is the most responsible for the environmental issues and its capture becomes prime importance. The objective of this work is the enhancement of the CO_2 absorption by employing nanofluids in the presence of magnetic field. The nanofluid used in this work is Al_2O_3/water in the concentration of 0.0015 %. The maximum flux obtained is 0.014 mol/m^2s(without magnetic field) and 0.015 mol/m^2s(with magnetic field) for lower CO_2 flow rate of 30 LPH. Hence the nanofluids along with magnetic field shows the positive performance towards the absorption of CO_2.

KEYWORDS: CO_2, Flux, Flow rate, Global warming

HIGHLIGHTS

- Experiments carried with/without the magnetic field of the structured packed absorption column
- Nanofluids were prepared by dispersing Al_2O_3 nanofluidsin water with/without magnetic field
- The mass transfer performance of the structured packed absorption column have been investigated

INTRODUCTION

Due to human activities and industrial revolution from the 17th century 289 ppm of CO_2 has been increased to 406 ppm in 2017 which results in 2°C raise in atmospheric temperature in the current status. To avoid dangerous ecological and environmental issues, CO_2 sequestration has to be carried out **[1,2]**. The major sources of green house gases **[3]** are from burning of fossil fuels, fermentation, deforestation, chlorofluorocarbons, agricultural activities **[4,5]** etc. CO_2 is the major contributor for global warming which results loss in biodiversity and extreme changes in agricultural activities **[6,7]**. CO_2 has been captured by various technologies **[8]** like adsorption, membrane separation, cryogenic method, biological fixation and wet scrubbing or absorption. This wet scrubbing is the cost effective and widely accepted method to capture the acid gas. Among all the chemical solvents **[9]**, amine based solvents are used to capture CO_2. But this method has several advantages and certain disadvantages like high power consumption, low gas loading, high equipment corrosion, amine degradation etc.,. Hence a novel solvent with low cost, higher capacity and efficiency with lesser decomposition and corrosion rate is the aqueous ammonia **[10,11]**. After the solvent selection; next absorption is to be carried out in the wetted wall column or packed column. Among these two, wetted wall column is most preferable with film promoter. The film promoter improves the formation of a stable liquid film and liquid distribution inside the column. Apart from the conventional solvents nanofluids **[12,13,14]** are used to enrich the absorption rate. With Al_2O_3, nanofluids **[15,16,17]**, the Brownian movement and grazing effect has been enhanced by the application of magnetic field.

CO_2 SEQUENSTRATION METHODS

There are various technologies available for acid gas removal. They are adsorption, membrane separation, cryogenic method and absorption process.

Absorption

Absorption or Wet scrubbing is the transfer of one or more species from the gas phase to a liquid phase. The species transferred to the liquid phase are known as solutes or absorbate. Absorption occurs by physical and chemical means. Physical absorption occurs when there is no chemical reaction between the solute and the solvent. Absorption with Chemical absorption involves the removal of impurities from gas phase and dissolving them into liquid.

Membrane Separation

The membrane permeates the chemical species more easily than the other conventional methods. The porous or semi- porous structured membranes like polymers, zeolite or inorganic membranes like palladium are used. These gas separation membranes will permeate one of the component more quickly than the other. The separation capacity is increased by performing multiple stage operation but it leads to high energy consumption, operational cost and difficulty of operation.

Cryogenic Method

Cryogenic method involves the gas compression and cooling it to the minimum temperature thereby the separation takes place by distillation up to 90 %. This method is used for high CO_2 concentrations, high pressure gases and high energy is needed for refrigeration. The unwanted component like water is to be removed before cooling the gas stream to avoid blockages. The light olefins and paraffins are studied by cryogenic distillation. Alternate to distillation, absorption was carried out with mass transport but distillation was carried with energy transport. The absorption reduces the capital cost and energy requirements of aromatic separations. Copper and Silver based solutions were used to separate olefins and paraffins.

Adsorption

The flue gas stream is passed through the solid adsorbent such as Zeolite, activated carbon and CO_2 is held on the surface particles of the adsorbent. These adsorbents are heated to desorb the CO_2. Desorption is carried out either reducing the pressure or by increasing the temperature . This method is not carried out for large scale separation of CO_2 from the flue gas and hence this method is used along with other methods like pressure or electric swing adsorption.

Other Methods

The other methods used for CO_2 capture are carbonation, calcination and biosorption etc., Carbonation means adding limestone or carbon compounds to separate CO_2 from the gas stream. Calcination means heating to very high temperatures in the presence of oxygen. Biosorption is the process of removing CO_2 from the gas stream by the usage of microorganisms. This process has been carried out by employing specific microorganisms suitable for CO_2 absorption. So, a thorough knowledge on the application and availability of microorganisms should be identified.

Reaction Mechanism

The reaction between aqueous ammonia and carbon dioxide mainly occurs in the liquid phase of the gas-liquid interface, the reactions in the liquid phase of the CO_2 - ammonia system are given as follows:

The total reaction involved in the process is described as follows

$$CO_2\,(g) + NH_3\,(aq) + H_2O \longleftrightarrow NH_4HCO_3\,(aq) \qquad (1)$$

The actual process is more complicated and can be described in the step by step process.

CO_2 (g) + $2NH_3$ (aq) ←→ NH_2COO- (aq) + NH_4^+ (aq) (2)

At the same time the complex balance of solute forms ionic reactions in the solution and the equation are as follows

NH_3(aq) + H_2O(l) ←→NH_4^+ (aq) + OH^- (aq) (3)

NH_4HCO_3(aq) ←→ NH_4^+ (aq) + HCO_3^-(aq) (4)

$(NH_4)_2CO_3$(aq) ←→ $2NH_4^+$ (aq) + CO_3^{2-}(aq) (5)

OH- (aq) + HCO_3^-(aq) ←→ CO_3^{2-}(aq) + H_2O (6)

CO_3^{2-}(aq) + CO_2(g) + H_2O(l) ←→ $2HCO_3^-$(aq) (7)

The reaction (2) is very fast and irreversible and instantaneous. The reaction (4) is too slow to influence the rate of the absorption directly. Therefore, the reaction between aqueous ammonia and carbon dioxide is mainly controlled by reaction (2). It is a second-order reaction with first- order for CO_2 and NH_3 respectively. On the other hand, reactions in (1) and (2) are reversible, with ammonium carbonate ($(NH_4)_2CO_3$) or bicarbonate (NH_4HCO_3) as the products. The forward reactions are dominant at room temperature. The backward reactions occur at temperatures of around 38-600 °C.

ENHANCEMENT OF CO_2 ABSORPTION

Enhancement Using Nano Fluids

Nano particles are of very small in size and dissolved in the solvent. These particles have very large surface area compared to that of the normal particles. Hence the increased surface area increases the more number of gas and liquid molecules to react simultaneously increasing the mass transfer rate. These fluids have novel properties useful in many applications like refrigerator, chiller, heat exchanger, pharma industries, fuel cells, microelectronics etc., Nano fluid synthesis include metallic particles, oxide particles, carbon nano tubes, graphene and ceramic particles. The thermo physical properties of nano fluids include viscosity and thermal conductivities(Farghali *et al.* 2013).

Also the brownian movement of the nano particles in the solvent increases the mass transfer rate. The effect known as grazing effect plays a major role in explaining the role of nano particles in the solvent. Here the nano particles adsorb the gas molecules from the bulk gas and after few seconds it releases the gas molecules into the bulk liquid. Here the nano particles are assumed that they only act as catalyst and does not take part in the chemical reaction of the process (Park *et al.* 2006, 2007, 2008). The major disadvantage in this case is that the nano particles have to be in suspension medium in the solvent. If the nano particle concentration increases beyond the optimized level agglomeration of the particles and hindrance to the absorption of gas molecules into the bulk liquid takes place. The nano fluids are employed in gas absorption studies and furthermore, external magnetic field is induced in the absorption column for the enhancement of gas absorption.

Enhancement Using Magnetic Field

CO_2 absorption was enhanced with the introduction of smaller sized nano paricles. The nano particles used in CO_2 absorption studies are Al_2O_3, TiO_2, SiO_2, NiO, Fe_3O_4, Fe_2O_3, graphene oxide etc., The iron oxide magnetic particles when dissolved in the base solvent and exposed to external magnetic field results in the segregation of nano particles in the nano fluid. As a result the interfacial area is increased. In shuttle mechanism, at the gas–liquid interface nano particles adsorb the carbon dioxide in the film layer and after some time desorb them in the solvent and this increases the rate of absorption. In brownian mechanism the nano particle disturbs the flow and enhances the mixing of the liquid leading to increase in residence time and good gas-liquid contact within the absorption column. This accounts to increase in rate of CO_2 absorption by nano fluids. The presence of magnetic field increases the randomness of the nano particles in the fluid accounting to increased mass transfer rate.

Other Methods for Absorption Enhancement

Other methods for CO_2 absorption enhancement includes the usage of solid absorbents like calcium based sorbents, sodium based sorbents, other metal based sorbents, pressure and temperature swing adsorption, mixed technology, acid catalyst, carbon nano tubes, microchannel reactors etc., Solid absorbents like calcium absorbents from limestone are low price, non-toxic in nature, CO_2 capture easily employed in fluidized bed reactor at very high temperature around 500^0C. The sorbent can be reused in cement production and hydrogen production. Carbon nano tubes are employed for low temperature and regeneration of the solvent is easy.

EXPERIMENTAL SETUP

The experimental setup consists of a wetted-wall column, solvent container, liquid distributor, CO_2 vessel, pump, and flow meters. Height and internal diameter of wetted wall column are 0.5 m and 0.012 m respectively. The liquid flows inside the wetted wall column to cover the internal surface of the wetted-wall column as a very thin layer. The liquid flow rate varies in the range of 0.7–1.1 LPH and the gas flow rate is adjusted at 30 and 90 litre per hour.

Fig. 1

To generate a homogeneous magnetic field, a coil of 1.1 mm-thick copper wire with 10000 windings is wrapped around the external surface of the absorption column. The coil is winded throughout the entire length of the inner tube of the column(0.5 m) and is placed in the middle of the column. A maximum electrical current of 2 A is applied to the coil with the aid of transformer. The nanofluid consist of mixture of Al_2O_3 and water. Average particle size of Al_2O_3 used is 53 nm. The CO_2(gas) and nanofluid(liquid) is passed into a wetted wall column in a counter current manner. The CO_2 is passed at the bottom of wetted wall column while nanofluid liquid is allowed to enter at the top of the column. The nanofluid starts to absorb CO_2 gas. A copper coil is wrapped around the wetted wall column to generate magnetic field. The heat generated during the process is cooled by circulating cold water through a coil which is placed between magnetic field coil and the outer column. The magnetic field is applied to enhance higher mass flux, residence time and mass transfer coefficient. The absorbed CO_2 is collected at the bottom of the wetted wall column. The absorbed CO_2 is titrated against sodium hydroxide by adding phenolphthalein as an indicator. By varying the flow rates, voltage the readings would be taken with and without magnetic field. Care is taken to ensure a uniform thin layer of nanofluid is established in the inner surface of the column.

Nano Fluid Preparation (λ-Al_2O_3):

Aluminum sulfate pentahydrate, $Al_2(SO4)_3 \cdot 18H_2O$, (4gram) and polyethylene glycol (5 gram) (abbreviated as PEG2000) are dissolved in 100 mL distilled water. Aqueous ammonia is added to the above solution, giving rise to milky precipitates at pH= 9. The mixture is then stirred for 1 hour at room temperature and subsequently placed in a Parr Teflon lined stainless steel vessel**[18].** The vessel is completely sealed and heated at 130°C for 48 hours. The mixture is gradually cooled until room temperature is reached. The resulting precipitate is filtered and washed three to four times using double distilled water and ethanol to remove polyethylene glycol and other impurities. The synthesized Al_2O_3 powder is dried at 100°C for 1 hour and is calcined at 400°C for 2 hours**[19,20].**

RESULTS AND DISCUSSION

From the experiments conducted in wetted wall column for the absorption of CO_2 using Al_2O_3 nanofluid, it was observed that the change in molar flux with respect to solvent flow rates follows a polynomial trend. The relationship between molar flow rate and solvent flow rate can be better described by the equation (1),

$$F = a \cdot S^2 \tag{8}$$

Where F is molar flux and S is the solvent flow rate and by neglecting the higher order terms.

1.EFFECT OF SOLVENT FLOW RATE ON MOLAR FLUX WITHOUT MAGNETIC FIELD AND NANOFLUID

The effect of solvent flow rate on molar flux are carried out for 30, 60 and 90 LPH of CO_2 flow rate. The observations are shown in the fig. 2.

Fig. 2

It is observed that at 60 LPH of CO_2 flow rate the change in molar flux with respect to the change in solvent flow rate follows a polynomial trend. It has been shown in the fig.3, the maximum flux is obtained at 60 LPH is 0.0058 mol/m^{2}s. At this point the solvent flow rate is 1.1 LPH without the aid of magnetic field.

Fig. 3

It was observed that as the flow rate of the solvent increases the molar flux also increases. This was due to the mechanism that as solvent flow rate increases more amount of solvent molecules to absorb the CO_2. As a result of more solvent molecules leads to viscosity effect shoots up.

2.EFFECT OF SOLVENT FLOW RATE ON MOLAR FLUX WITH MAGNETIC FIELD WITHOUT NANOFLUID

The following fig.3 illustrates the influence of solvent flow rate on molar flux in presence of magnetic field, for different CO_2 flow rates.

Fig. 4

From fig. 4 it can be interpreted that when magnetic field is used, greater molar flux can be obtained for the same molar flow rates. Molar flux of >0.002 mol/m^{2}s can be achieved at lower flow rate of solvent (1.1 LPH) and CO_2 (30 LPH) when magnetic field is used. At higher CO_2 flow rate of 90 LPH, high molar flux was observed even at lower solvent flow rate. fig.5 shows greater change in molar flux for increasing solvent flow rates and as high as flux of >0.009 mol/m^2 s was observed at 1.1 LPH of Solvent flow rate.

Fig.5

With the presence of magnetic field the flux increases even in the higher solvent flow rates. The movement of solvent molecules would be promoted by the magnetic field produced in the wetted wall column.

3.INFLUENCE OF NANOFLUID (0.0015) ON CHANGE OF FLUX WITH RESPECT TO SOLVENT FLOW RATE (WITHOUT MAGNETIC FIELD).

Fig. 6

The fig.6 shows that there is a increase in the rate of change of molar flux (positive slope). The higher values of molar flux at a low CO_2 flow rate of 30 LPH. The change in flux at this flow rate is better described in fig. 7.

Fig. 7

A maximum molar flux of 0.0014 mol/m^2 s was observed for 30 LPH of CO_2 flow rate. The presence of nano particle increases the mixing effect which was due to the Brownian motion. The increase in the Brownian motion was due to the fact that it increases upto a critical value and beyond this value the interparticle interaction hinders its motion. This mechanism is known as shuttle or grazing effect. These nano particles may increases the interfacial area by covering the gas bubble surface and preventing the coalescence of these bubbles[21,22].

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4.INFLUENCE OF NANOFLUID (0.0015 %) ON CHANGE OF FLUX WITH RESPECT TO SOLVENT FLOW RATE (WITH MAGNETIC FIELD).

The Brownian movement and micro convection of nanoparticle is responsible for the achievement of high mass transfer. Brownian movement of nanoparticle is the random movement of smaller sized particles in the fluid. The mass diffusion occurs by the diffusive transport of species due to concentration difference. When magnetic field is applied the absorption increases than the without magnetic field by 66.10%. Nano particles with magnetic field promotes the Brownian movement and the grazing effect, thereby the removal efficiency increases with high mass transfer rate**[23]**. The fig.8 depicts the change of molar flux with solvent flow rates for different CO_2 flow rates. The graph reveals that higher flux can be best achieved at a lower CO_2 flow rate of 30 LPH. This higher molar flux for different solvent flow rates even at low CO_2 flow rate was achieved because of the use of nanofluids (0.015 mol/m^{2}s). It was also observed that when nanofluids were used in presence of magnetic field, higher CO_2 flow rate resulted in lower molar fluxes for different solvent flow rates.

Fig. 8

Hence lower CO_2 flow rate was considered sufficient for obtaining better molar fluxes when nanofluids are used in presence of magnetic field.

CONCLUSION

The CO_2 gas absorption studies have been carried out in wetted wall column. Selection of suitable solvent, comparing the performance of random and structured packing material, enhancement of absorption by adding nano materials in solvent and induction of magnetic field were detail studied. An effort has been made to enhance CO_2 sequestration with the nanofluid addition and in the presence of magnetic field. The mass transfer flux increases with the magnetic field than without the magnetic field. When nanofluid (0.0015 %) is used along with magnetic field, the flux increases. Further study can be carried out for higher ratio of nanofluids (>0.0015 %). For the nanofluids, as the volumetric ratio increases the absorption also increases. Hence the acid gas absorption has been carried out in a wetted wall column with higher mass transfer rate with lower solvent flow rate and CO_2 gas flow rate. The following conclusions were drawn out of the study.

1.Among different solvents considered in this study NH_3 found to be better for CO_2 gas absorption. It was observed that the structured packing resulted in the higher efficiency than random packing.

2.It was also observed that the % CO_2 removal was found to be increased with increase in the NH_3 concentration.

3.With the introduction of nano fluids the % CO_2 removal increased. Increase of ammonia concentration and decrease of temperature have a positive effect on the density and viscosity. Aqueous ammonia also have a non-volatile state and a high-surface tension, which could be useful for providing a better CO_2 absorption performance.

4.Pka is a major physio-chemical parameter which is used to analyse the reaction mechanism and its molecular structure. Since ammonia was having higher reactivity than amine solutions.

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FIGURE CAPTIONS

Fig. 1 Schematic diagram of wetted wall column

Fig. 2 SEM image of Al_2O_3

Fig.3 Molar flux vs. Solvent Flow rate for Different CO_2 Flow Rates: 30, 60, and 90 LPH(without magnetic field)

Fig. 4 Molar flux vs. Solvent Flow rate for CO_2 Flow Rates of 60 LPH (without magnetic field)

Fig. 5 Molar flux vs. Solvent Flow rate for Different CO_2 Flow Rates: 30, 60, and 90 LPH (with magnetic field)

Fig. 6 Molar flux vs. Solvent Flow rate for CO_2 Flow Rate of 90 LPH (with magnetic field)

Fig. 7 Molar flux vs. Solvent Flow rate for Different CO_2 Flow Rates: 30, 60, and 90 LPH, using Nano fluid (0.0015 %) without magnetic field

Fig. 8 Molar flux vs. Solvent Flow rate for CO_2 Flow Rate of 60 LPH, using Nanofluid (0.0015 %) without magnetic field

Fig. 9. a) Molar flux vs. Solvent Flow rate for Different CO_2 Flow Rates: 30, 60, and 90 LPH, using Nanofluid (0.0015 %) with magnetic field; b) Molar flux vs. Solvent Flow rate for CO_2 Flow Rate of 30 LPH, using Nanofluid (0.0015%) with magnetic field

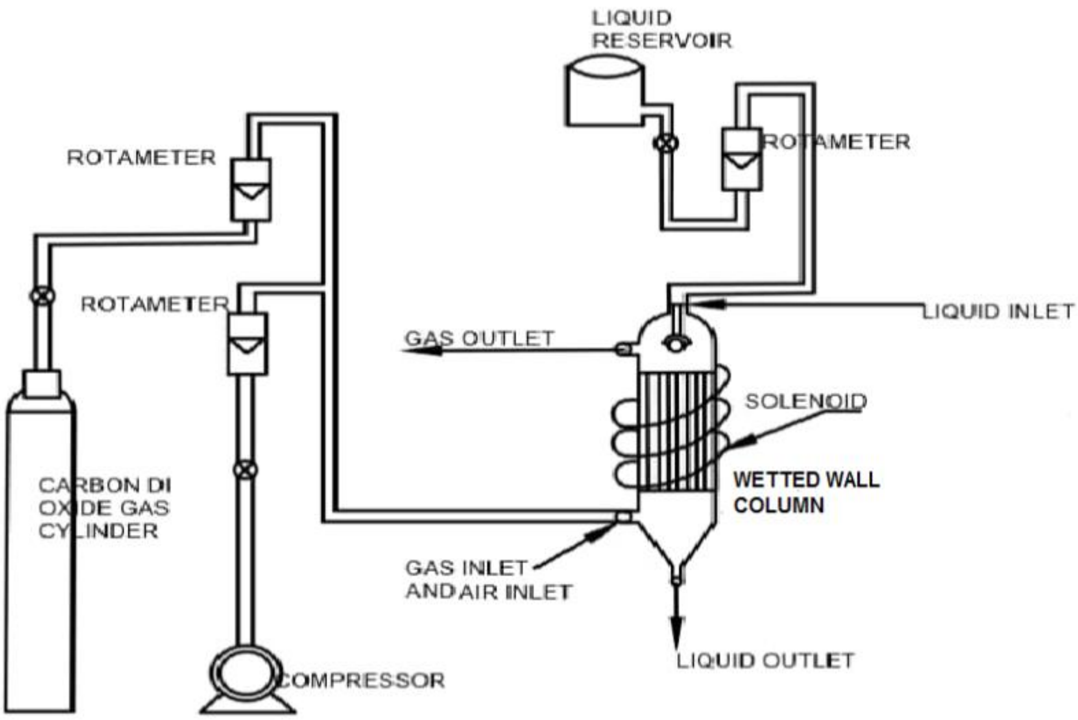

LIQUID
RESERVOIR
ROTAMETER
ROTAMETER
ROTAMETER
LIQUID INLET
GAS OUTLET
SOLENOID
WETTED WALL
COLUMN
CARBON DI
OXIDE GAS
CYLINDER
GAS INLET
AND AIR INLET
LIQUID OUTLET
COMPRESSOR

Fig. 1

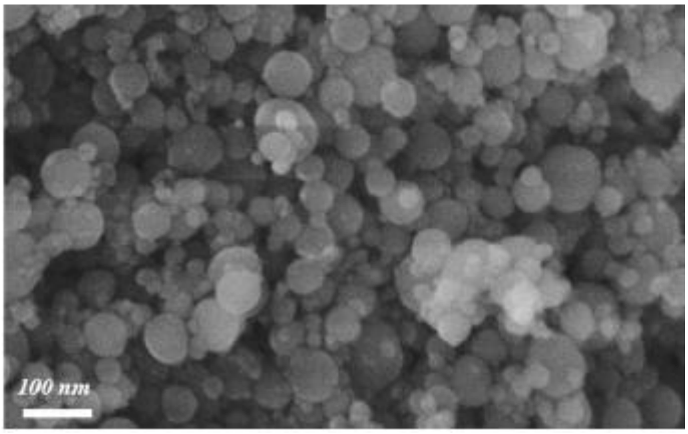


Fig. 2

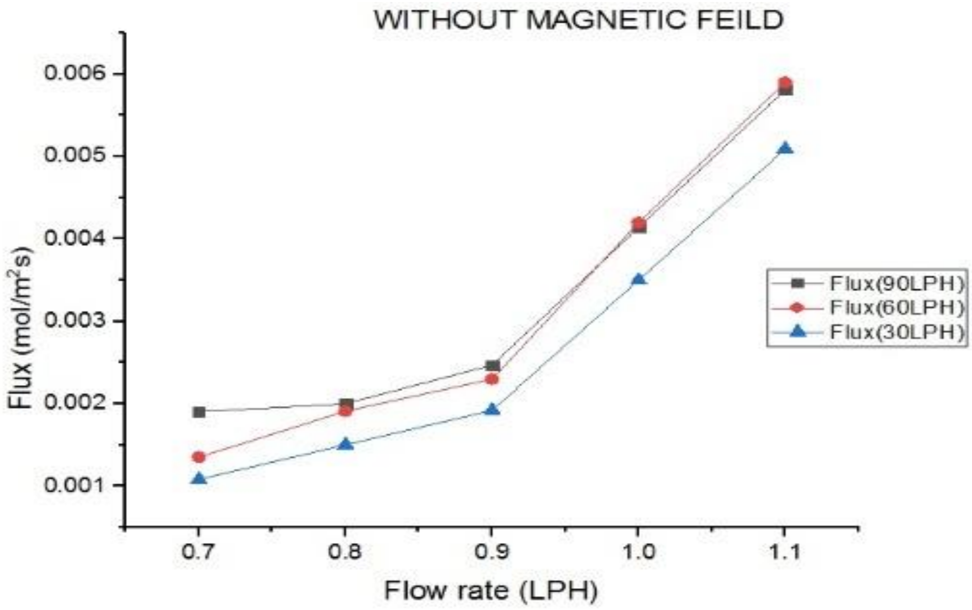

WITHOUT MAGNETIC FEILD
0.006
0.005
0.004
0.003
0.002
0.001
Flux (mol/m^{2}s)
Flux(90LPH)
Flux(60LPH)
Flux(30LPH)
0.7
0.8
0.9
1.0
1.1
Flow rate (LPH)

Fig. 3

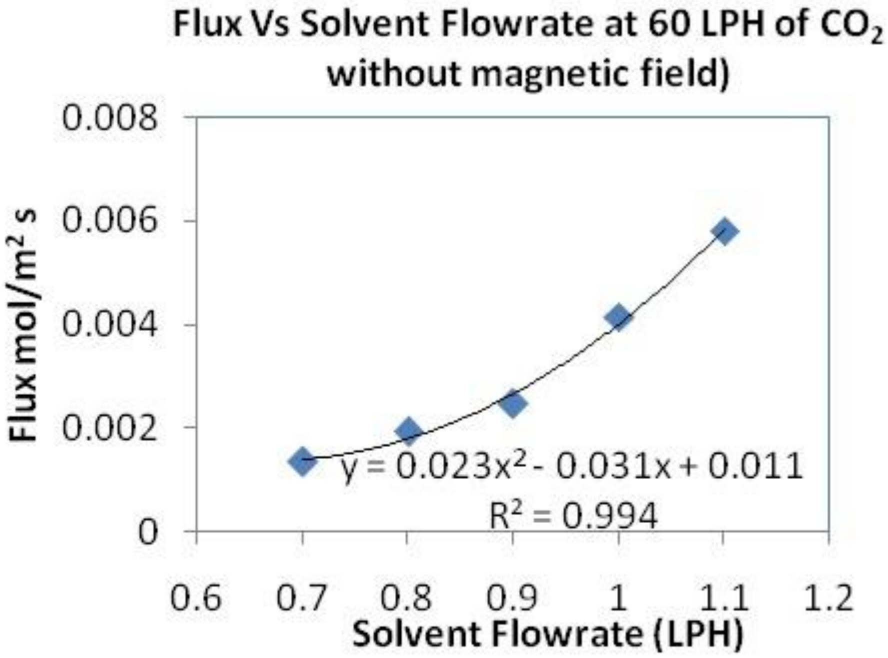

Flux Vs Solvent Flowrate at 60 LPH of CO2
without magnetic field)
0.008
0.006
0.004
0.002
0
Flux mol/m2 s
y = 0.023x2 - 0.031x + 0.011
R2 = 0.994
0.6
0.7
0.8
0.9
1
1.1
1.2
Solvent Flowrate (LPH)

Fig. 4

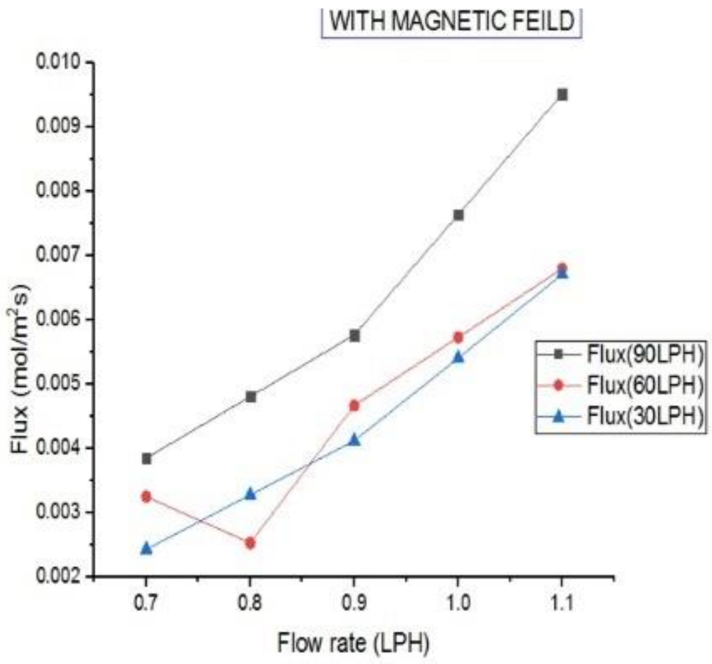

WITH MAGNETIC FEILD
Flux (mol/m²s)
0.010
0.009
0.008
0.007
0.006
0.005
0.004
0.003
0.002
0.7
0.8
0.9
1.0
1.1
Flow rate (LPH)
Flux(90LPH)
Flux(60LPH)
Flux(30LPH)

Fig.5

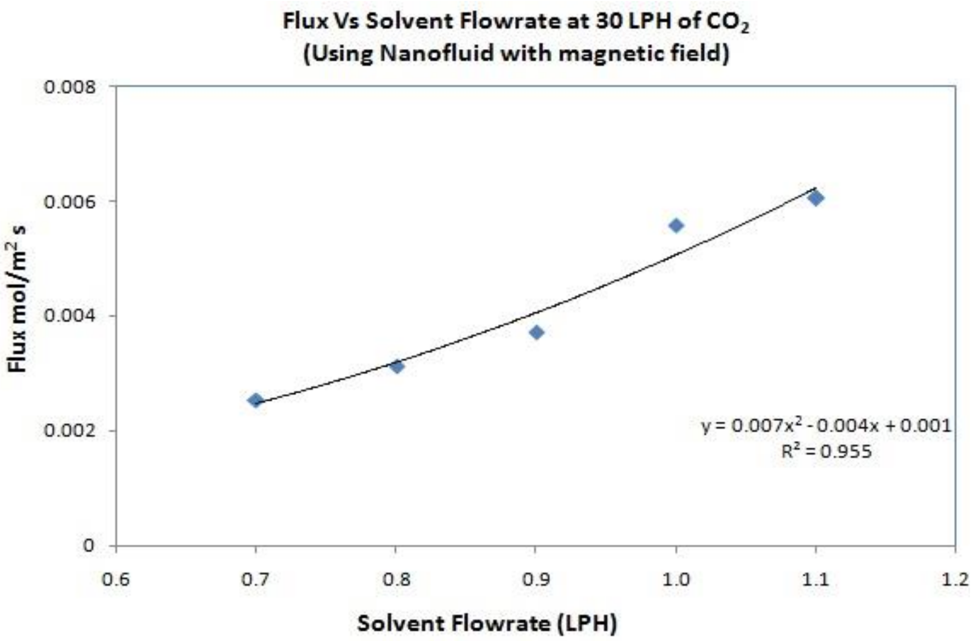

Flux Vs Solvent Flowrate at 30 LPH of CO_2
(Using Nanofluid with magnetic field)
Flux mol/m^2 s
0.008
0.006
0.004
0.002
0
0.6
0.7
0.8
0.9
1.0
1.1
1.2
Solvent Flowrate (LPH)
$y = 0.007x^2 - 0.004x + 0.001$
$R^2 = 0.955$

Fig.6

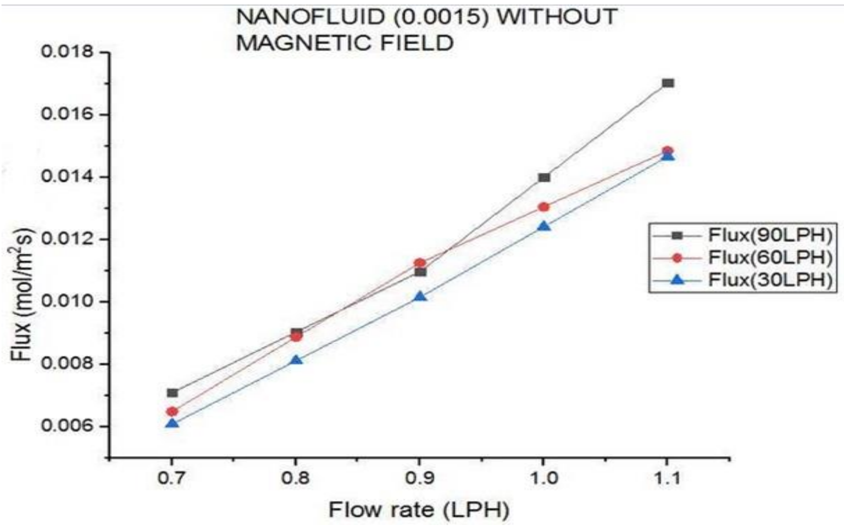
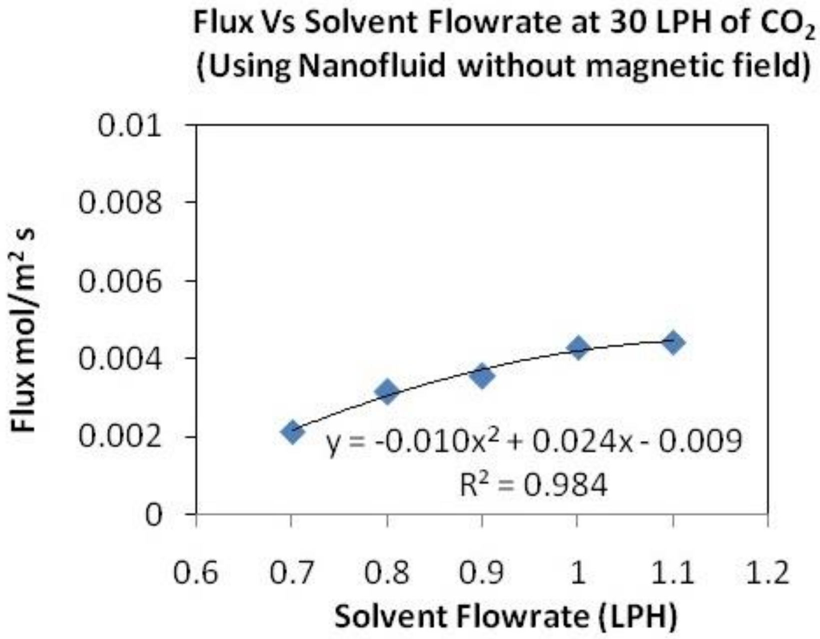
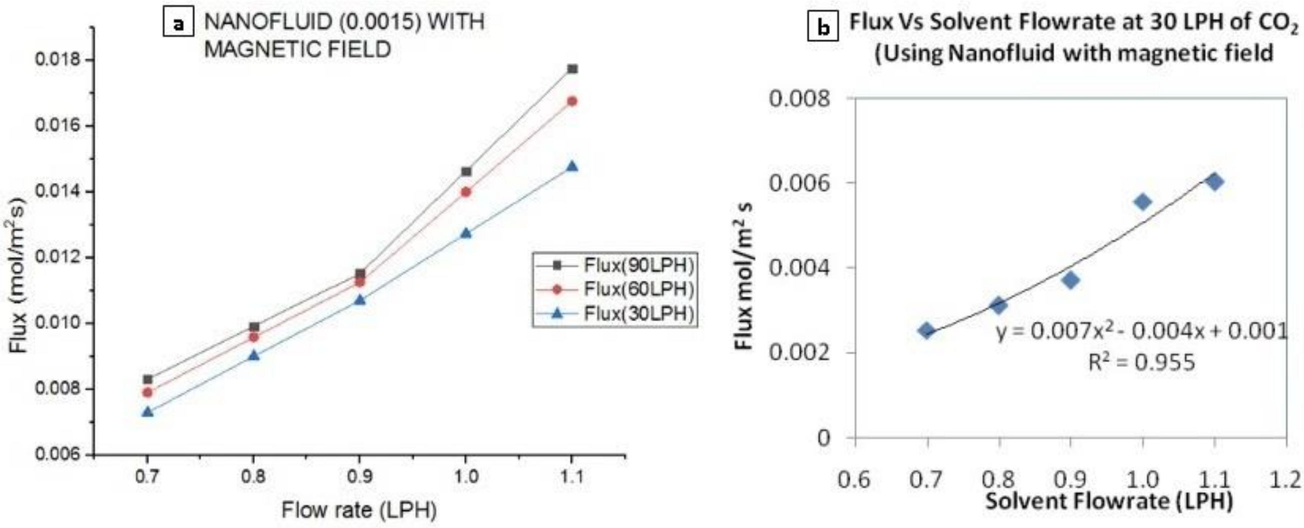

NANOFLUID (0.0015) WITHOUT MAGNETIC FIELD
0.018
0.016
0.014
0.012
0.010
0.008
0.006
Flux (mol/m2s)
0.7
0.8
0.9
1.0
1.1
Flow rate (LPH)
Flux(90LPH)
Flux(60LPH)
Flux(30LPH)

Fig 7



Flux Vs Solvent Flowrate at 30 LPH of CO2 (Using Nanofluid without magnetic field)
0.01
0.008
0.006
0.004
0.002
0
Flux mol/m2 s
y = -0.010x2 + 0.024x - 0.009
R2 = 0.984
0.6
0.7
0.8
0.9
1
1.1
1.2
Solvent Flowrate (LPH)

Fig.8



a
NANOFLUID (0.0015) WITH MAGNETIC FIELD
Flux (mol/m2s)
0.018
0.016
0.014
0.012
0.010
0.008
0.006
0.7
0.8
0.9
1.0
1.1
Flow rate (LPH)
Flux(90LPH)
Flux(60LPH)
Flux(30LPH)
b
Flux Vs Solvent Flowrate at 30 LPH of CO2
(Using Nanofluid with magnetic field
Flux mol/m2 s
0.008
0.006
0.004
0.002
0
y = 0.007x2 - 0.004x + 0.001
R2 = 0.955
0.6
0.7
0.8
0.9
1.0
1.1
1.2
Solvent Flowrate (LPH)

Fig.9