Geo-Sequestration of CO2 in Deep Saline Carbonate Aquifers: The Role of Reservoir Pressure, Salinity and Temperature

Presented by:

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Indian Institutes of Technology

- Autonomous public institutes of higher education
- Governed by the Institutes of Technology Act, 1961 which has declared them as institutions of national importance

History and Journey of IITR: 175+ years

Origin: 1847

Erstwhile Thomason College: The first engineering institution in the British empire

Founded in 1847 to manage the water resources of the Ganges River, the institution carries the mantle of a long and distinguished tradition of teaching and learning.

Strong international character from inception.

E.g., William Willcocks, a civil engineer who graduated from Roorkee in 1872, is a renowned irrigation engineer, having proposed the first Aswan Dam and undertaken major projects of irrigation in South Africa and Turkey.

1949 Erstwhile University of Roorkee

2001 Indian Institute of Technology Roorkee

THE WORLD UNIVERSITY RANKINGS 401-500 **#3 in India**

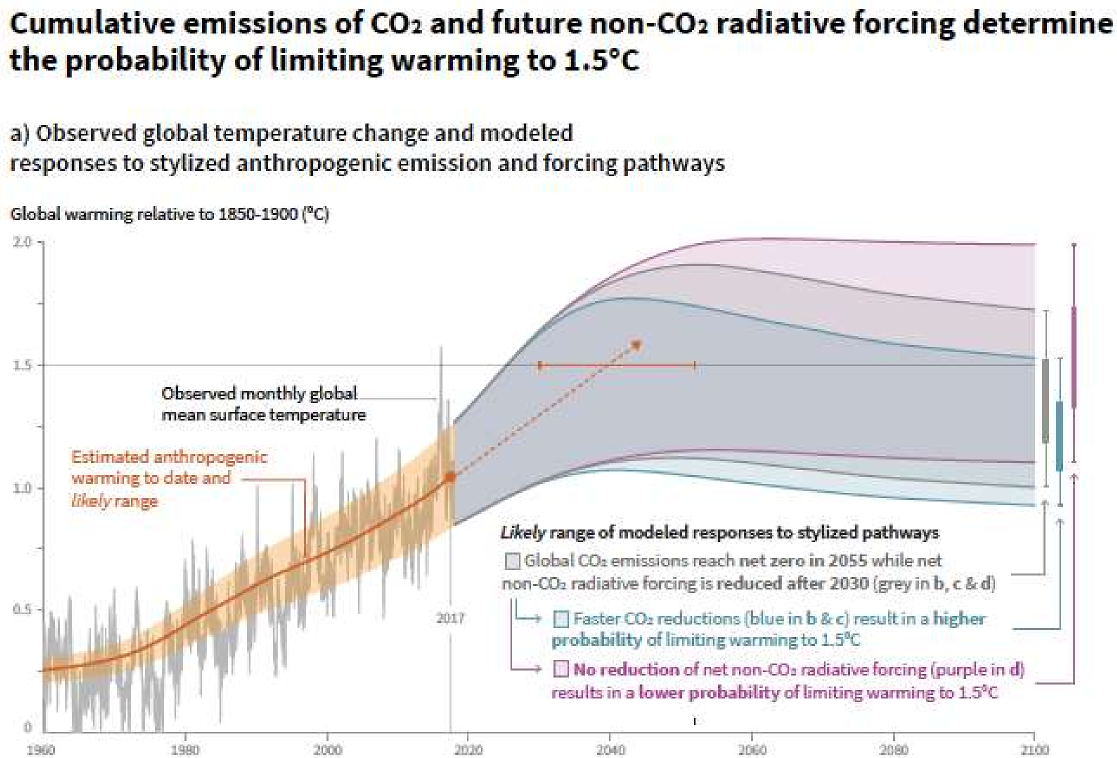
QS WORLD UNIVERSITY RANKINGS 381 **#7 in India**

Introduction

- Since pre-industrial times, increased anthropogenic emissions of greenhouse gases have lead to marked increase in greenhouse gas concentrations.
- Present CO_2 concentration is 413 ppm (NOAA, 2020)

Impacts

- Concern about global climate change
- Potential impact include : sea level rise, floods, erosion, leaching of soil nutrients, change in precipitation pattern, ecology and biodiversity.

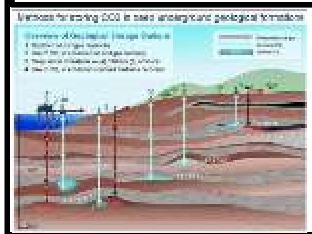


CO_2 Sequestration

CO_2 Sequestration

Bio-sequestration

1. Plant sequestration
2. Plankton sequestration
3. Algae sequestration

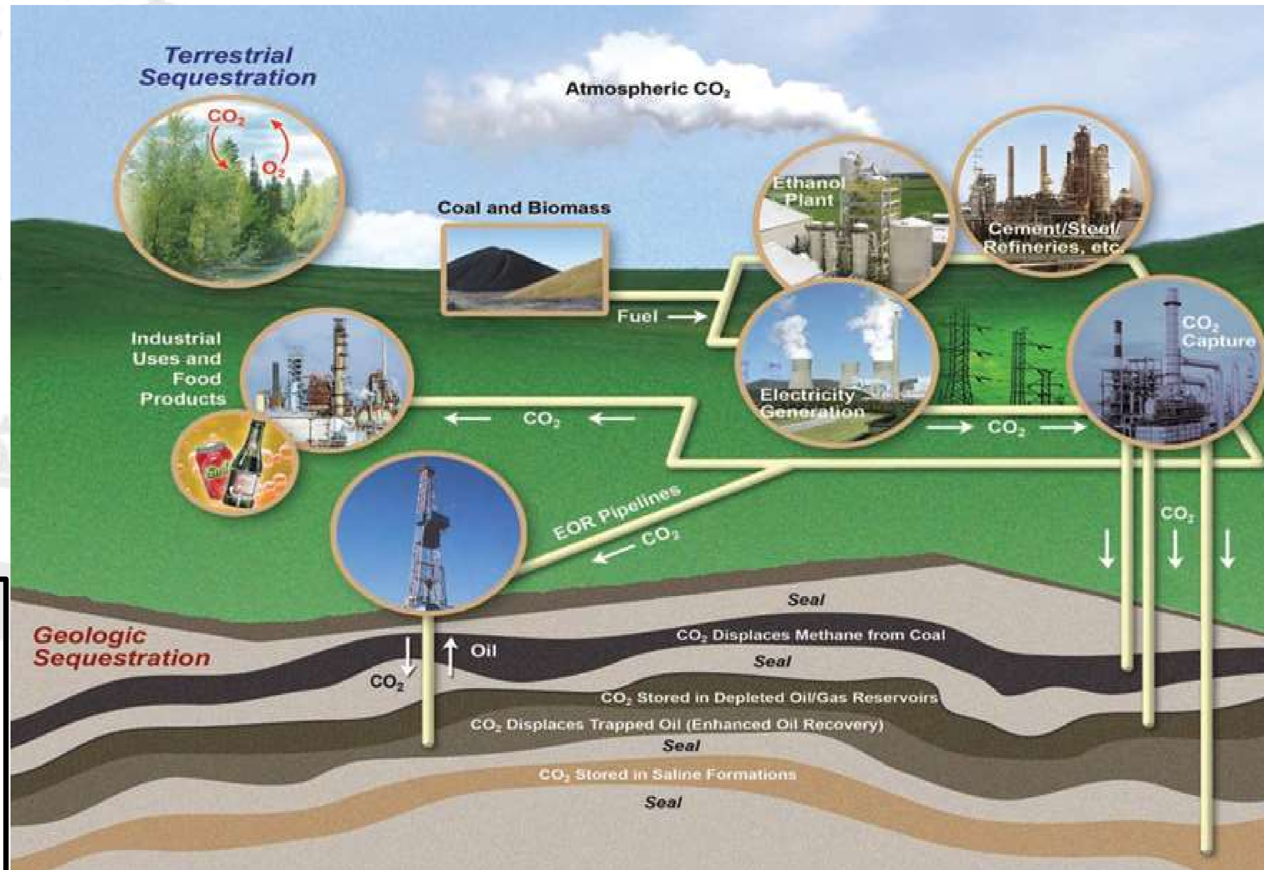


Geo-sequestration

1. Unminable Coal formations
2. Depleted oil and gas reservoirs
3. Deep saline aquifers
4. Ocean sequestration

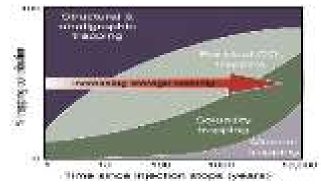
Carbon dioxide Capture Sequestration is a process:

- Capture of carbon-dioxide
- Transport of captured and compressed carbon dioxide
- Underground injection and geologic sequestration

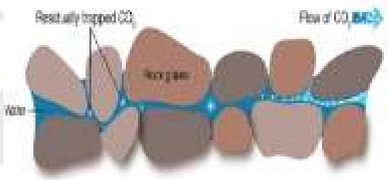


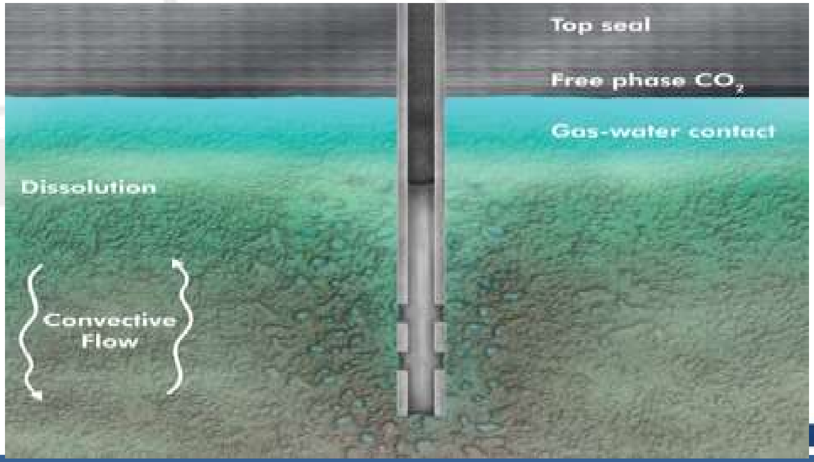
CO_2 trapping mechanisms

CO_2 trapping mechanisms



- Geological trapping
  - Structural trapping
  - Residual trapping
- Geochemical trapping
  - Solubility trapping
  - Mineral trapping





Research gaps

- Majority of earlier studies focused on understanding the different processes related to geological storage of CO_2 in sandstone, shales and coal media.
- Studies related to CO_2 sequestration in carbonate saline formations does not addressed:
  - Impact of salinity level on CO_2 geosequestration
  - Effect of injection pressure and temperature on CO_2 movement
  - Cyclic brine-CO_2 injection
- Major challenges associated with the multiple phases when the supercritical CO_2 is injected along with the reactivity of carbonate minerals at high pressure and salinity conditions.
- Better understanding of how CO_2-brine-rock interaction causes petrophysical changes in porous media is needed.

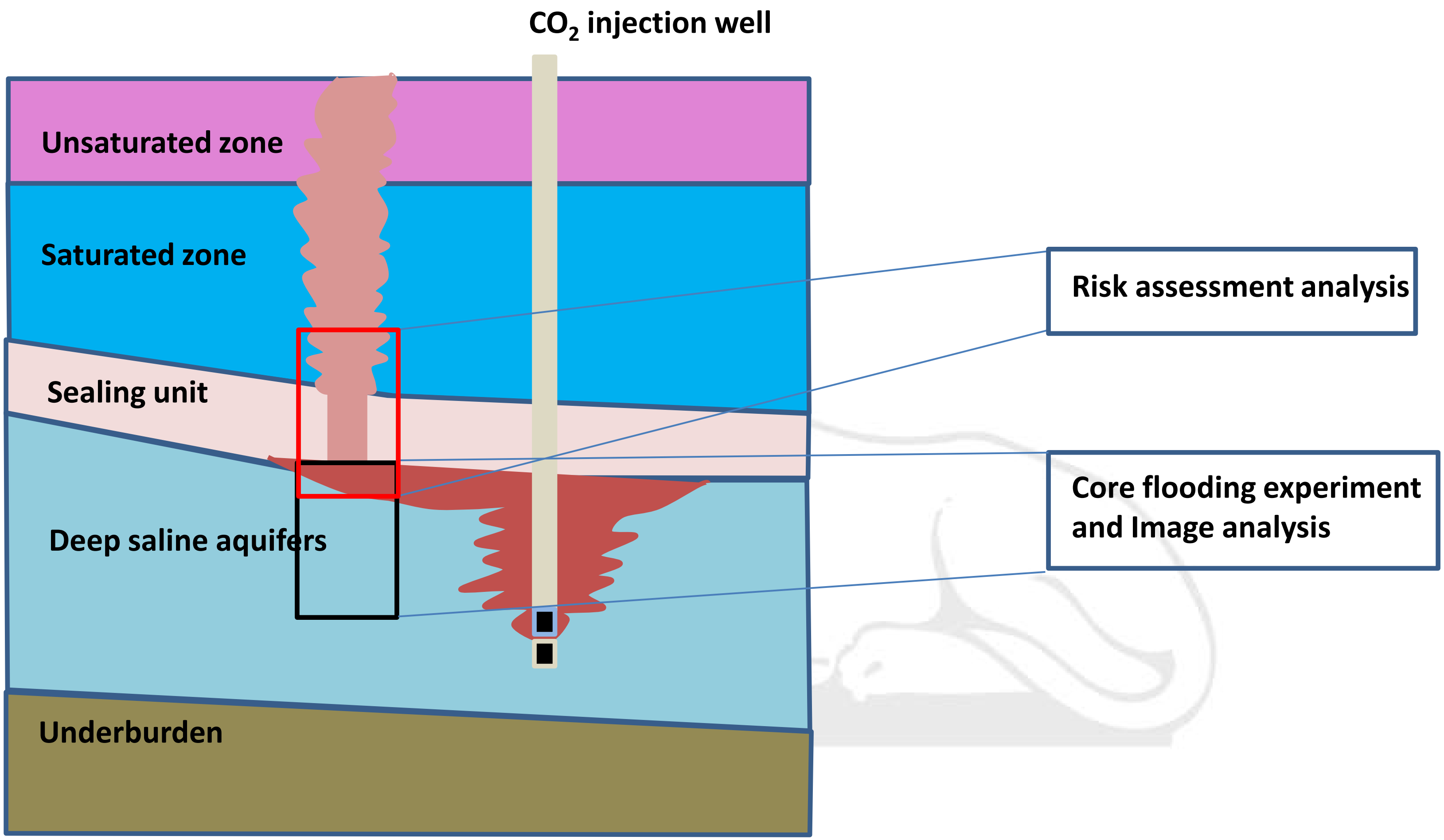
Objectives of Research

The main focus of this study was to investigate the geological sequestration of CO_2 in deep saline formations and its associated risk using a series of laboratory experiments and numerical runs.

Specific objectives are:

1. To understand the multiple-phase flow through porous media under high pressure and salinity conditions.
2. To investigate the effect of cyclic CO_2-brine flooding on multiphase flow behavior during geosequestration under in-situ reservoir conditions.
3. To investigate the impact of CO_2-brine-rock interactions on dissolution/ precipitation of minerals.
4. To evaluate the release dynamics and associated risk assessment of CO_2 migration from subsurface to overlying atmosphere.

Schematic diagram of methodology



Core flooding experimental set-up

- Core flooding experiments have been performed using Grace flooding apparatus at TAMUQ.
- Rock and fluid samples are changed

in various set of experiments.

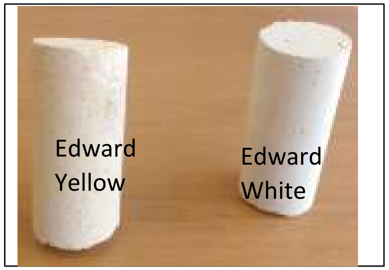
Table 1. Parameters table of in-situ reservoir conditions

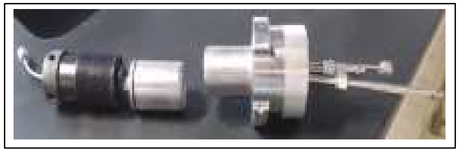
Core sample	Injection pressure	Salinity
CD1a	8 MPa	3 %
CD2a	8 MPa	3 %
CD1b	8 MPa	7.5 %
CD2b	8 MPa	7.5 %
CD1c	10 MPa	7.5 %
CD2c	10 MPa	7.5 %

*CD1: Edward White limestone **CD2: Edward Yellow limestone

Confining pressure of formation: 13.78 MPa

Methodology framework for core flooding experiments





Limestone core samples and adjustable end plugs to fit core samples

Vacuum chamber for saturation of cores and saturation cell

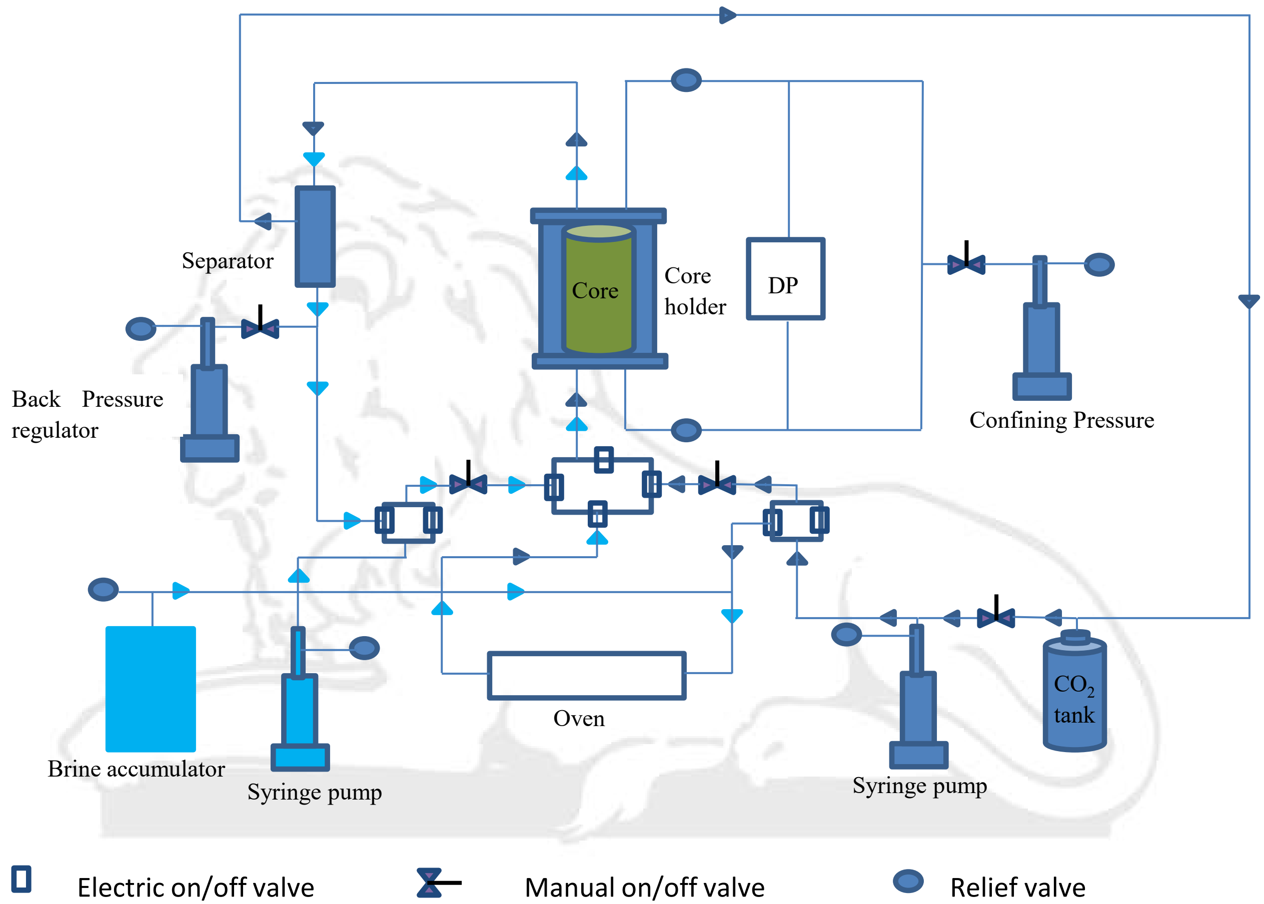
Front panel for maintaining in-situ reservoir conditions

Grace core flow tester for performing flooding experiments

Impact of:

- Salinity
- Injection pressure
- Cyclic flooding

Schematic of the flooding apparatus used



Methodology for image analysis

CO_2-brine-rock interaction using core images

- Reservoir formation was subjected to CO_2-water-rock interaction experiment
- The changes are observed using XRD, FeSEM-EDS and TEM method
- Petrophysical and Minerological changes were observed

Non-destructive way of Christological, chemical and physical properties; adsorption-desorption analysis, internal structure on nano-scale

X-Ray Diffraction

Field emission Scanning electron Microscopy

Transmission electron Microscopy

Modeling of CO_2 movement

Mass conservation equation for CO_2 movement involving wetting fluid (brine) and non-wetting fluid (CO_2):

$$\frac{\partial}{\partial t}\left(S_\gamma \phi \rho_\gamma\right) - \nabla.\left(\rho_\gamma v_\gamma\right) - \rho_\gamma q_\gamma = 0$$

Mass transport of CO_2:

$$\frac{\partial C}{\partial t} + v.\nabla C = \nabla.(D\nabla C)$$

Multiphase flow for wetting and non-wetting fluids

COMSOL Multiphysics

Transport of species in porous media

Impact of injection pressure and flux

Migration of CO_2

Concentration profile of CO_2

Risk assessment of CO_2 migration

- The theory of double effective stress of porous medium which considers the deformation mechanism and material structure of the porous medium

$$p_F = \frac{1}{1-\phi\left(\frac{1-2\mu}{2(1-\mu)}\right)+\phi_c}\left(\left(S_t + p_\pi + \left(2-\phi\left(\frac{1-2\mu}{2(1-\mu)}\right)\right)p_p + \frac{E\alpha_m}{3(1-\mu)}[T_w - T_o]\right) + \left(\frac{2\mu}{1-\mu}+3\beta-\alpha\right)S\right)$$

- It is combined with one-dimensional diffusivity equation

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial p}{\partial r}\right) = \frac{\mu c \phi}{2.634 * 10\text{^}(-4)K}\frac{\partial p}{\partial t}$$

$$\frac{\partial^2 p}{\partial r^2} + \frac{1}{r}\frac{\partial p}{\partial r} = \frac{\mu c \phi}{2.634 * 10\text{^}(-4)K}\frac{\partial p}{\partial t}$$

For the case of: infinite reservoir and constant rate injection solution of the above equation in dimensionless form is:

$$p_D = -\frac{1}{2}E_i(-\frac{r_D^2}{4t_D}) = -\frac{1}{2}\left[\ln\left(\frac{r_D^2}{4t_D}\right) + 0.5772\right]$$

Results of core flooding experiments

- Preliminary experiments have also been performed for estimating the porosity and permeability of the samples used.

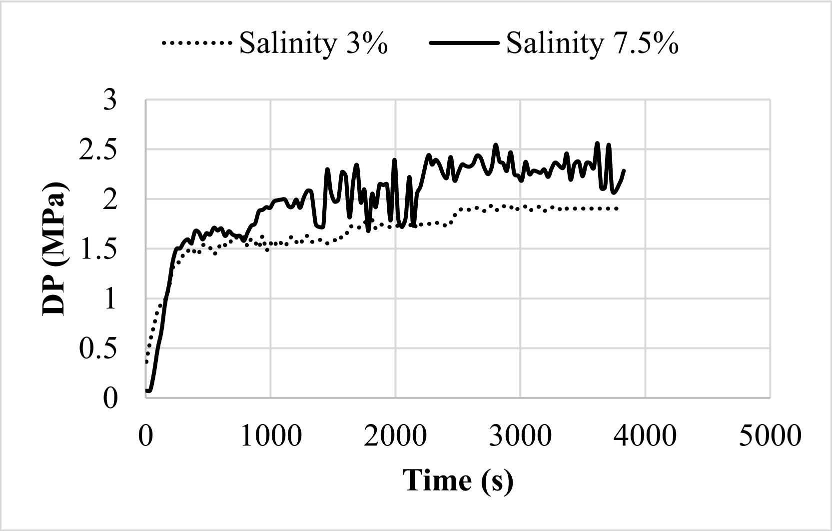
Table 2. Porosity calculation table

Core sample	Length (cm)	Diameter (cm)	Area (cm_2)	Bulk Volume	Dry Weight	Saturated Weight	Porosity	Permeability, k (md)-millidarcy
CD1a	15.765	4.275	14.359	226.376	337.780	383.390	19.75	1.2
CD2a	15.747	4.271	1.333	225.695	348.628	388.123	17.15	8.1
CD1b	14.342	3.789	11.284	161.838	317.018	361.99	26.33	1.3
CD2b	15.316	3.738	10.983	168.228	347.621	388.591	23.19	9.7
CD1c	13.487	3.789	11.284	152.194	296.587	337.557	25.54	1.38
CD2c	14.351	3.738	10.983	157.626	327.477	364.427	22.24	8.2

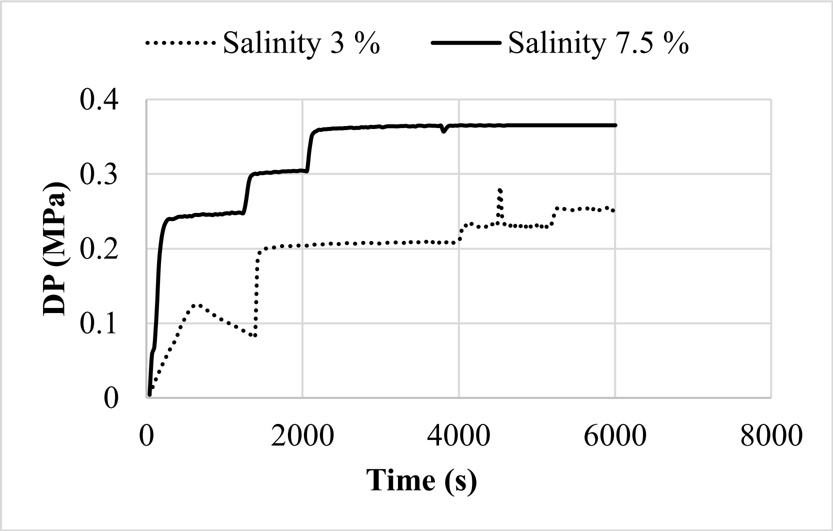
- Permeability was estimated using Darcy's law:

$$q = \frac{KA}{\mu L}(\Delta p)$$

Effect of salinity level



Differential pressure across the **Edward white** sample for two salinity conditions

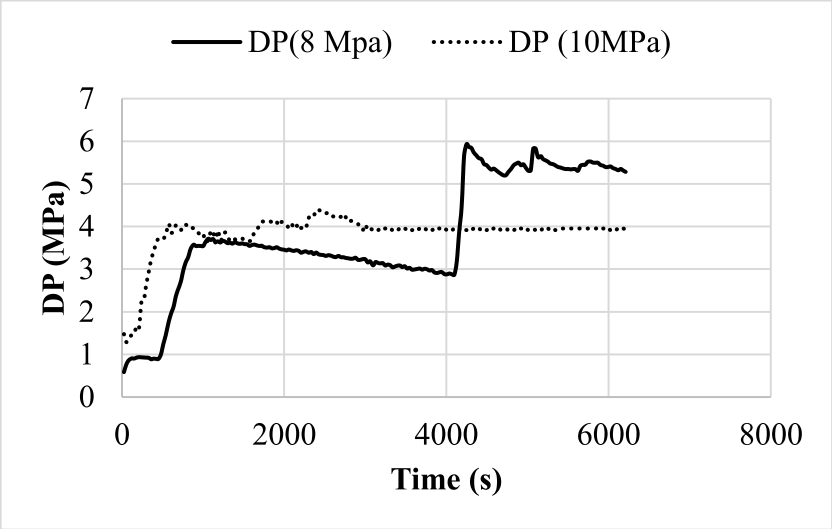


Differential pressure across the **Edward yellow** sample for two salinity conditions

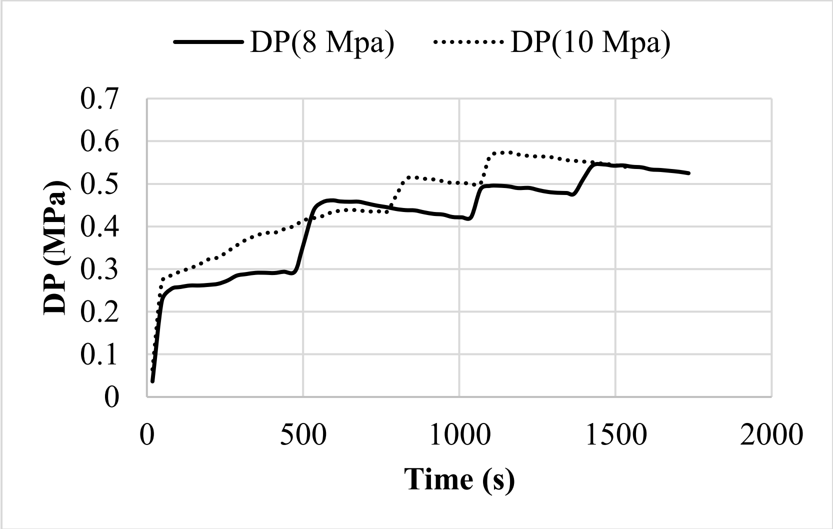
At high salinity condition the solubility of CO_2 in brine decreases due to precipitation of salts owing to the "salting-out-effect". The solubility of CO_2 in brine decreases with increasing salinity

Reference: Nighswander et al (1989), Rochelle and Moore, (2002) and Lee et al (2016)

Impact of injection pressure



Differential pressure across the **Edward white** sample for two injection pressure conditions

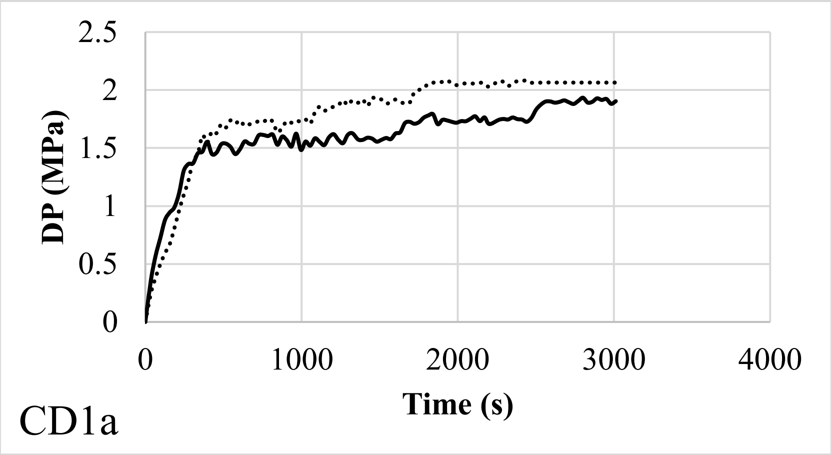


Differential pressure across the **Edward yellow** sample for two injection pressure conditions

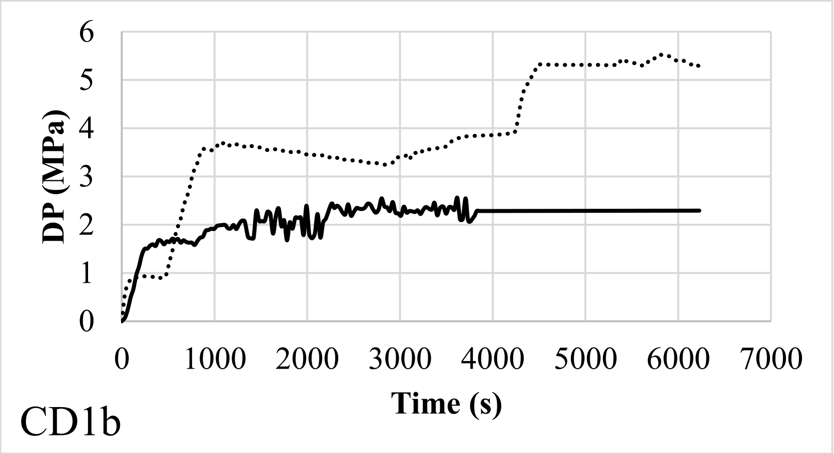
Pressure drop across the porous medium is low when low injection pressure was used and high when high injection pressure was used.
Phase change variation as the process involves injection of CO_2 in deep formations at high pressure to be in supercritical state, but CO_2 can exist in gas phase also.

Results: Differential pressure across cores for two brine injection cycles performed (CD1)

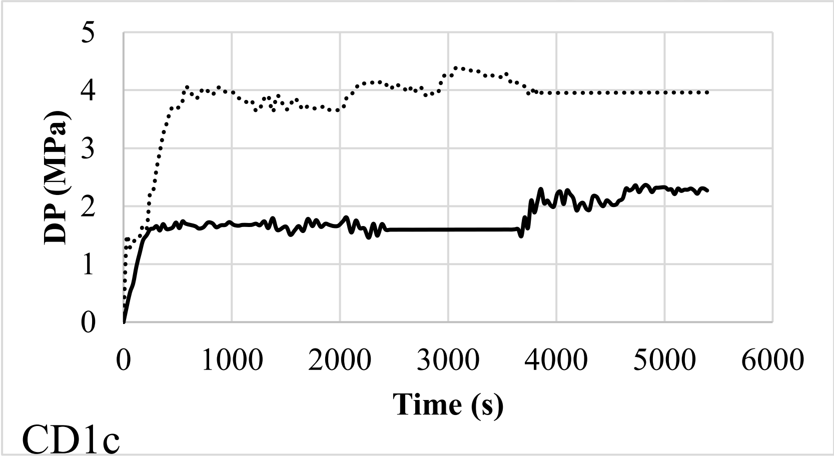
Salinity 3 % and injection pressure 8 MPa



Salinity 7.5 % and injection pressure 8 MPa



Salinity 7.5 % and injection pressure 10 MPa

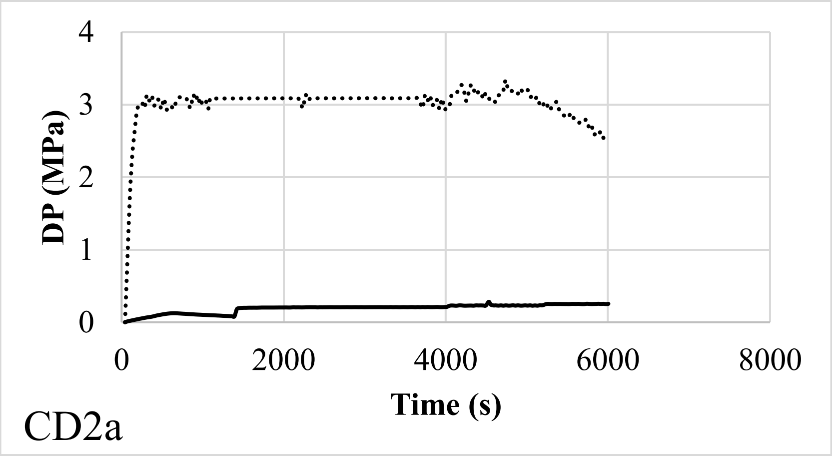


- The differential pressure first increases rapidly and the rate starts decreasing before it gets stabilized after a certain period of time.
- With consecutive injection cycles performed pressure drop across the porous medium increases which tends to level off gradually.

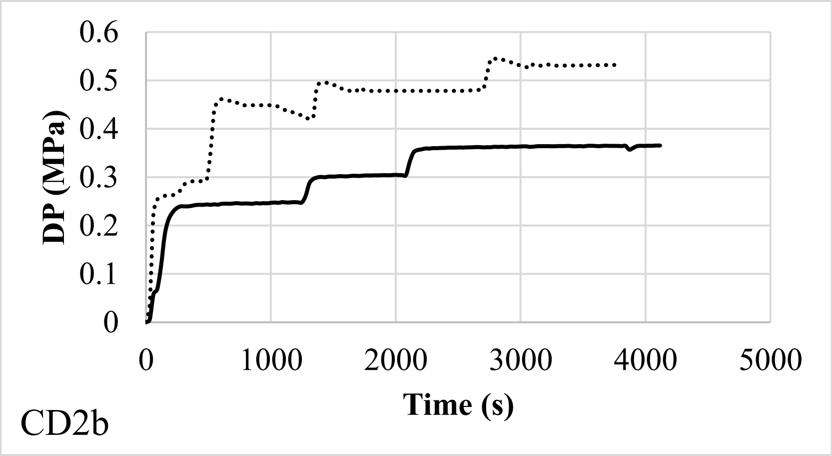
*solid line: 1st injection cycle
**dotted line: 2nd injection cycle

Results: Differential pressure across cores for two brine injection cycles performed (CD2)

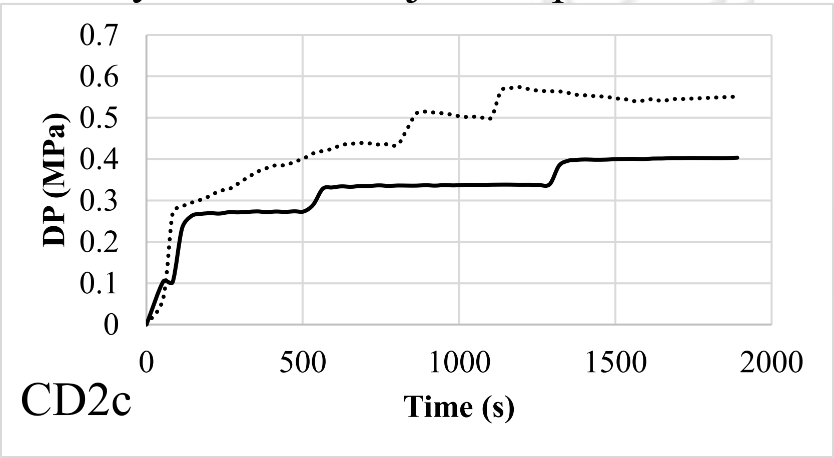
Salinity 3 % and injection pressure 8 MPa



Salinity 7.5 % and injection pressure 8 MPa



Salinity 7.5 % and injection pressure 10 MPa



*solid line: 1st injection cycle
**dotted line: 2nd injection cycle

- A smooth profile with some discrete steps of differential pressure increment is obtained which initially increased and then further stabilizes with time.

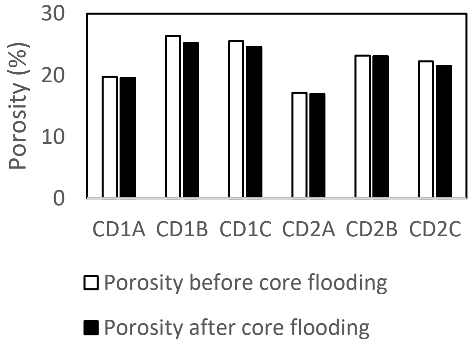
Reason:

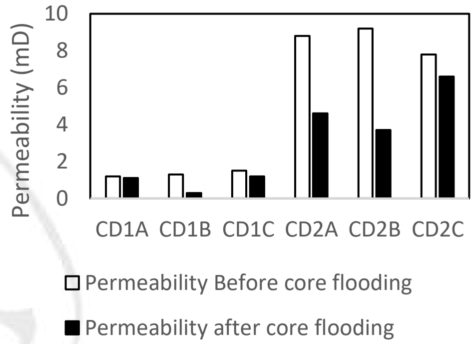
- Presence of acidic brine solution
- Hysteresis effect
- Physical compaction of the pore throats
- Existence of multiple phases

Characterization of petrophysical properties of cores

Change in porosity and permeability of core samples after flooding experiments:

S. No	Sample	Porosity Before	Porosity After	Porosity reduction (%)	Permeability Before	Permeability After	Permeability reduction (%)
1.	CD1a	19.753	19.559	1.0	1.2	1.1	8.3
2.	CD1b	26.339	25.198	4.3	1.3	0.3	76.9
3.	CD1c	25.540	24.585	3.7	1.5	1.2	20
4.	CD2a	17.156	16.936	1.3	8.8	4.6	47.7
5.	CD2b	23.194	23.106	0.4	9.2	3.7	59.8
6.	CD2c	22.240	21.535	3.2	7.8	6.6	15.4

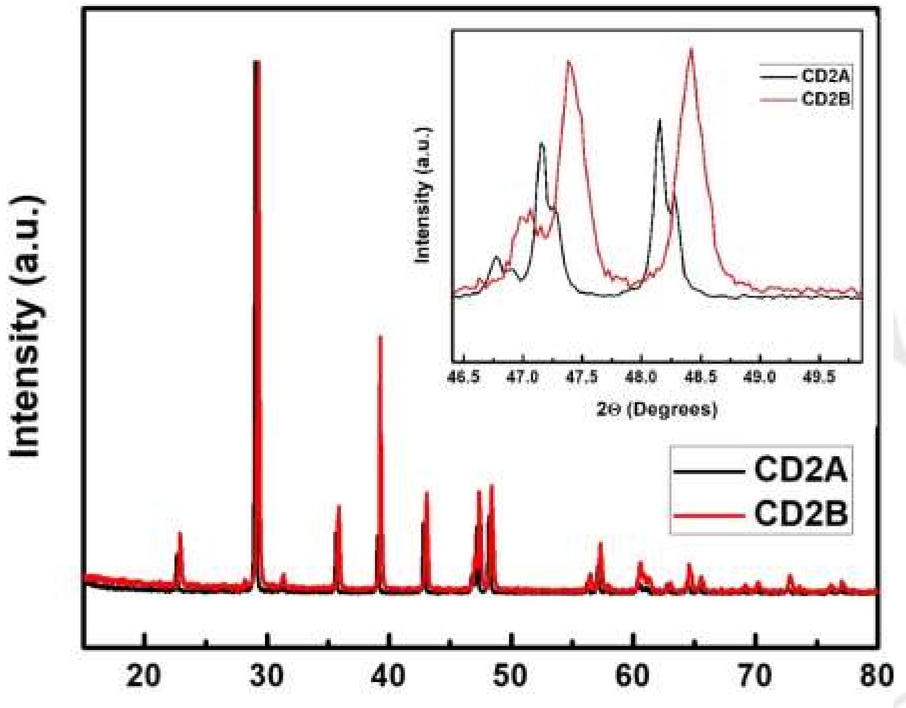




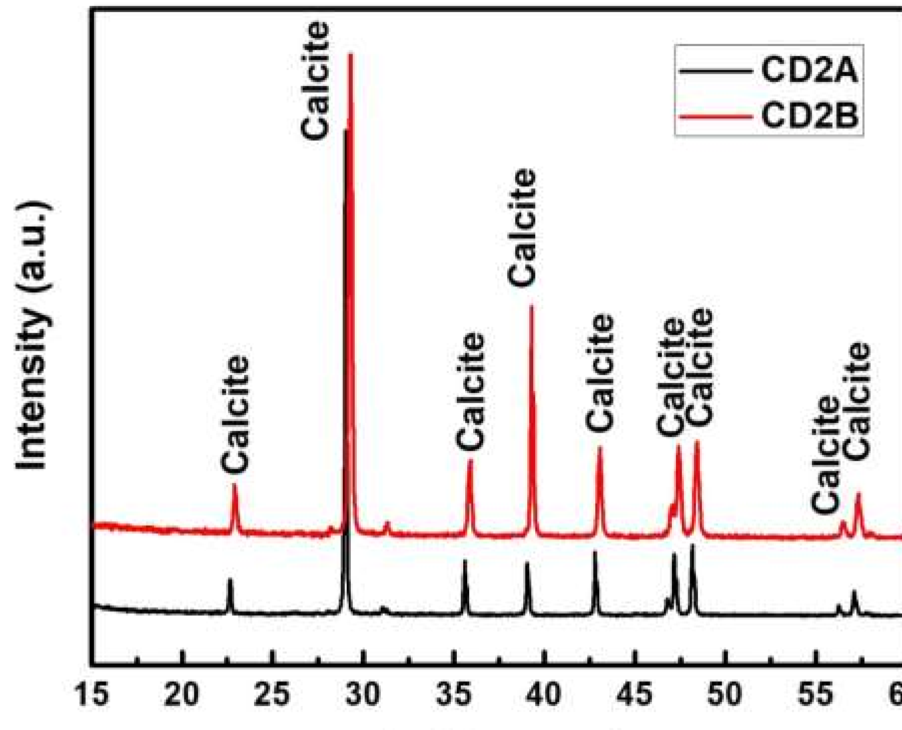
The results show decrease in porosity values after the flooding experiments in a range of 5% than the original values. The results are in accordance with the Saeedi, 2012 study performed on sandstone core samples.

XRD analysis

XRD analysis of Edward yellow limestone core sample before and after performing the flooding experiments.



(c)



(d)

B: before flooding A: after flooding

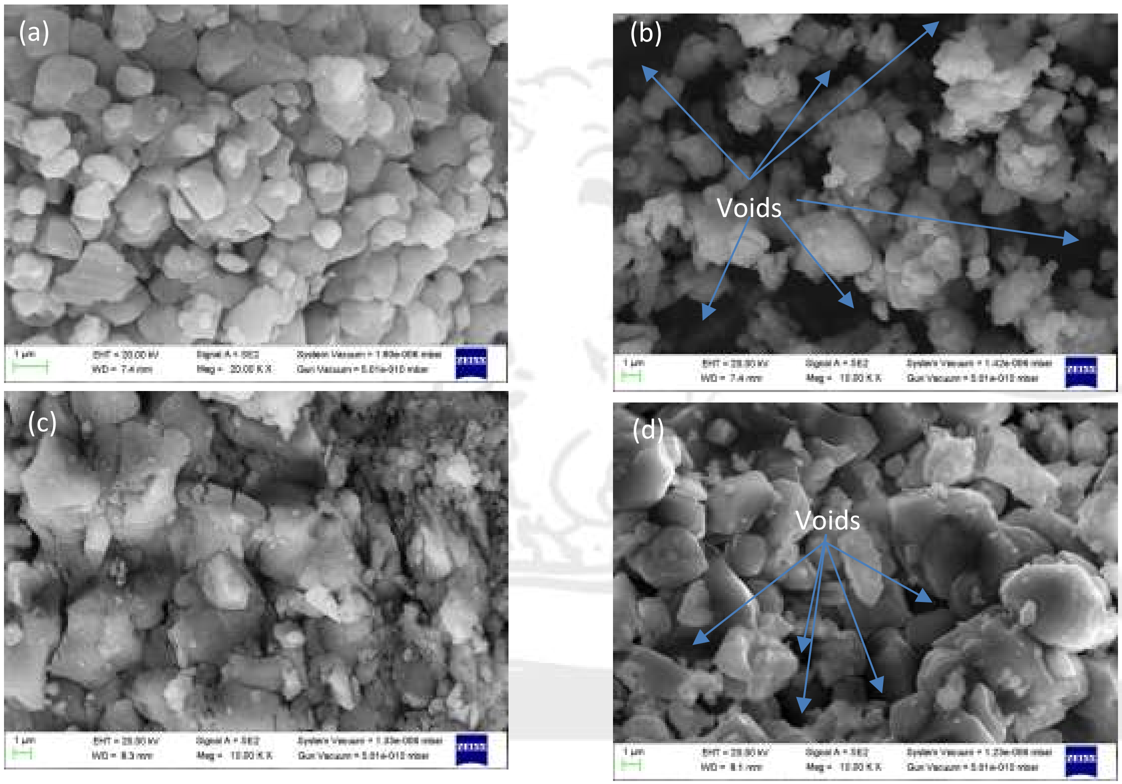
From the graph c it can be observed that the peak before the flooding experiments are more intense as compared to the other one.

Reference: Al-Jaroudi et. al, 2007

Results: FeSEM images of core samples

FeSEM analysis (Dissolution/Precipitation)

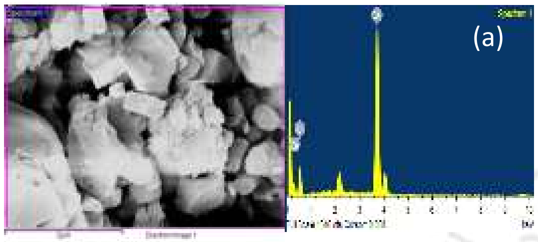
Before flooding | **After flooding**



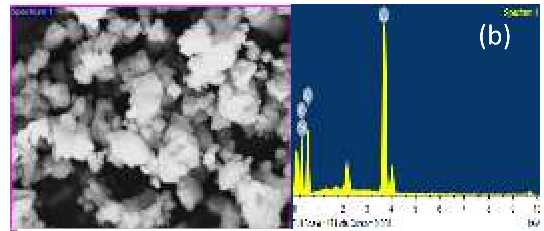
Dissolution of carbonate minerals is visible after flooding experiments on both Edward white and Edward yellow carbonate core samples

EDX results of CD1 core sample

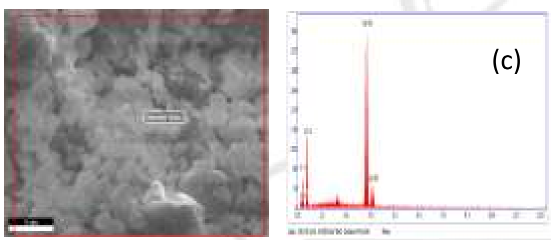
Edward white core sample before and after flooding experiment



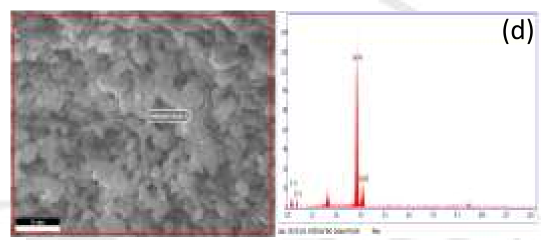
Element	Weight%	Atomic%
O K	54.23	74.80
Ca K	45.77	25.20
Totals	100.00	



Element	Weight%	Atomic%
C K	21.16	31.43
O K	49.95	55.71
Ca K	28.89	12.86
Totals	100.00	



Element	Weight %	Atomic %	Net Int.	Error %
C K	14.73	23.2	24.03	10.59
O K	51.44	60.83	56.95	12.43
CaK	33.83	15.97	211.79	2.92

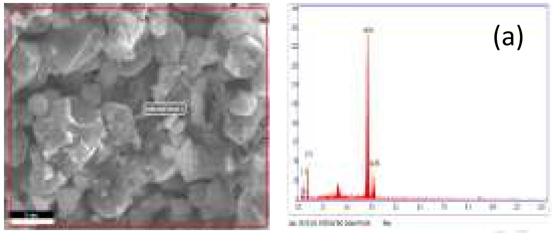


Element	Weight %	Atomic %	Net Int.	Error %
C K	11.37	24.19	4.12	16.73
O K	20.13	32.15	2.82	24.29
CaK	68.49	43.66	93.59	3.35

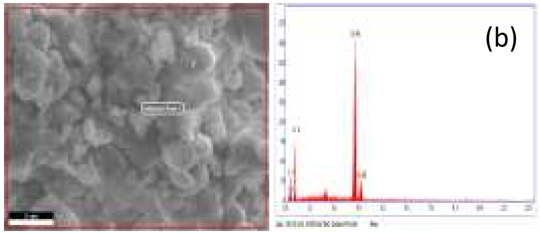
Weight percentage of element changes after performing the flooding experiment
Ca decreases as well as increases after flooding.

EDX results of CD2 core sample

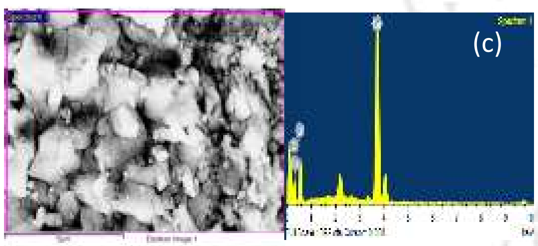
Edward yellow core sample before and after flooding experiment



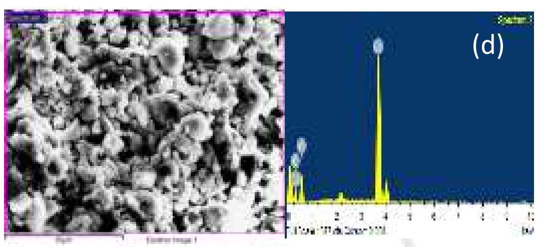
Element	Weight %	Atomic %	Net Int.	Error %
C K	11.58	20.45	12.34	12.76
O K	41.13	54.52	23.66	14.63
CaK	47.29	25.03	194.12	2.92



Element	Weight %	Atomic %	Net Int.	Error %
C K	14.88	23.99	19.03	10.57
O K	47.98	58.07	38.42	12.73
CaK	37.15	17.95	181.38	2.48



Element	Weight%	Atomic%
C K	9.42	15.58
O K	53.00	65.80
Ca K	37.58	18.62
Totals	100.00	

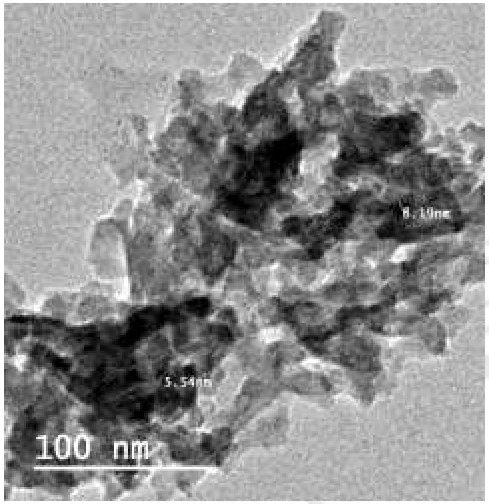


Element	Weight%	Atomic%
C K	10.34	17.48
O K	48.65	61.74
Ca K	41.01	20.77
Totals	100.00	

Weight percentage of element changes after performing the flooding experiment (Ca decreases as well as increases after experiment)

Results: TEM analysis and SAED of Edward white limestone core sample

Before flooding



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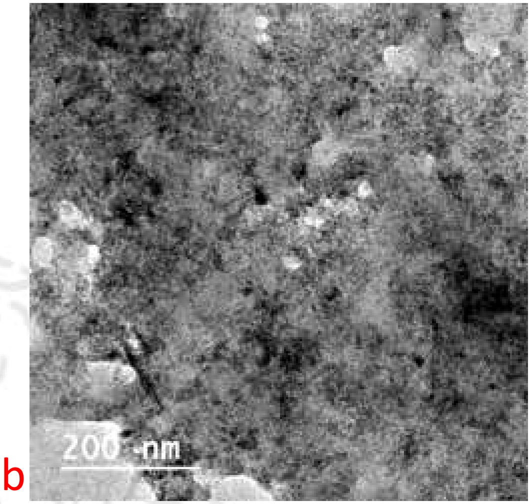

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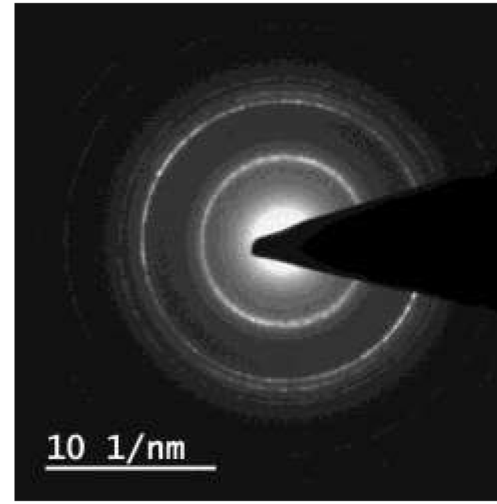
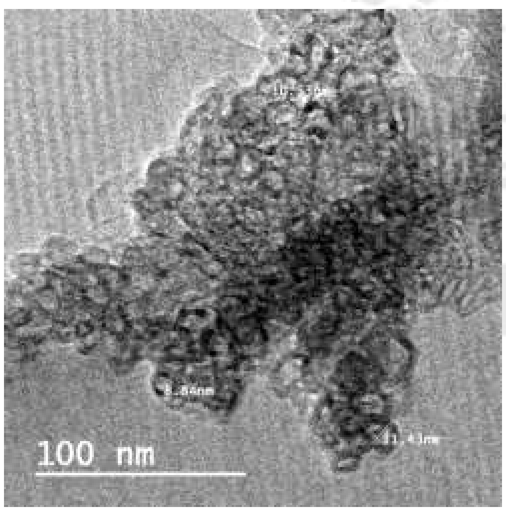


c

After flooding



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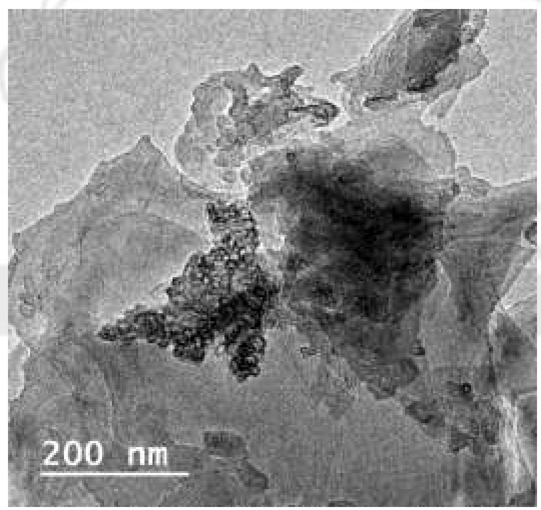
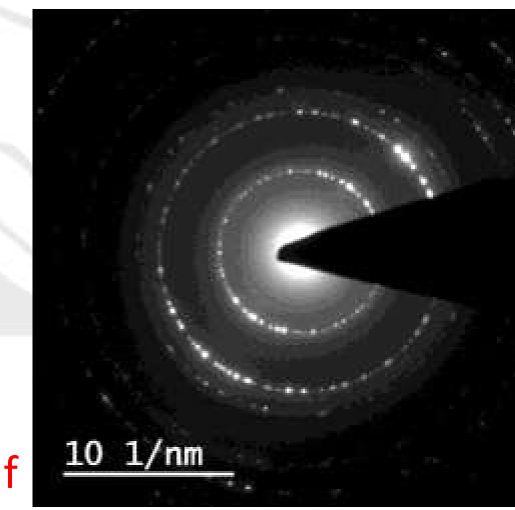


e



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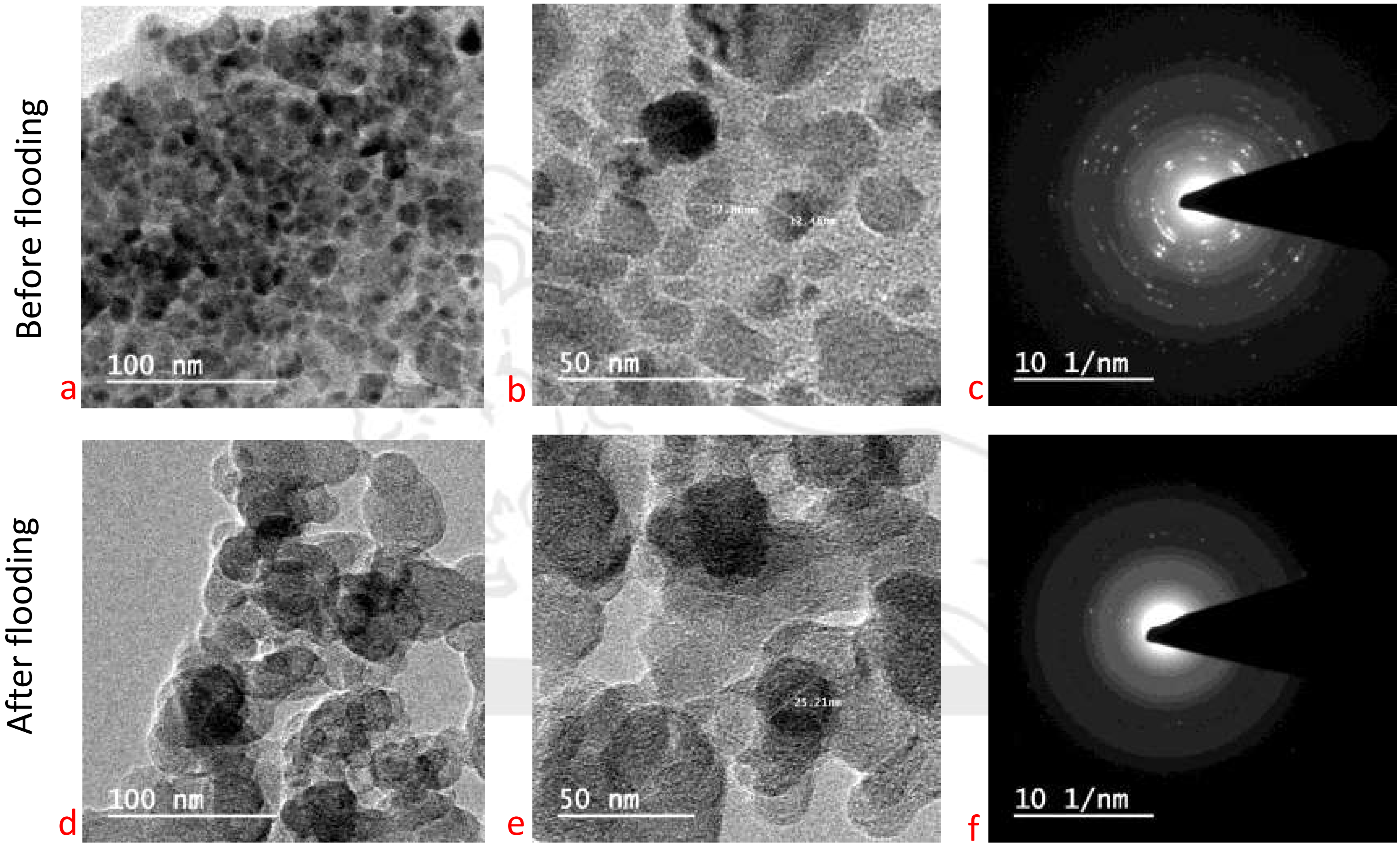
Results: TEM analysis and SAED of Edward yellow limestone core sample



Discussions

- There is dissolution as well as precipitation of carbonate minerals in cores

- Petrophysical changes indicates towards dominance of precipitation process

- Edward white average pore size observed:
  Before flooding: 5-8 nm.
  After flooding: 8-11.5 nm
- Edward yellow sample:
  Before flooding: 12 nm
  After flooding: 25 nm

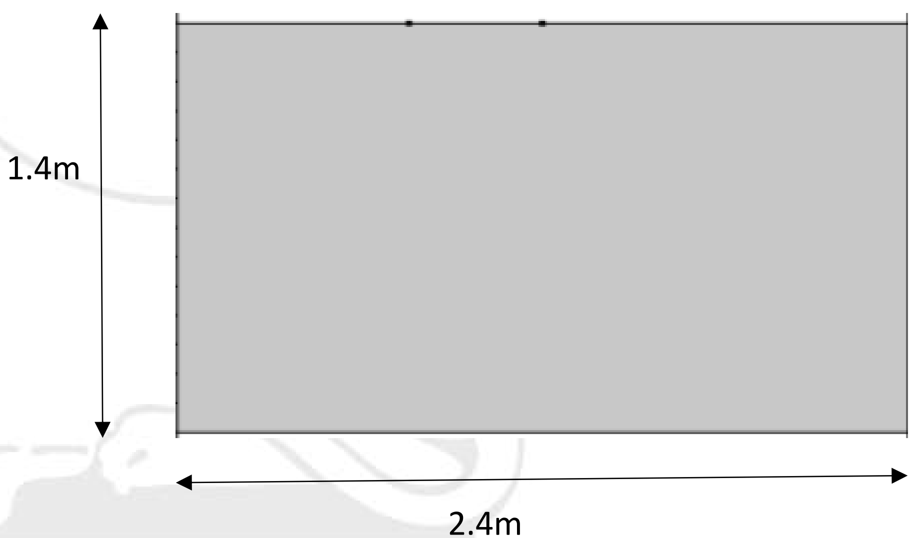
- The pore size of both the carbonate cores used increases after performing flooding experiments that indicates towards dominant dissolution process.

- Formation of rings in SAED of TEM images show that the material is crystalline, which is also confirmed from the sharp peaks obtained in XRD

Model formulation

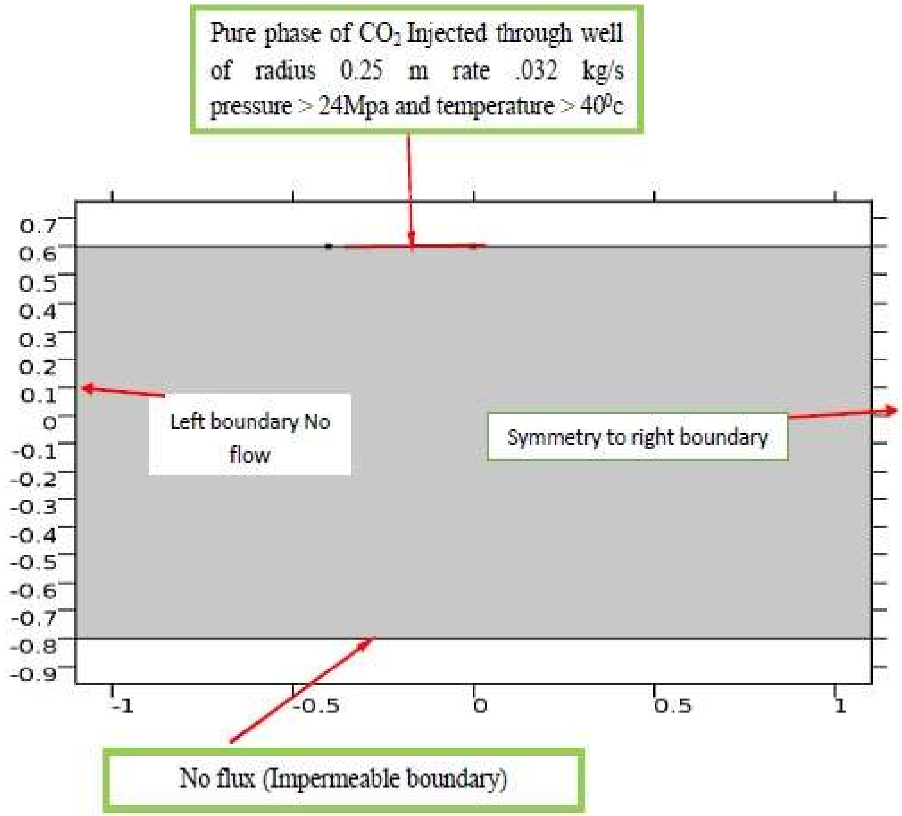
- Darcy's law: Vertical flow.
- Saline water and CO_2 are two immiscible fluid.
- CO_2flux is injected through upper boundary well and lower boundary has no flux
- Left boundary: no flow condition
- Right boundary is symmetry to the left boundary

Domain geometry:- 2D domain of width 2.4 m and depth 1.4 m



Boundary Conditions:

Dirichlet type of boundary conditions are considered in this study for both lower and upper boundaries (P_0=0, P_1 and P_2=28 to 36 Mpa , T_0=0 and C_0=0, C_1=1).

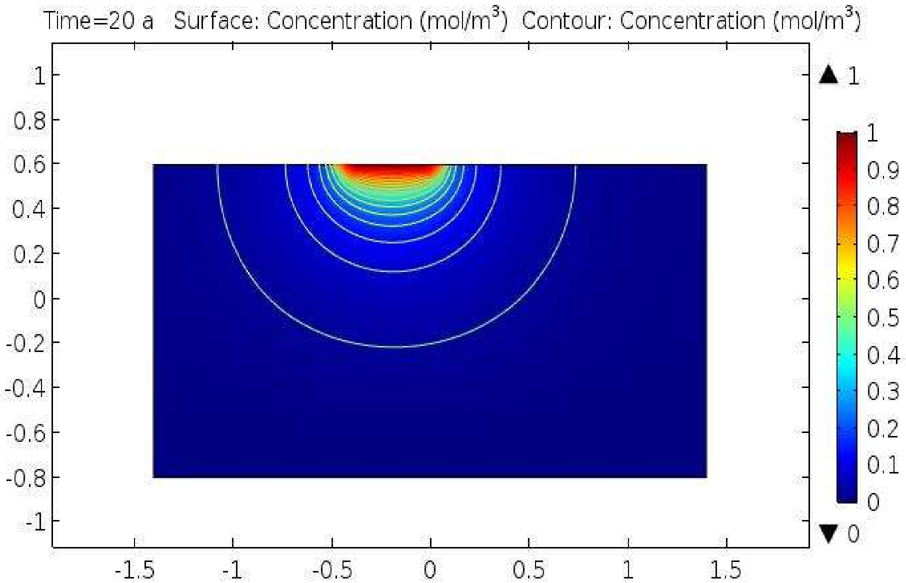


Model input parameters:

Parameters can be obtain by well logging and core flooding experiments			
PROPERTIES	SYMBOL	VALUE	UNIT
Porosity	Φ	10-20 %	
Permeability	k	200	mD
Density of saline water	ρ_s	1200	kg/m^3
Density of CO_2	P_{CO2}	500-900	kg/m^3
Viscosity of saline water	μ_s	0.00039	kg/(m.s)
Viscosity of CO_2	μ_{CO2}	0.0025	kg/(m.s)
Gravity	g	9.81	m/s^2
Injection rate	i	0.32	kg/s
Time of injection	t_i	20	year
Pressure at injection point	P_i	28	Mpa
Initial reservoir Temperature	T	50	^0C
Initial reservoir Pressure	P_i	36	Mpa
Area of reservoir	A	2.4x 1.4	m^2
Storage efficiency	E	0.1-6%	
Diffusion co-efficient	De	$2.49x10^{-9}$	m^2/s

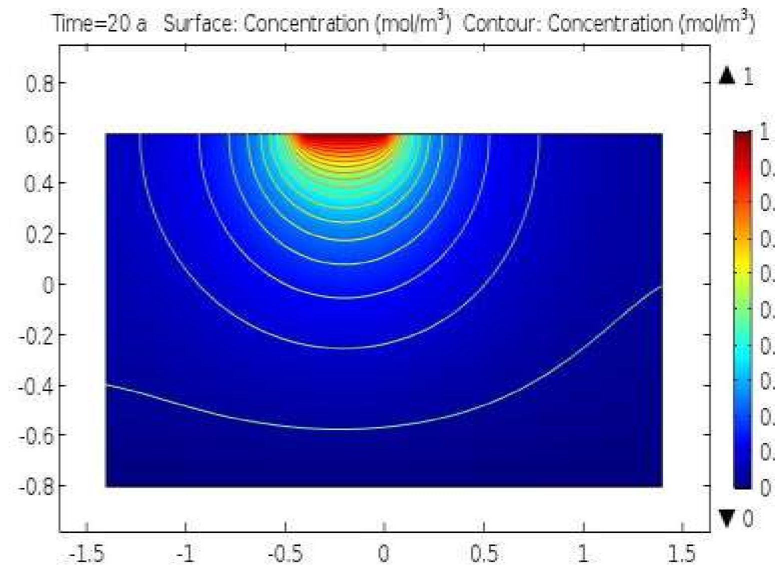
Results of CO2 Migration at different pressure

Case 1: When subsurface domain pressure kept 36 MPa and pressure of injection point kept 28 MPa



At t=20 yr Concentration profile of CO_2 migration in subsurface

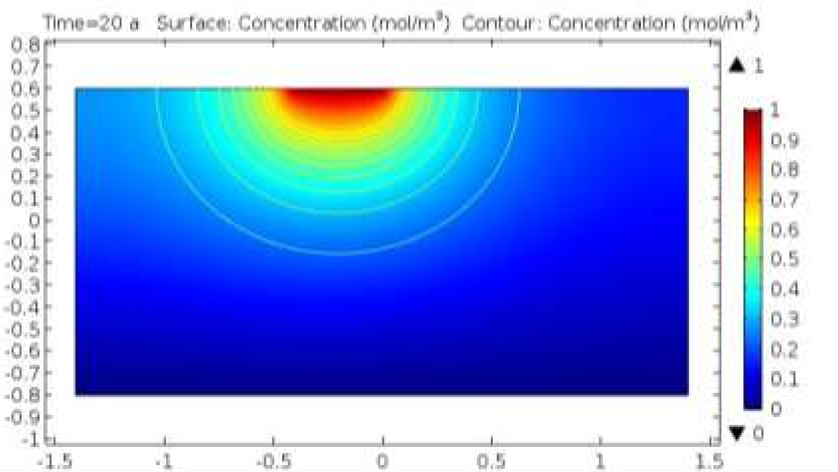
Case 2: When subsurface domain pressure kept 36 MPa and pressure of injection point kept 32 MPa



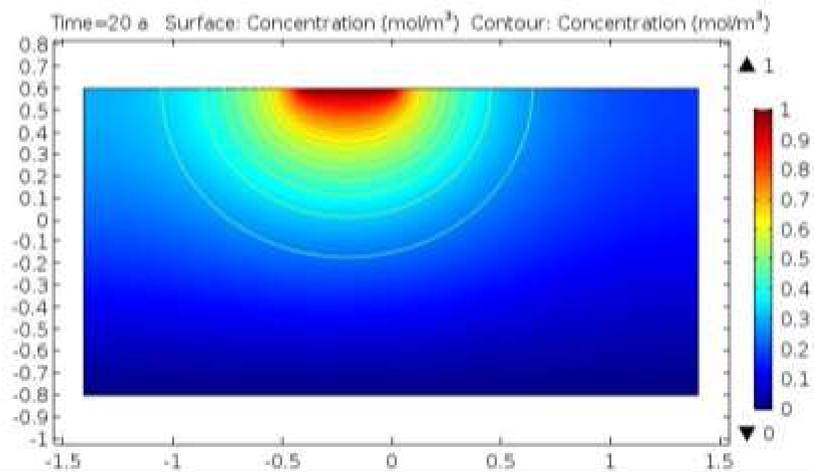
At t=20 yr Concentration profile of CO_2 migration in subsurface

Results: Impact of temperature

Concentration contours are plotted for two different temperatures, 50^0c and 60^0c respectively



At temperature 50^0c *and t=20 yr Concentration profile of* CO_2 *migration in subsurface*

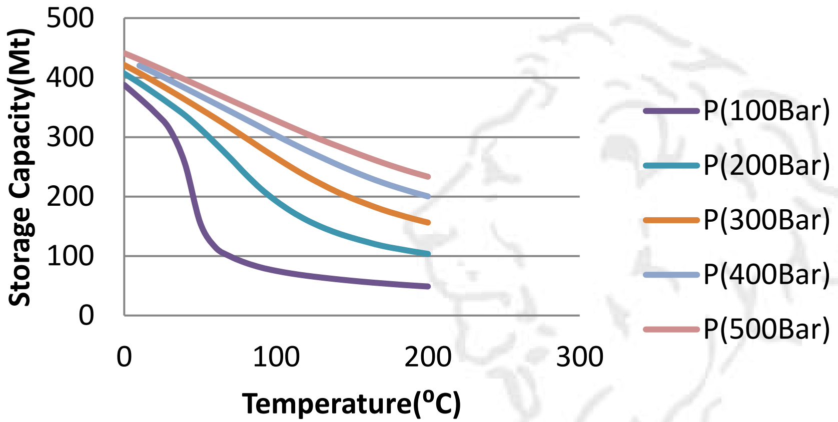


At temperature 60^0c *and t=20 yr Concentration profile of* CO_2 *migration in subsurface*

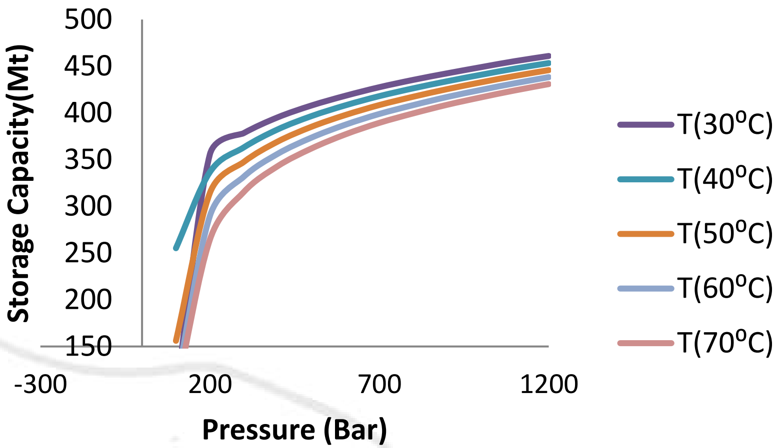
Results show that subsurface temperature of the geological formation is the less sensitive parameter as compared to pressure.

Impact of Temperature

Storage capacity depends on density which is function of subsurface temperature and pressure.



Relationship between storage capacity and temperature at different pressure



Relationship between storage capacity and pressure at different temperature

Storage capacity increases with increase in subsurface pressure, while, it decreases for higher temperature

Pressure is more sensitive parameter than temperature

Conclusions and Way Forward

- Core flooding experiments performed for multiple injection cycles under in-situ reservoir conditions to evaluate the effect of salinity, injection pressure and temperature on CO_2 fate in saline carbonate formations.

- Such experiments can be extended for a greater number of injection cycles of CO_2 and brine for getting detailed long term information about the solubility trapping, and CO_2 migration.

- Core samples having permeability beyond the range of selected cores (i.e., 1–10 mD) can be used for wide application of such types of findings in the field conditions.

- The petrophysical changes are observed clearly along with mineralogical changes in the form of XRD, FeSEM, and TEM images on selected cores.

Conclusions and Way Forward

- Upscaling from core Reservoir scales is needed to include subsurface heterogeneity, for improved filed data availability, and validation

- Use of similar modeling approach H2 storage in subsurface can be planned for its enhanced availability in pure phase.

- Simulation study can be improved by incorporating all the trapping mechanisms and further quantifying the different trapped forms of CO_2 under a given set of site specific salinity, pressure and temperature conditions.

- Studying other formations and potential sites like abandoned coal mines, oil production sites for enhance methane and oil recovery needed to explored with associated uncertainties and risks.

Thank You

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Publications

Journals:

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- Shachi, Yadav B. K., (2020), "Core flooding experiments for CO_2 geosequestration: Unravelling CO_2 interactions with deep saline carbonate aquifers" ASCE: International Journal of Geomechanics (Under review)
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- Shachi, Yadav B. K., Rahman M. A., Pal M. (2019), "CO_2-brine-rock interaction: dissolution / precipitation of carbonate minerals during geological sequestration". European Geosciences Union General Assembly (EGU-2019), Vienna, Austria, 7-12 April, 2019
- Shachi, Yadav B. K., Rahman M. A., Pal M. (2018), "Multiphase flow behavior during CO_2 sequestration in saline carbonate formations using core flooding experiment". American Geophysical Union Fall Meeting (AGU2018), Washington D.C., 10-14 December 2018
- Shachi, Yadav B. K., Rahman M. A. (2018), "Geological CO_2 storage in saline carbonate formations using core flooding experiment". European Geosciences Union General Assembly (EGU-2018), Vienna, Austria, 8-13 April, 2018
- Shachi, Yadav B. K., Rahman M. A., Pal M. (2018), "Multiphase flow behavior during CO2 geo-sequestration in carbonate formations". The Eleventh international conference on thermal engineering theory and applications (ICTEA-2018), Doha Qatar, 25-28 February, 2018
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- Shachi, Kumar V., Yadav B. K., Pal M. (2017), "Sequestration of CO_2 in subsurface under varying geological conditions". *Proceedings of International Conference on Modeling of environmental and water resources systems (ICMEWRS-2017),* HBTU Kanpur, 24-26th March, 2017 (ISBN 978-93-85926-53-2).