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Gordon Research Seminar

DTU

Visualization of CH_4 Hydrate Dissociation Under Permafrost Temperature Conditions Using High-Pressure Micromodel*

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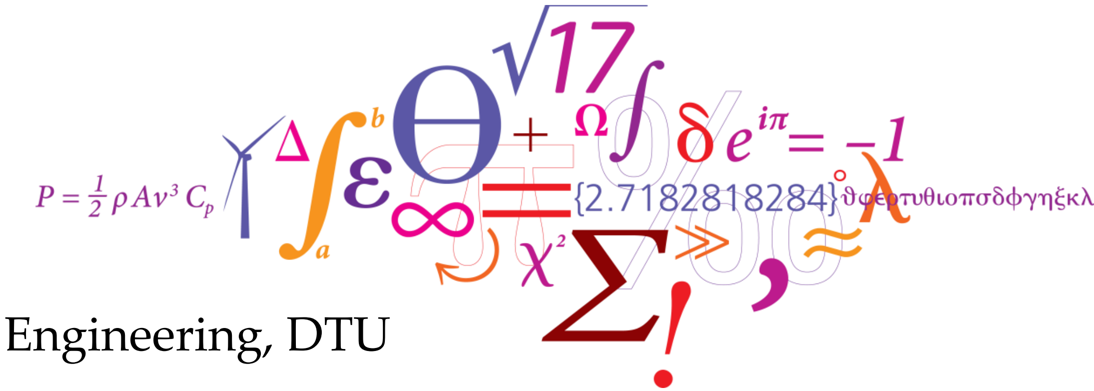

* Experimental work is performed at University of Bergen

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Gas Hydrate Studies using High Pressure Micromodels

Methane hydrate dissociation below subzero

CH_4/CO_2 Mixed Hydrate, Formation & Dissociation Behavior

Oral Presentation in GRS

Poster Presentation in GRC

CO_2 Injection into Methane Hydrate

Low Saturation
High Saturation
Liquid CO2
Gaseous CO2

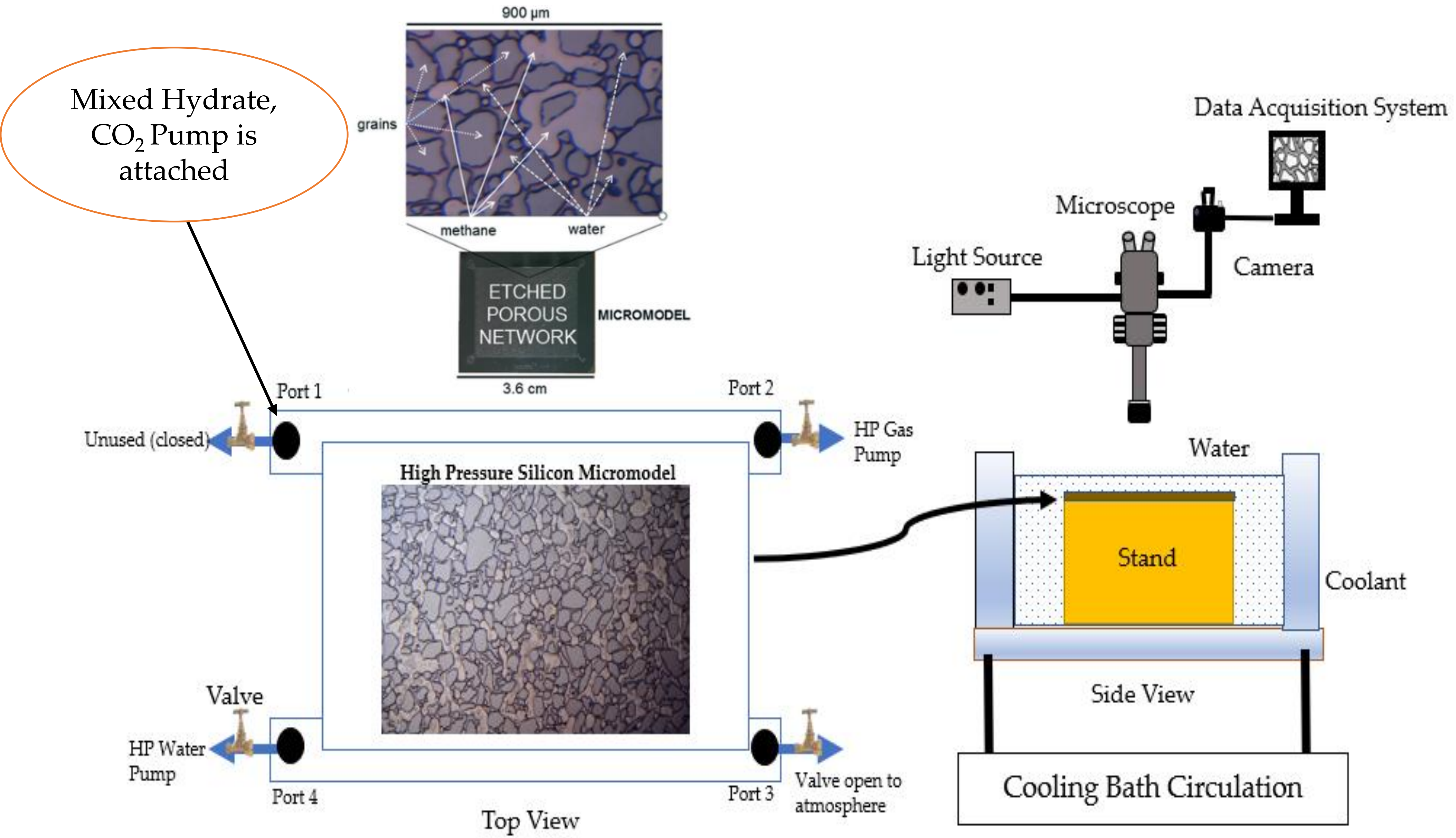
Supporting Studies

- Almenningen, S.; Flatlandsmo, J.; Kovscek, A.R.; Ersland, G.; Fernø, M.A. Determination of pore-scale hydrate phase equilibria in sediments using lab-on-a-chip technology. *Lab Chip* **2017**, *17*, 4070–4076.
- Almenningen, S.; Iden, E.; Fernø, M.A.; Ersland, G. Salinity Effects on Pore-Scale Methane Gas Hydrate Dissociation. J. Geophys. Res. Solid Earth 2018, 123, 5599–5608.
- Almenningen, S.; Gauteplass, J.; Fotland, P.; Aastveit, G.L.; Barth, T.; Ersland, G. Visualization of hydrate formation during CO2 storage in water-saturated sandstone. Int. J. Greenh. Gas Control 2018, 79, 272–278.

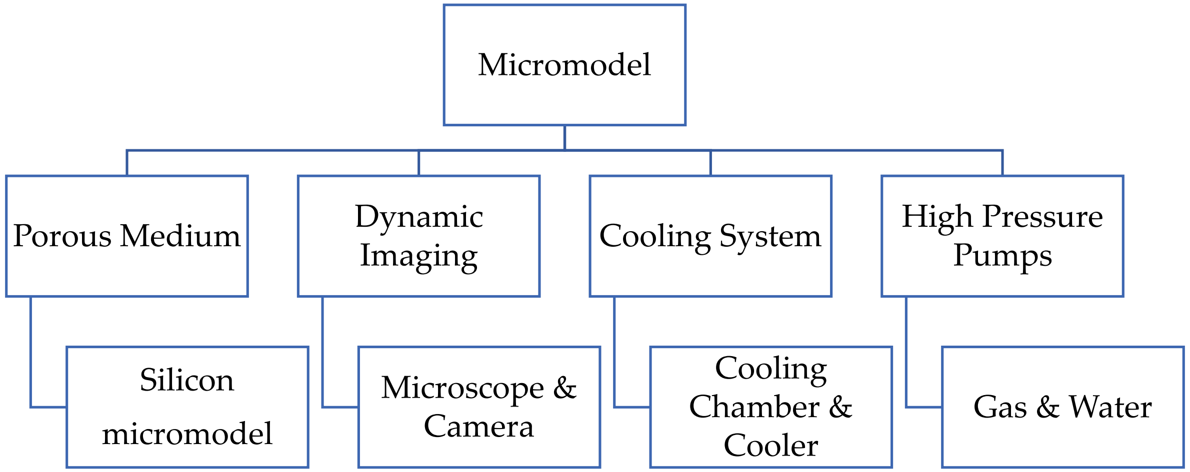
Methodology

1. Hydrate formation above 0°C
2. Cooling down to near 0°C or below 0°C
   - To observe the ice formation / change in the hydrate texture
3. Dissociation by pressure reduction
   - To observe the dissociation behavior below 0°C and/or in the presence of ice/supercooled water
4. Dissociation at constant pressure
   - To observe the dissociation behavior below the stability pressure in the presence of ice/supercooled water
5. Dissociation by thermal stimulation

Setup Diagram



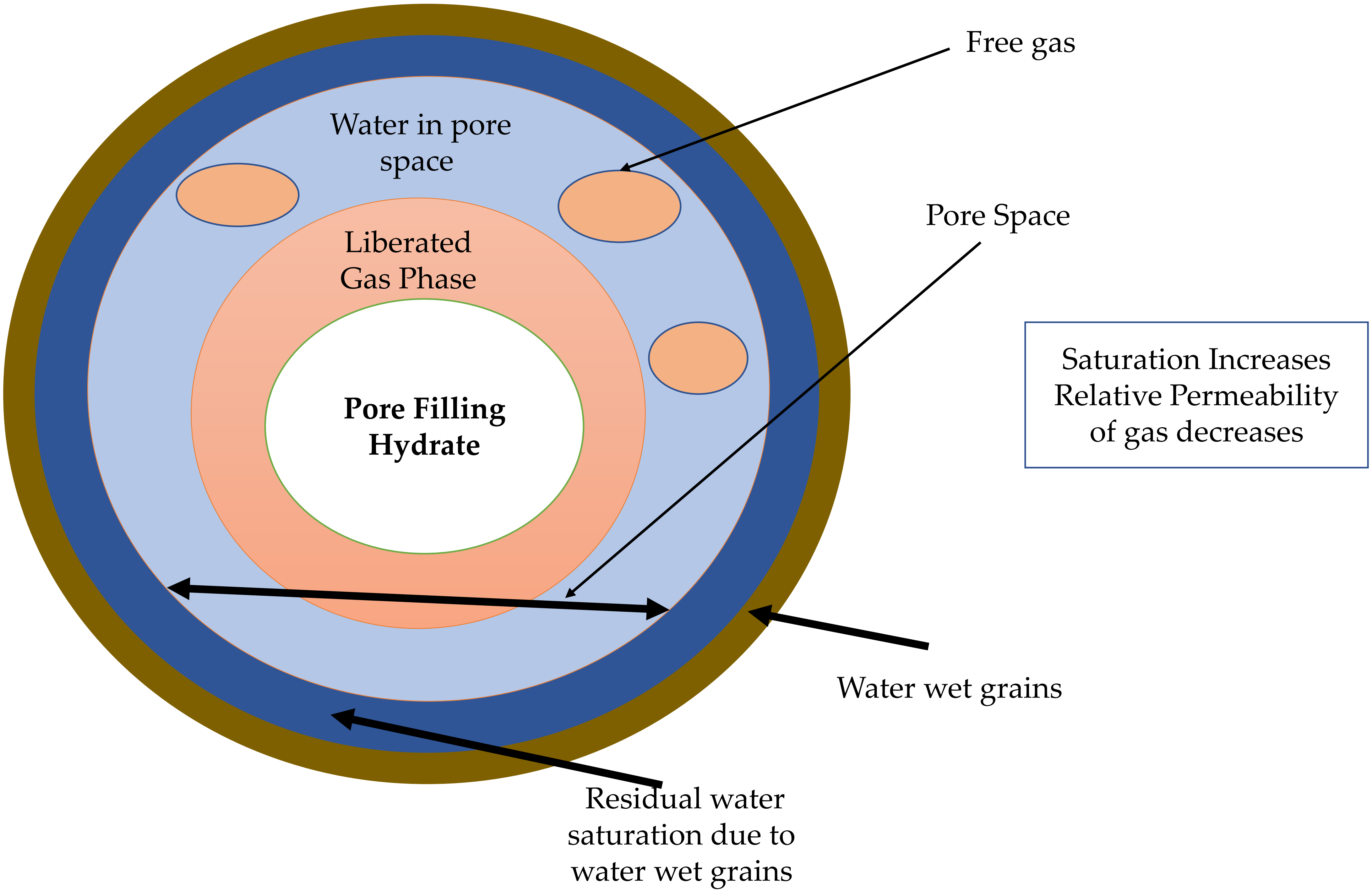
Laboratory setup

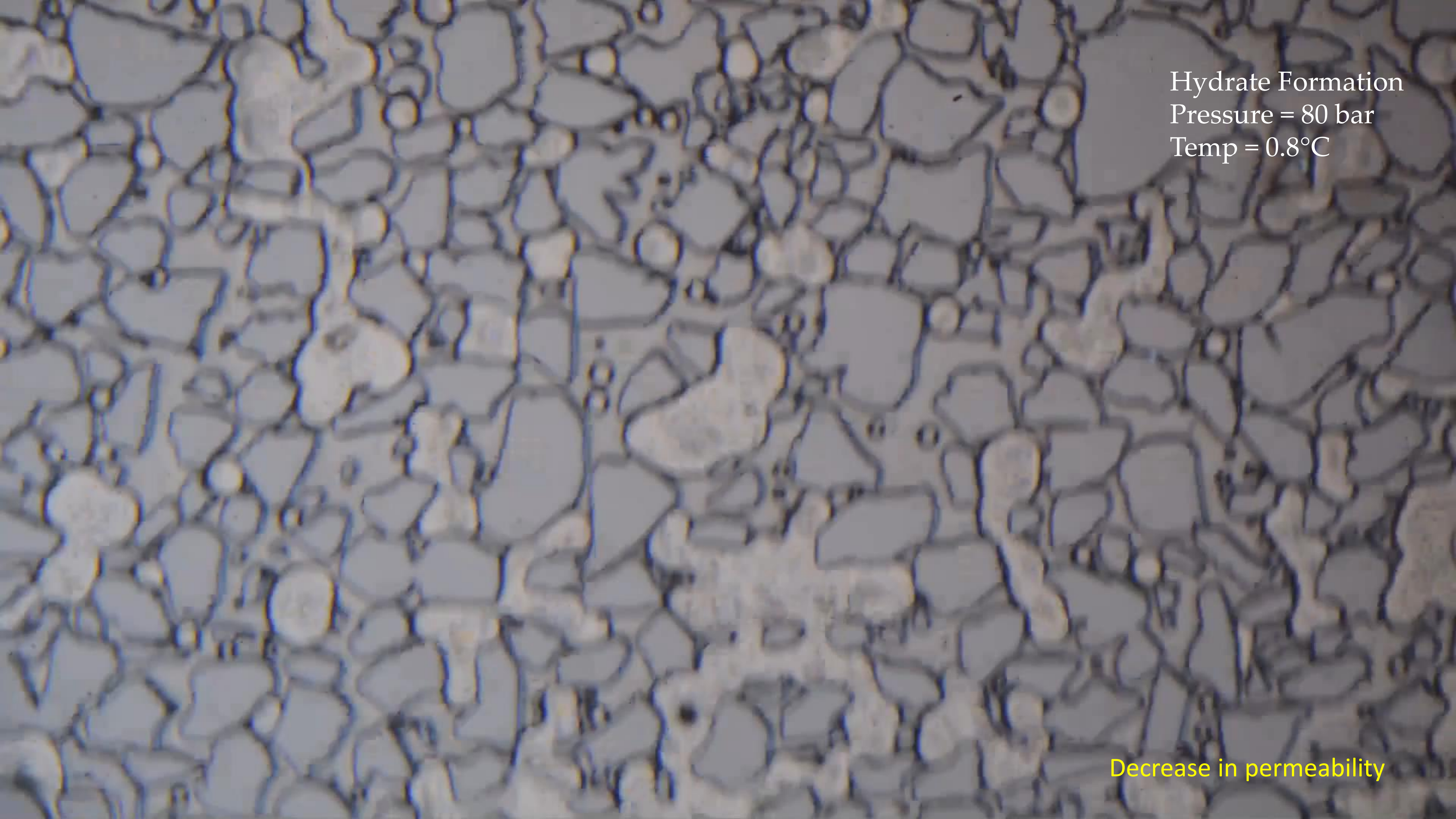


- Silicon wafer etched thin section (DRIE)
- Pore network of Berea Sandstone
- Water wet
- Constant vertical depth = 25 μm
- Porosity=0.61
- Pore diameter = 100 μm
- High Pressure= 100 bar
- Borosilicate glass, anionic bonding, oxide layer
- Aluminum manifold with nanoport

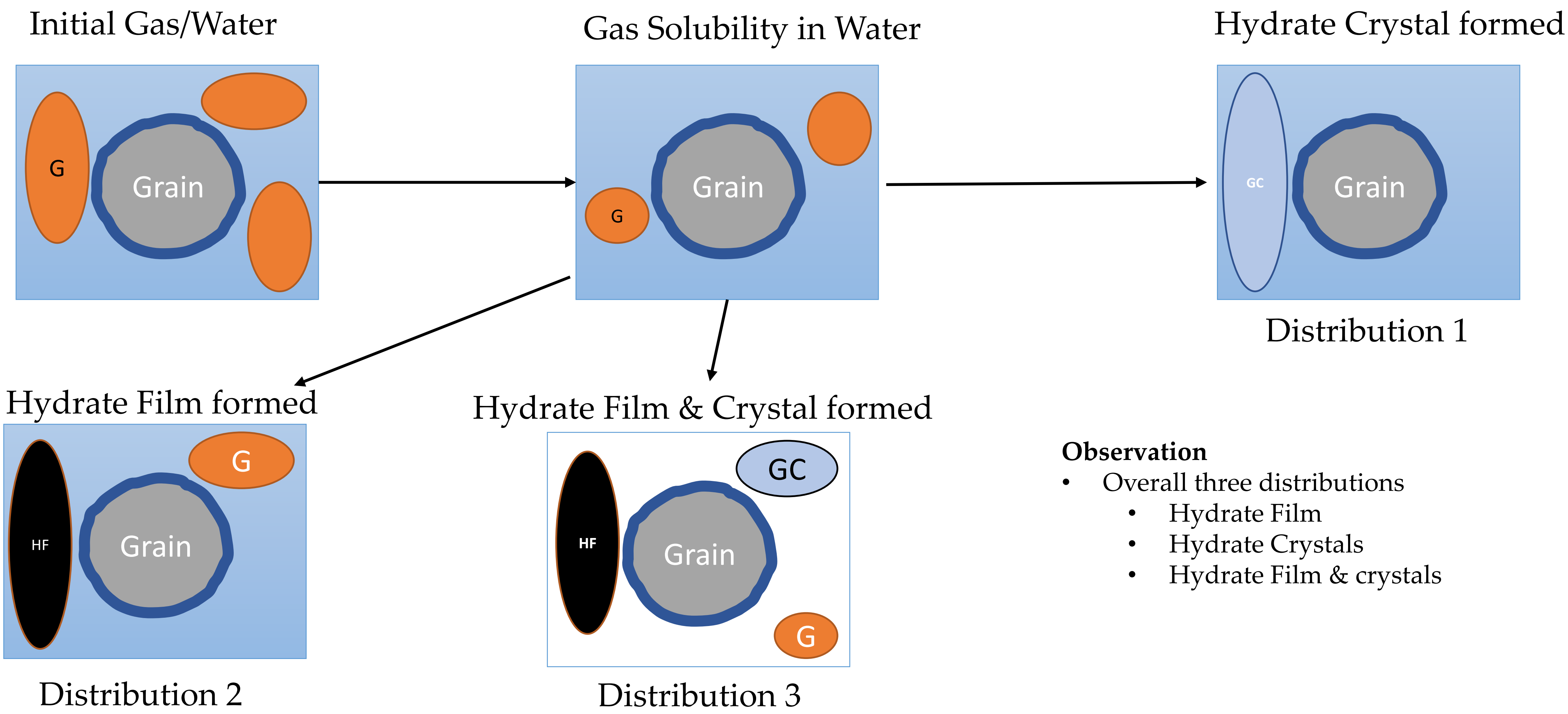
Capillary pressures were insignificant as pore sizes > 1 μm

Water wet, so pore filling (PF) hydrates observed


Free gas
Water in pore space
Pore Space
Liberated Gas Phase
Pore Filling Hydrate
Saturation Increases Relative Permeability of gas decreases
Water wet grains
Residual water saturation due to water wet grains

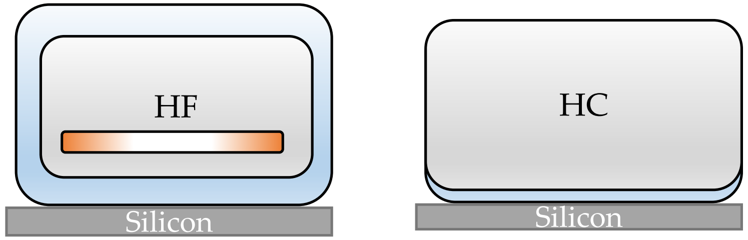

Hydrate Formation
Pressure = 80 bar
Temp = 0.8°C
Decrease in permeability

Formation Mechanism



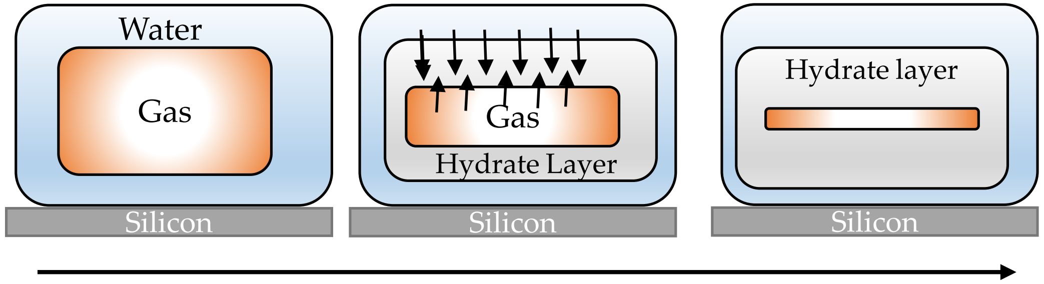
Observation

- Overall three distributions
  - Hydrate Film
  - Hydrate Crystals
  - Hydrate Film & crystals

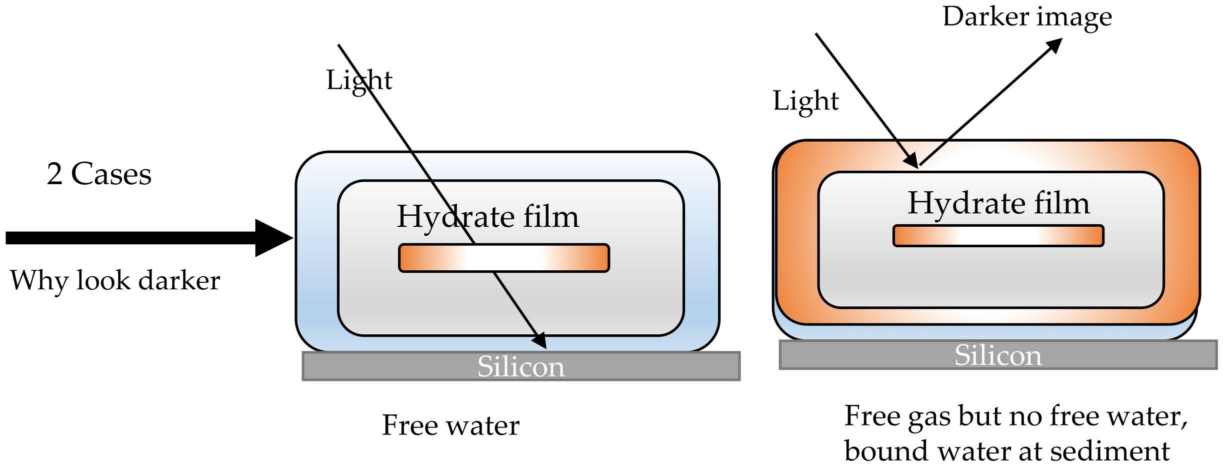


Hydrate arrangement

- **Observation**
- Hydrate can be formed
  - Methane saturated water crystallize
  - Gas pockets surrounded by water



Hydrate development at gas water interface for hydrate film

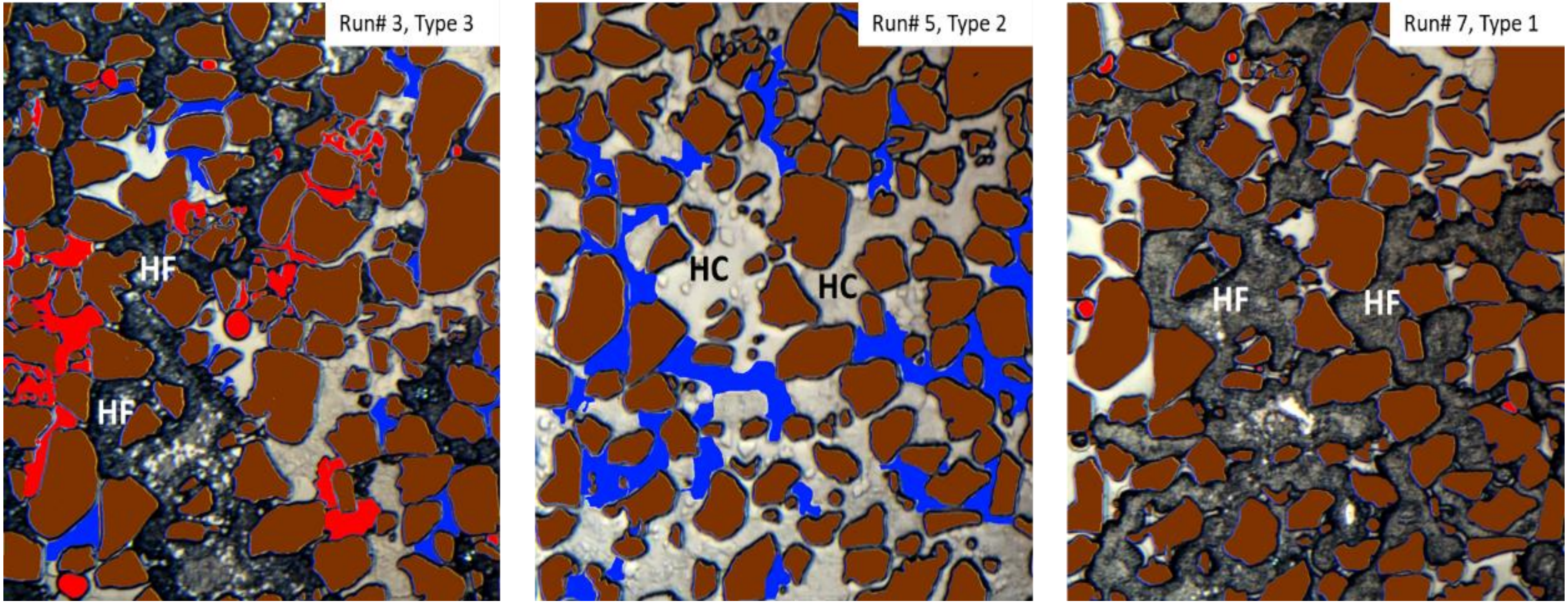


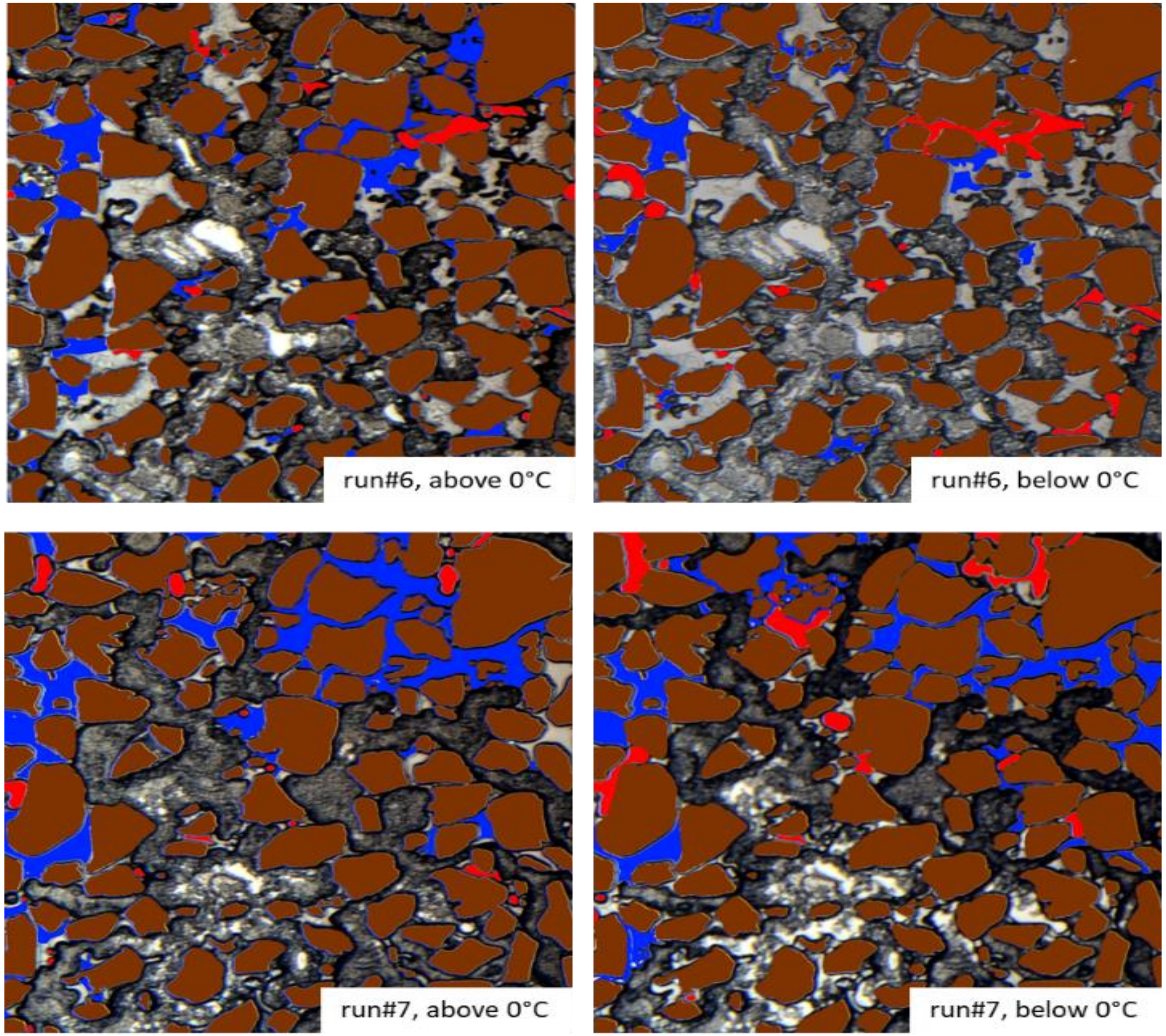
Take away & Known information

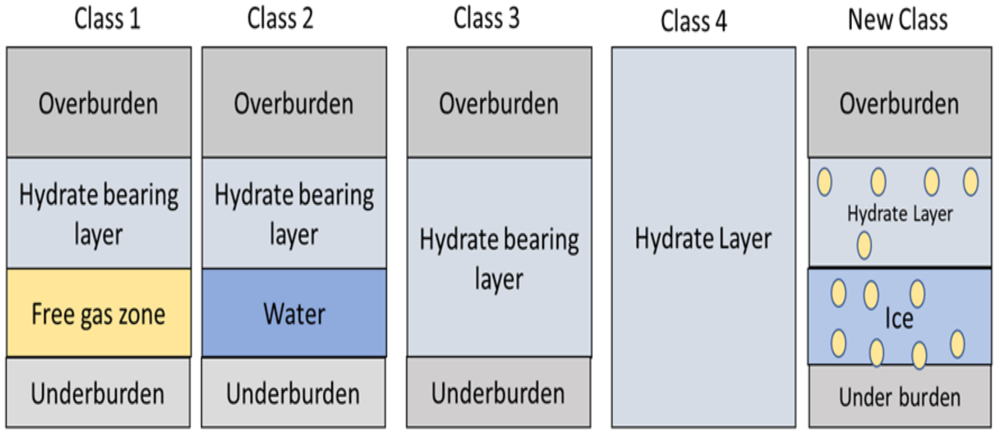
- Hydrate formed at gas-water interface are more porous in nature
- Hydrate film color is based on layer of gas around it, Higher the gas thickness above, darker is the hydrate shells
- Thickness of hydrate around gas is controlled by mass transfer/insufficient gas pressure
- Porous /non porous hydrate could be inferred from image analysis
- Isolated gas bubbles in small pores space converted into non porous hydrate
- Excess gas, hydrate film, if excess water, hydrate crystals.
- Initial water & gas availability control the hydrate redistribution & hydrate saturation
- Hydrate rearrangement is independent of driving force

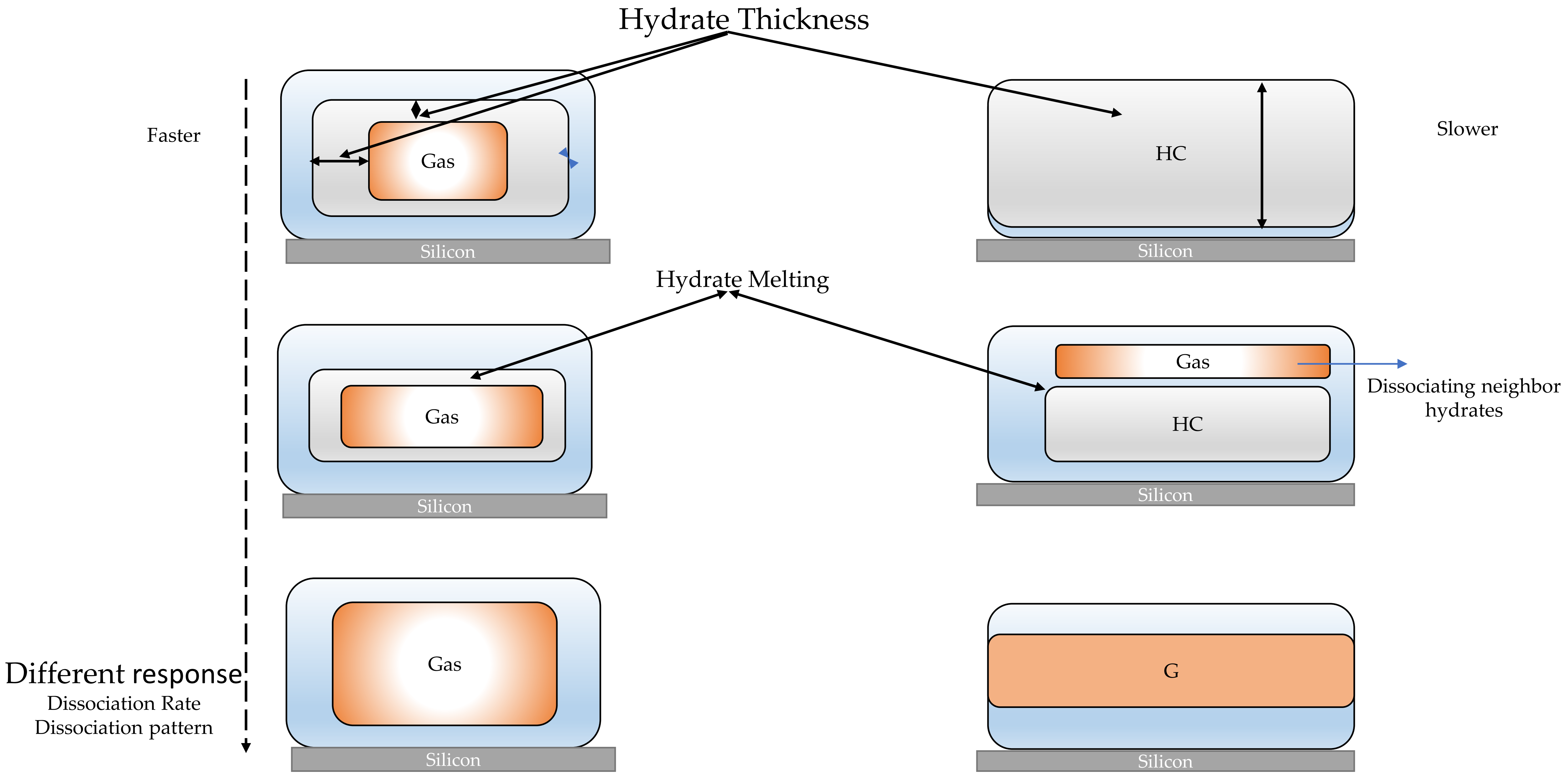
Experimental Plan

		Before formation		After formation				
run	T(°C)	Siw	Sig	SW	Sg	S_H	Hydrate Saturation	Morphology observed
run#1	0.9	10 %	90 %	1 %	6 %	93 %	High	HF, FG, FW
run#2	0.8	93 %	7 %	92 %	0 %	8 %	Low	HC, FW
run#3	1.0	61 %	39 %	3 %	8 %	88 %	High	HF, HC, FG
run#4	1.4	57 %	44 %	1 %	5 %	95 %	High	HF+FG+FW
run#5	1.5	56 %	44 %	16 %	0 %	84 %	High	HC+FW
run#6	1.7	50 %	50 %	7 %	2 %	91 %	High	HF+HC+FW+FG
run#7	1.9	53 %	47 %	12 %	1 %	87 %	High	HF+FW+FG
run#8	2.0	87 %	14 %	74 %	1 %	25 %	Low	HF+HC+FW+FG

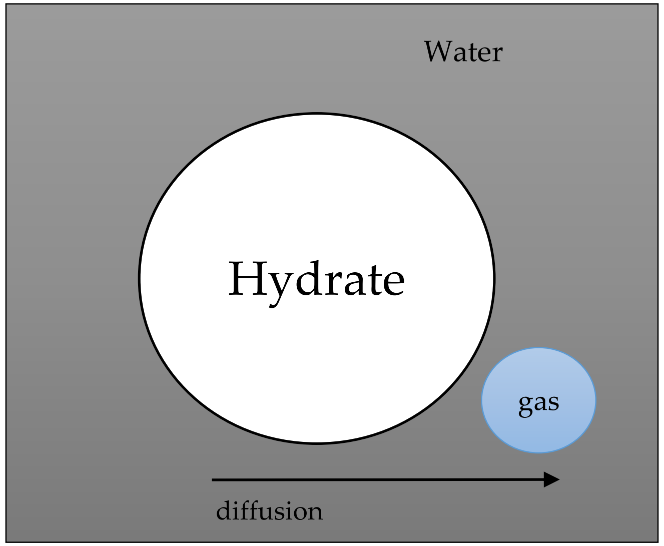

Run# 3, Type 3
HF
HF
Run# 5, Type 2
HC
HC
Run# 7, Type 1
HF
HF

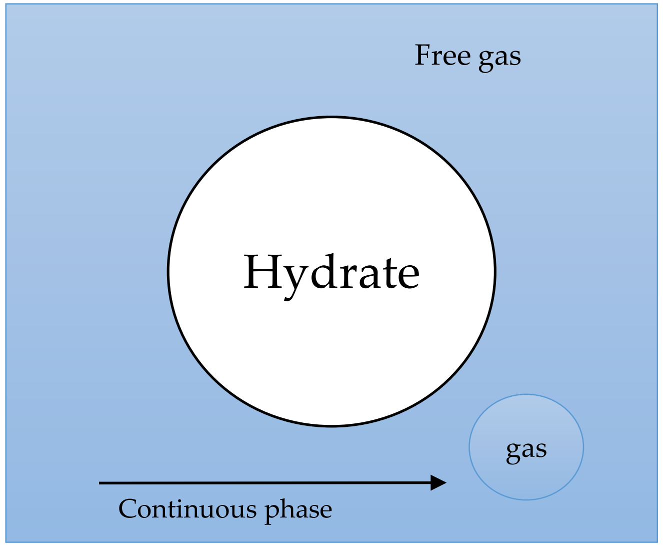

run#6, above 0°C
run#6, below 0°C
run#7, above 0°C
run#7, below 0°C


Class 1
Class 2
Class 3
Class 4
New Class
Overburden
Hydrate bearing layer
Free gas zone
Underburden
Overburden
Hydrate bearing layer
Water
Underburden
Overburden
Hydrate bearing layer
Underburden
Hydrate Layer
Overburden
Hydrate Layer
Ice
Under burden

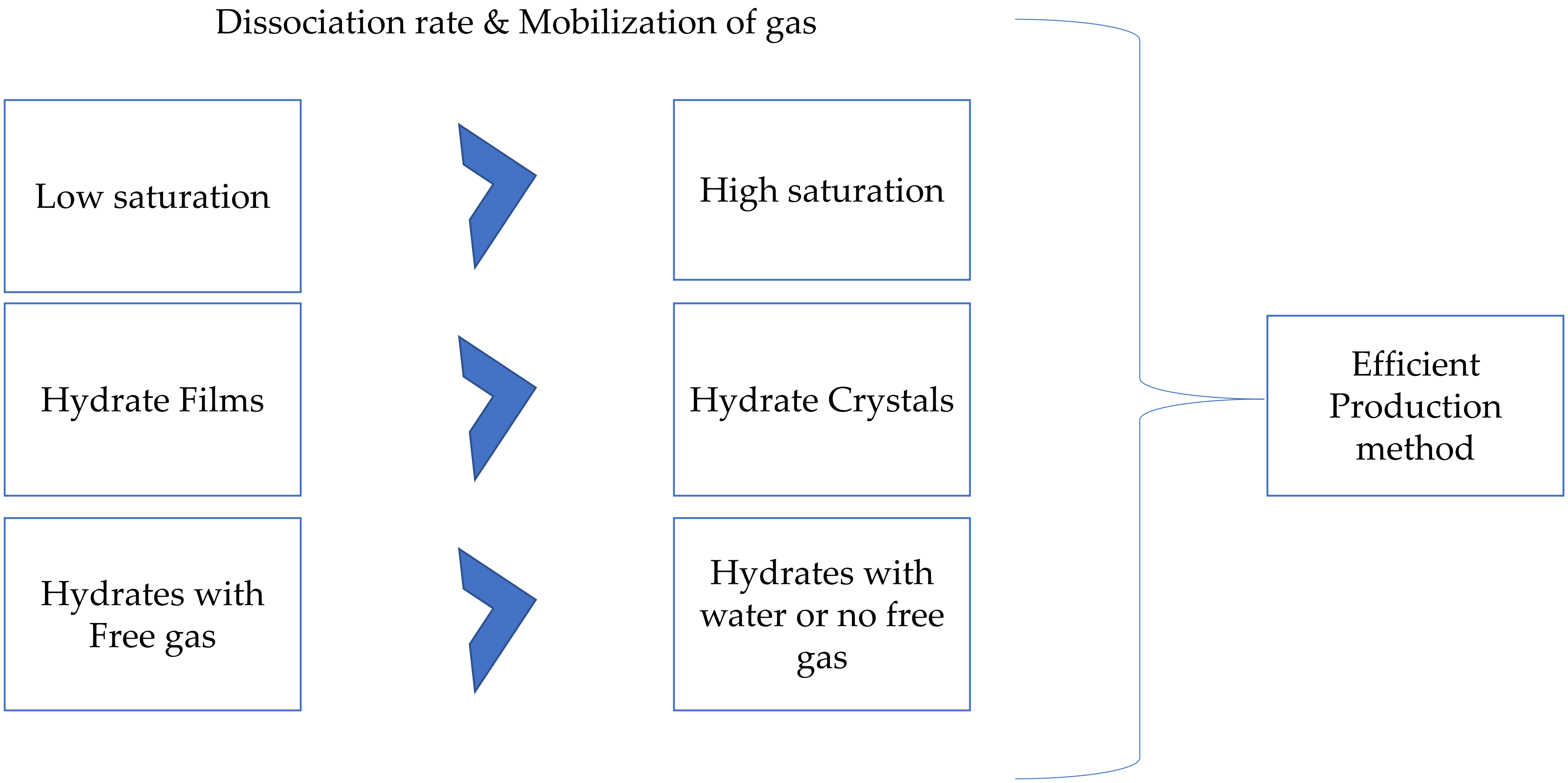

Hydrate Thickness
Faster
Gas
Silicon
HC
Silicon
Slower
Hydrate Melting
Gas
Silicon
Gas
HC
Silicon
Dissociating neighbor hydrates
Different response
Dissociation Rate
Dissociation pattern
Gas
Silicon
G
Silicon

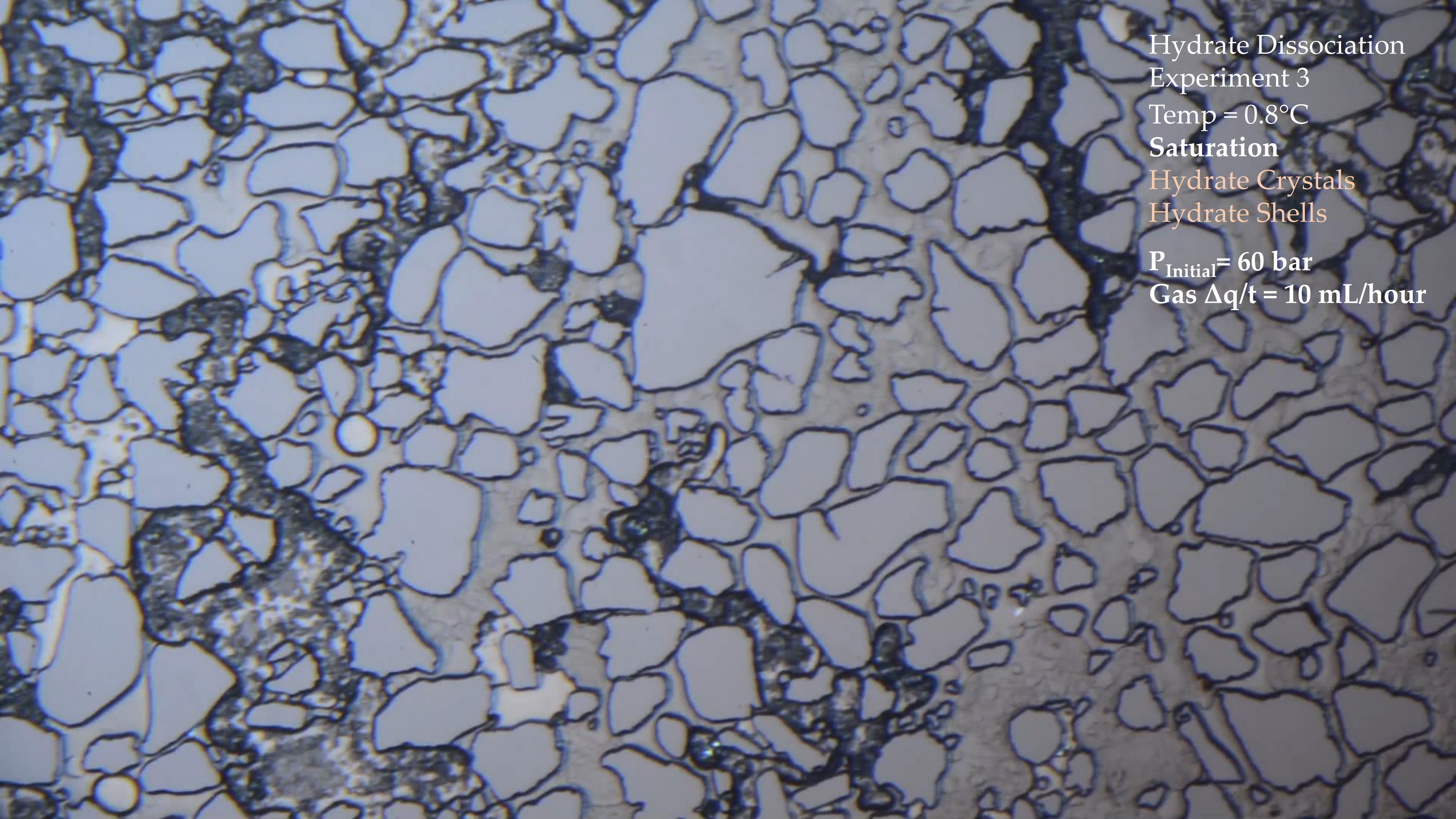
Free gas assisted dissociation





- **Take away**
- Free gas lead to accelerated hydrate dissociation by depressurization
  - Faster mass transport through continuous gas phase
- Hydrate Reservoirs with high hydrate saturation and no free gas
  - Depressurization not efficient method and combination with other methods are recommended.


Dissociation rate & Mobilization of gas
Low saturation
High saturation
Hydrate Films
Hydrate Crystals
Hydrates with Free gas
Hydrates with water or no free gas
Efficient Production method


Hydrate Dissociation
Experiment 3
Temp = 0.8°C
Saturation
Hydrate Crystals
Hydrate Shells
P_Initial = 60 bar
Gas Δq/t = 10 mL/hour

Run#	T(°C)	Sh	Hydrate pattern	ΔP				Δt (min)	Observations	Method
				Start (P_i)	P_d	ΔP	P_f			
run#1	0.9	93 %	HF+FG+FW	85	34,8	50,2	14	44.4	FD	Dep
run#2	0.8	8 %	HC+FW	76	20,4	55,6	14	38	FD	Dep
run#3	1.0	88 %	HF+HC+FG	60	18,3	41,7	14	50	FD	Dep
run#4	-0.5	93 %	HF+FG+FW	79	23,6	55,4	20	42	FD	Dep
run#5	-0.2	90 %	HC+FW	71		71	1,4	2905	SP, no FD	Dep plus temp
run#6	-2.8	93 %	HF+HC+FW+FG	80	18	62	5,6	2928	SP, no FD	Dep plus temp
run#7	-2.6	87 %	HF+FW+FG	55	15	40	14,3	75	FD	Dep
run#8	-2.6	24 %	HF+HC+FW+FG	80	13	67	5	3438	SP, no FD, RF	Dep plus temp

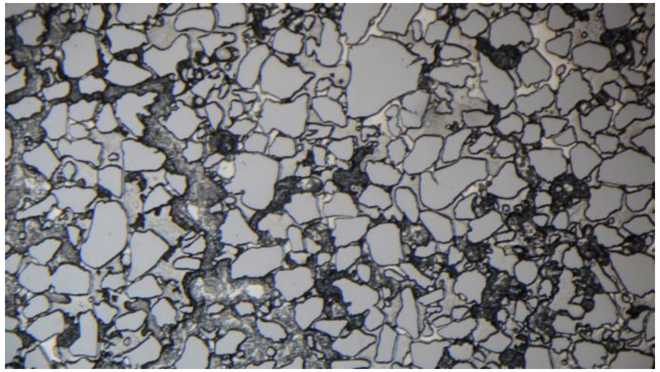
Effect of Temperature on Dissociation Rate

Take away 4

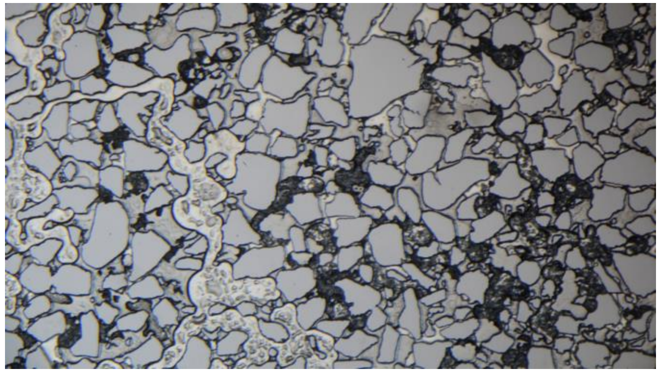
- No ice reformation during depressure
- Hydrate Crystals, depressurization, not sufficient

Exp -4

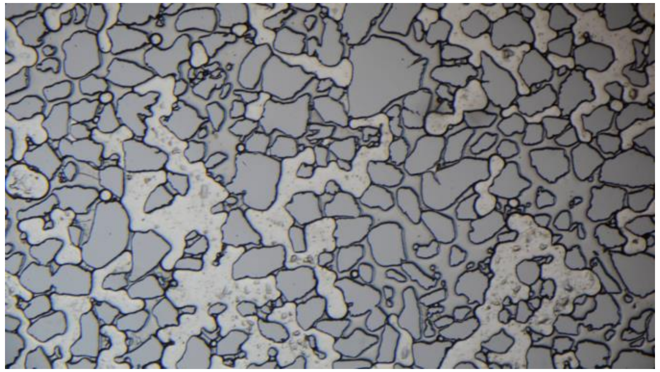
Hydrate shells + free gas + water **T=-0.5°C**



T=-0.5°C



T=-0.5°C

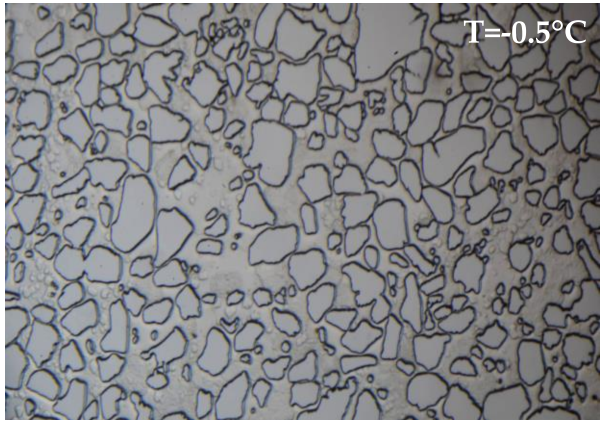


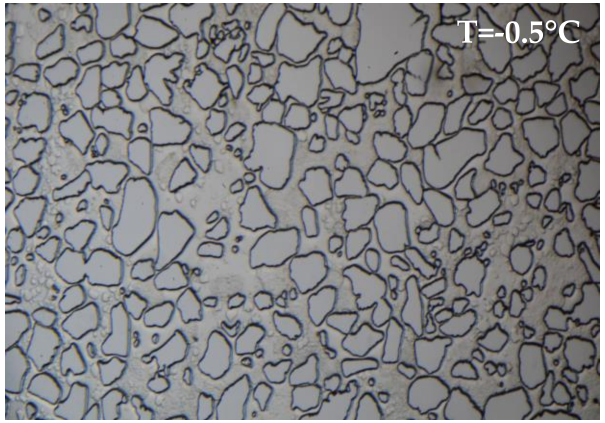
Hydrate Fully dissociated

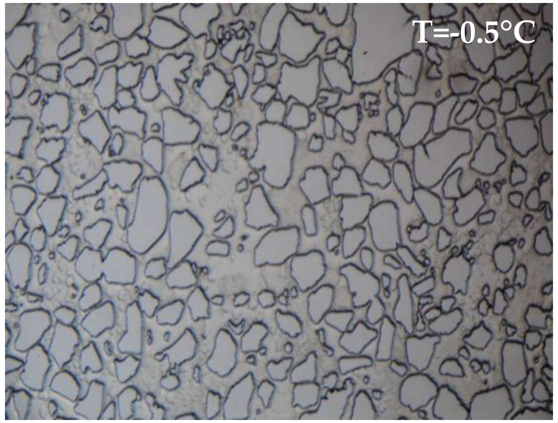
P= 71 bar Δt= 33 min → P= 22.8 bar Δt= 9 min → P= 20 bar

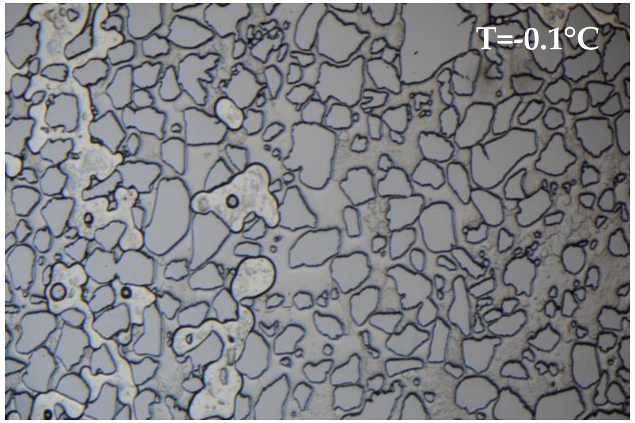
Exp -5

Hydrate Crystals





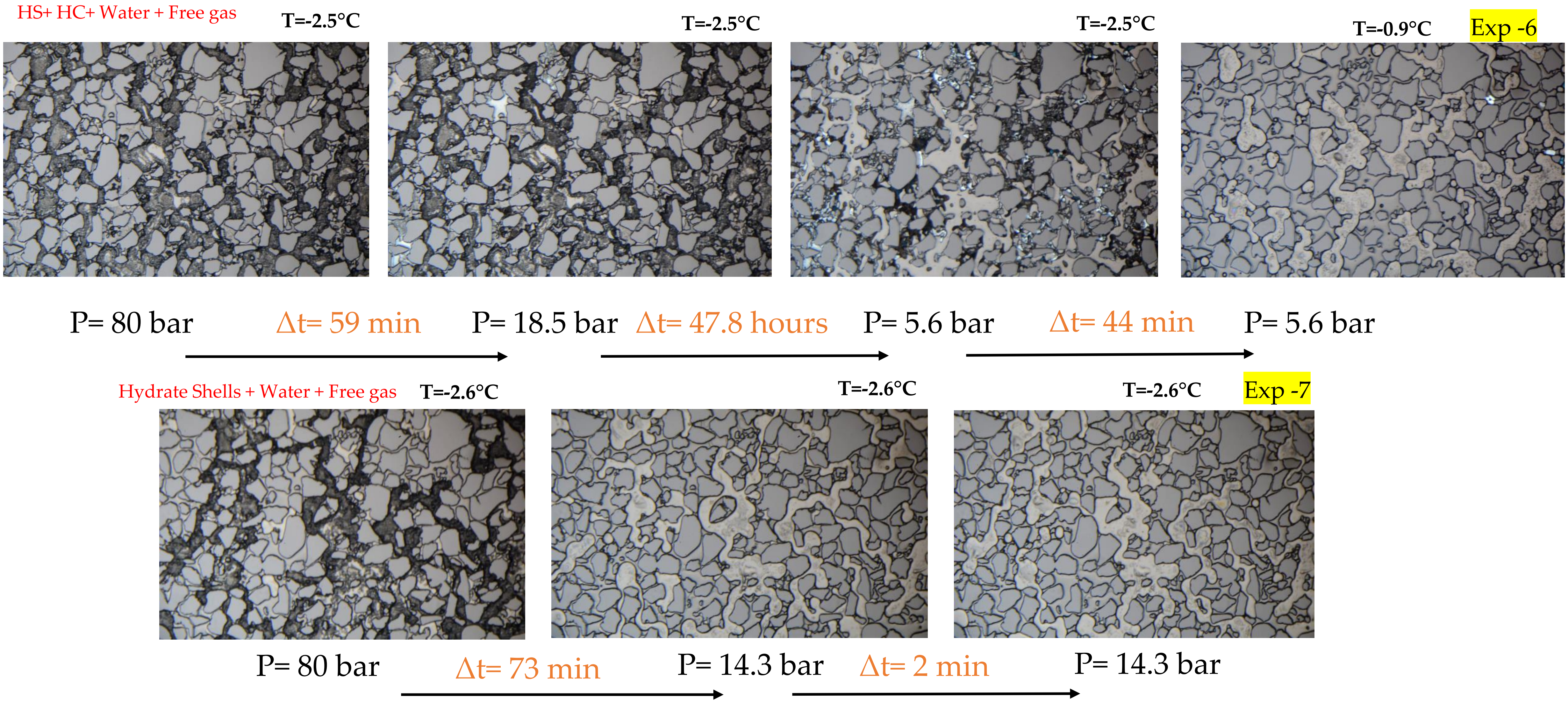




P= 71 bar Δt= 85 min → P= 12 bar Δt= 47.8 hours → P= 1.4 bar Δt= 6.15 min → P= 1.4 bar

No Dissociation during depressurization

Temperature increase

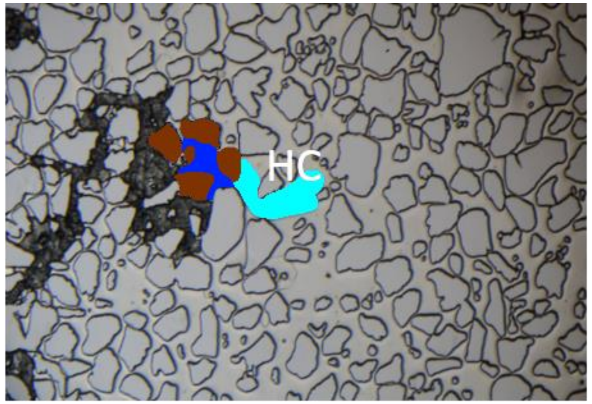


HS+ HC+ Water + Free gas
T=-2.5°C
T=-2.5°C
T=-2.5°C
T=-0.9°C
Exp -6
P= 80 bar
Δt= 59 min
P= 18.5 bar
Δt= 47.8 hours
P= 5.6 bar
Δt= 44 min
P= 5.6 bar
Hydrate Shells + Water + Free gas
T=-2.6°C
T=-2.6°C
T=-2.6°C
Exp -7
P= 80 bar
Δt= 73 min
P= 14.3 bar
Δt= 2 min
P= 14.3 bar

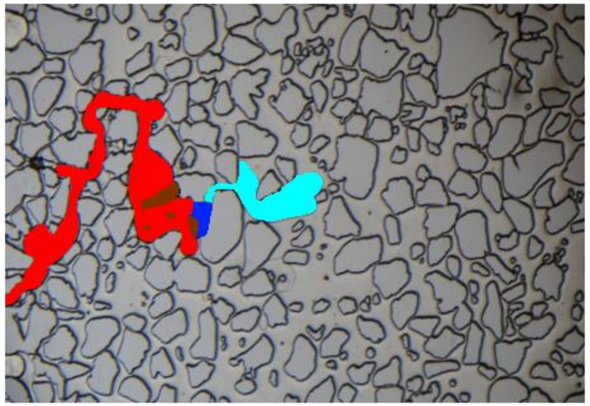
HS+ HC+ Water + Free gas

Exp -8

T=-2.9°C

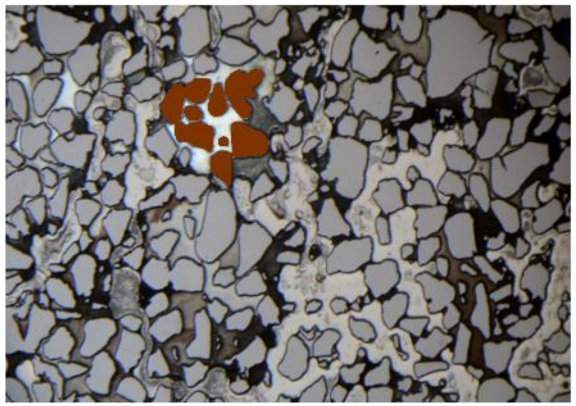


T=-2.9°C

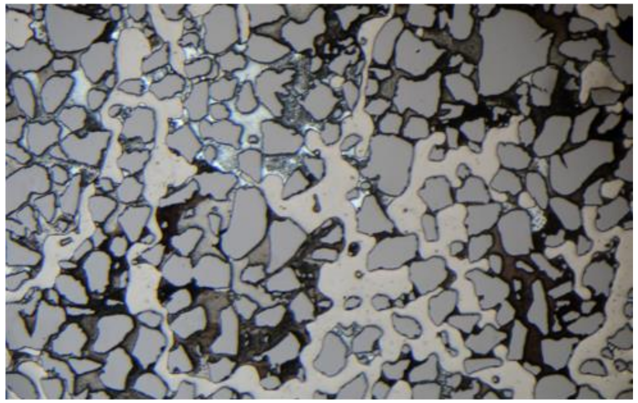


Reformation

T=-2.9°C

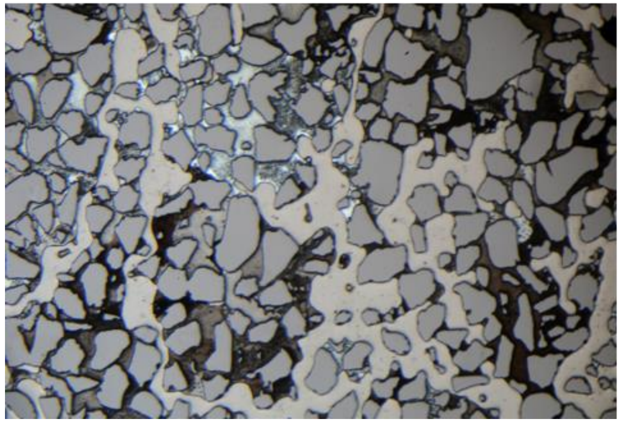


T=-2.9°C



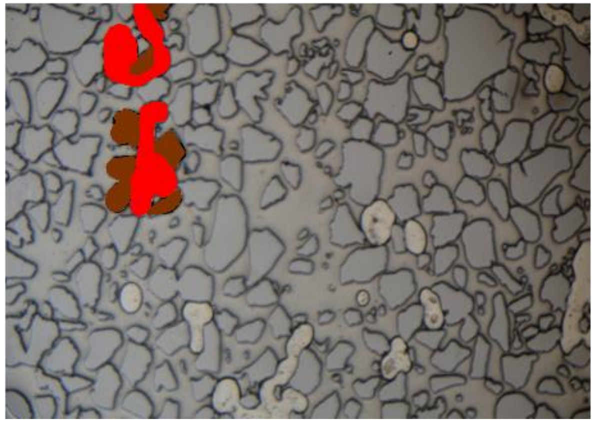
P= 80 bar   Δt= 83min   Δt= 7 min   P= 13 bar   Δt= 55.8 hours   P= 13 bar

T=-2.7°C

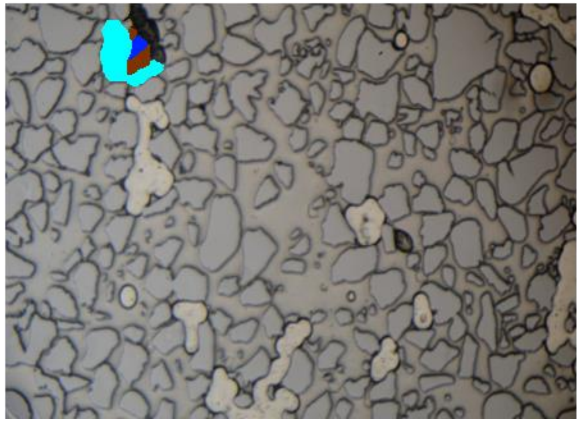


Reformation

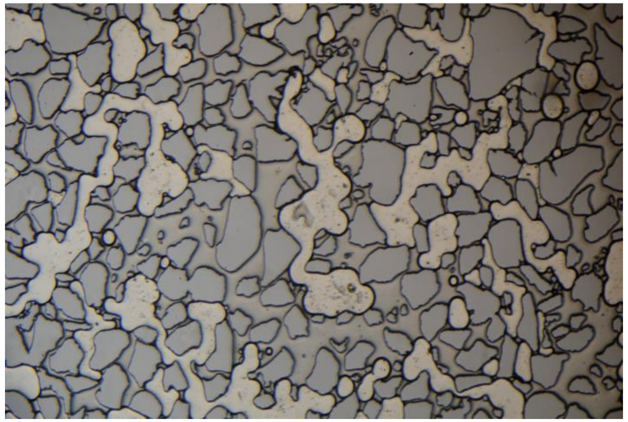
T=0.4°C



T=0.8°C



T=1.1°C



P= 5 bar

Δt= 48 mins →   Δt= 10 mins →   Δt= 28 mins →

P= 5 bar

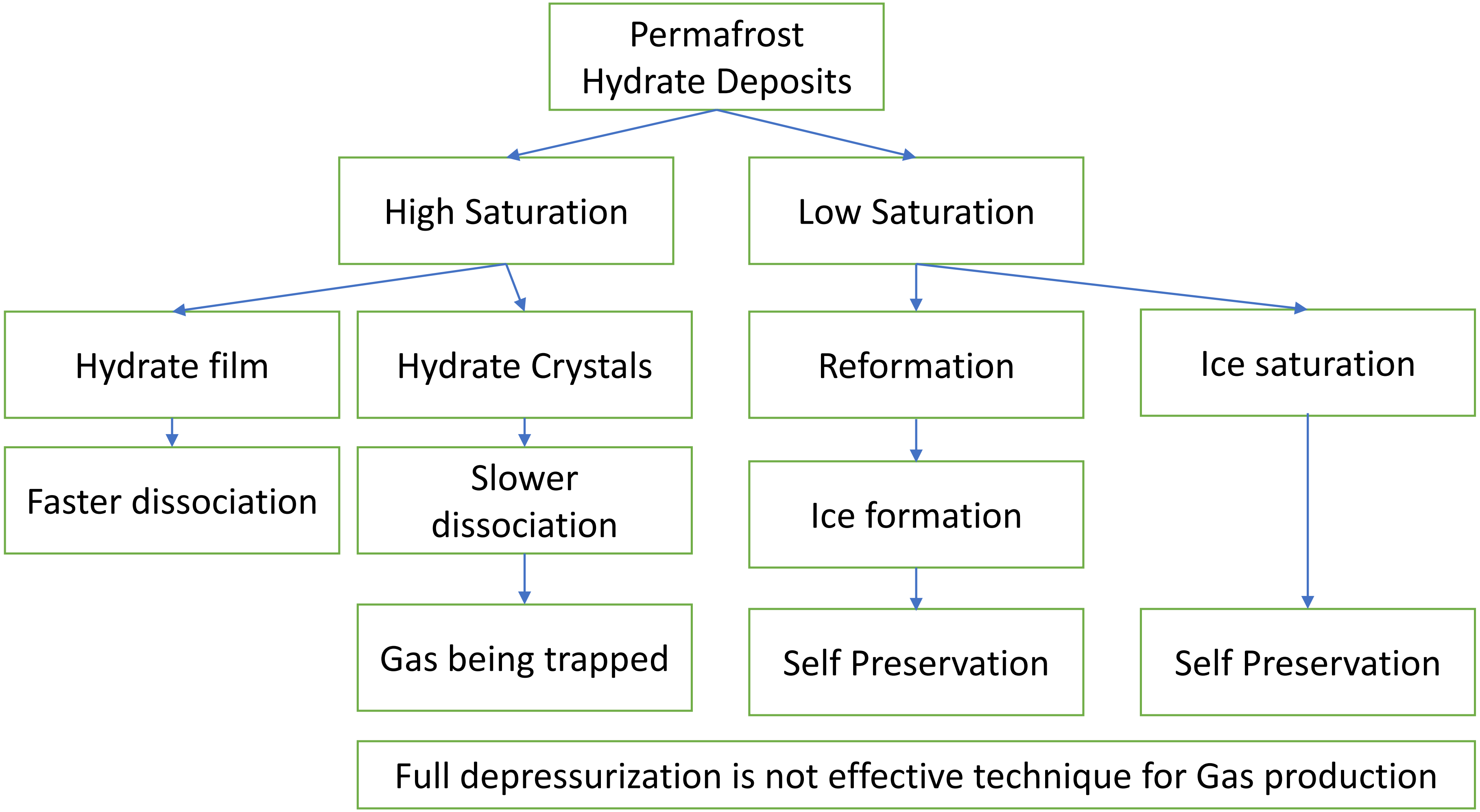
Take away 5

- Ice reformation is observed during depressurization
- High Self preservation
- Temp stimulation took longer time due to ic melting first

Self preservation & reformation

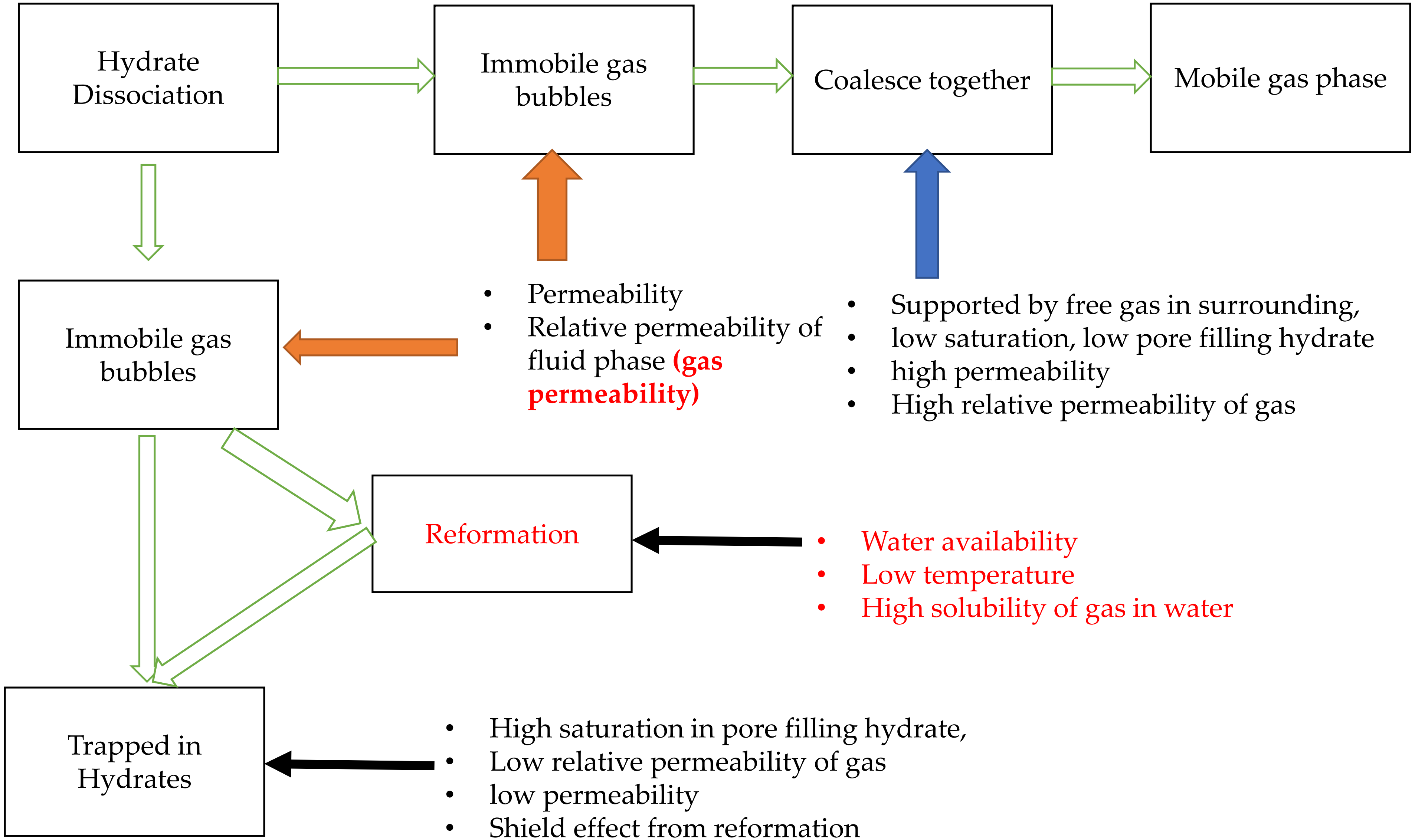
- Hydrate film show weaker self preservation tendency (no self preservation) compare to hydrate crystals
- Reformed hydrates are in the form of hydrate films, not crystalline in nature, hence porous and less stable hydrates.
- Excess water leads to higher risk of reformation/ice formation
- Risk of reformation higher at negative temperature
- Permafrost gas deposits could either coexists with supercooled water or ice along with isolated gas pockets.

Dissociation behavior in Permafrost at high negative temperature (-2^{o}C or below)



Pore filling hydrates

- Self Preservation due to ice/supercooled water
- Hydrate crystals, Hydrate films



Final Conclusion

- Micromodel based pore level study provide insights about kinetics of hydrate formation and dissociation
- Initial information such as hydrate saturation, free gas presence is critical for selection of efficient production technique.
- Subzero temperature, make dissociation slower due to self preservation tendency shown by hydrate as well as increase risk of ice and hydrate reformation. Thus, depressurization is not efficient method for gas production in permafrost hydrate reservoirs.

Acknowledgement

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Center for Energy Resources Engineering
CERE

DTU Chemical Engineering
Department of Chemical and Biochemical Engineering


Thank you!