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A new approach of experimental and modelling study of mixed gas hydrates under non-equilibrium conditions

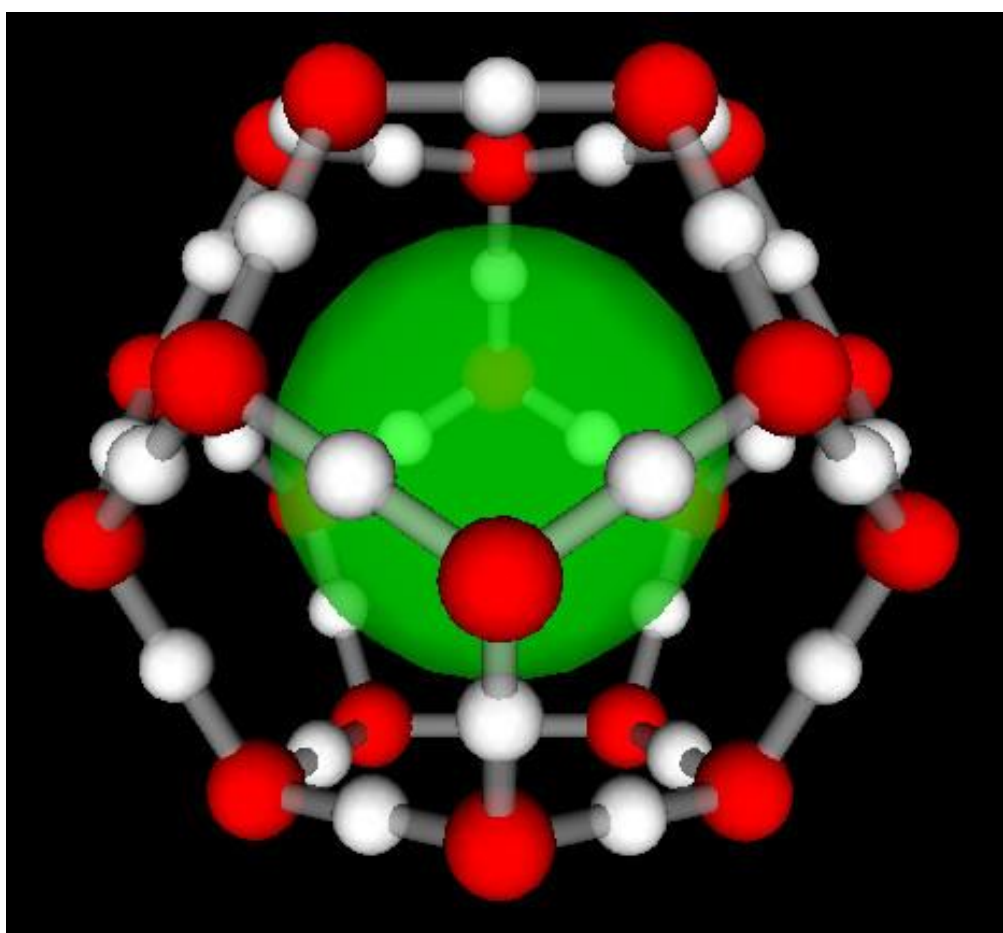


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Introduction

Gas hydrates are crystalline solids composed of water and gas. The gas molecules (guests) are trapped in water cavities (host) that are composed of hydrogen-bonded water molecules.

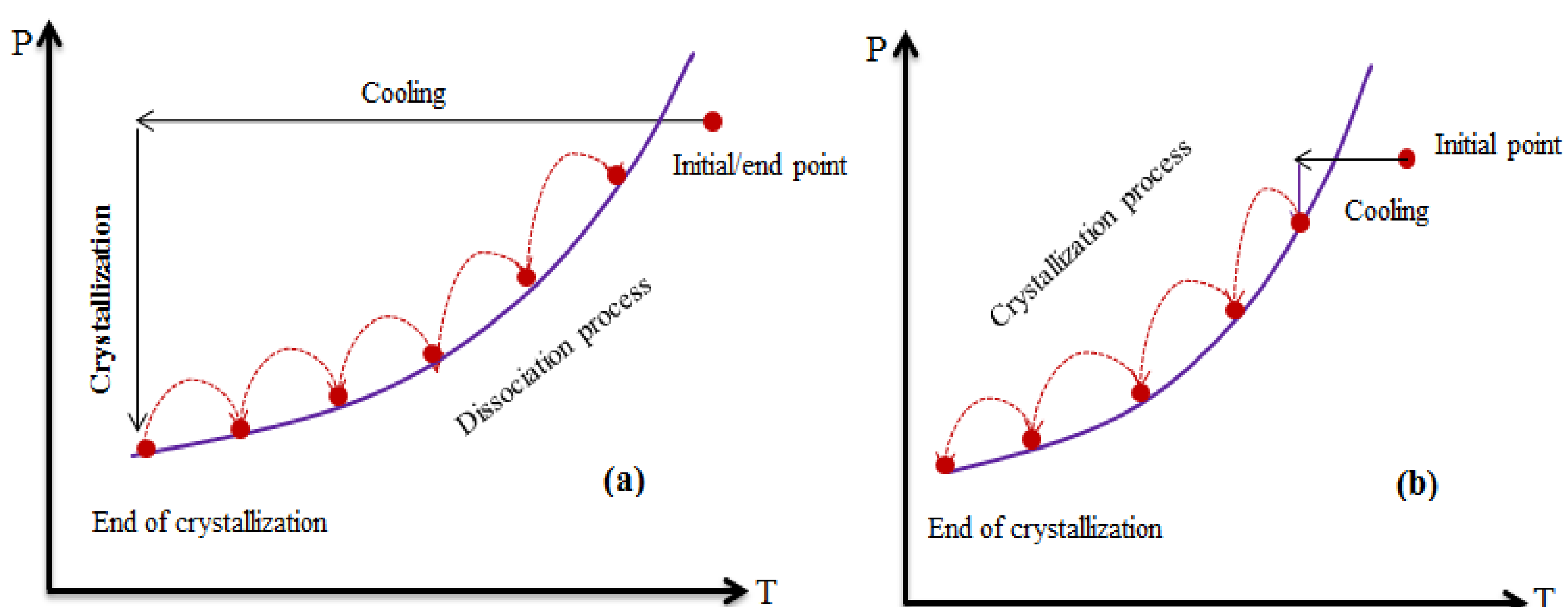


Gas hydrates are a crucial issue in many fields, from oil & gas industry, to carbon capture and storage, air conditioning, or even planetary science. They also have an enormous potential as an energy resource



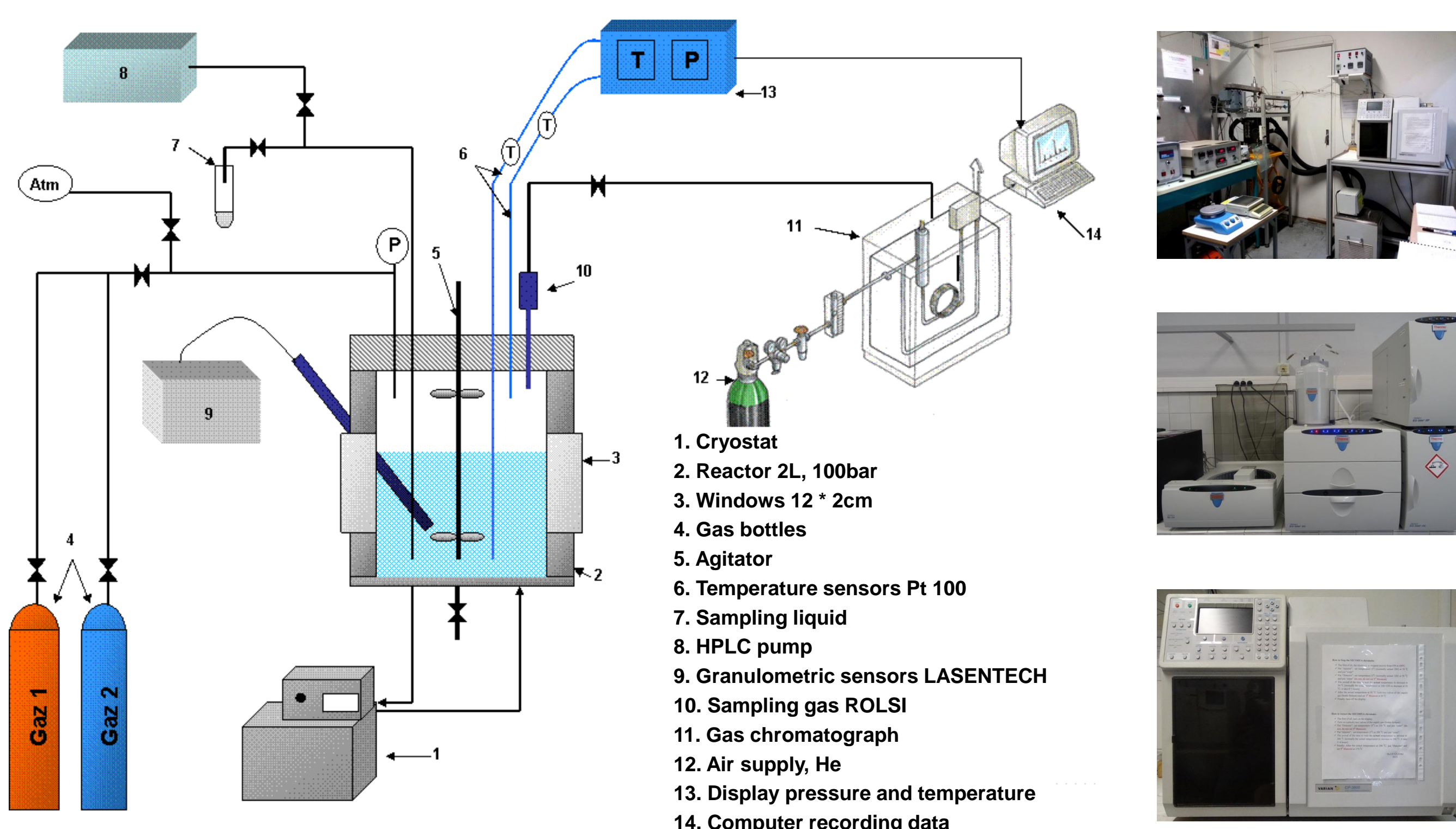
Objectives

➤ Effects of crystallization rate



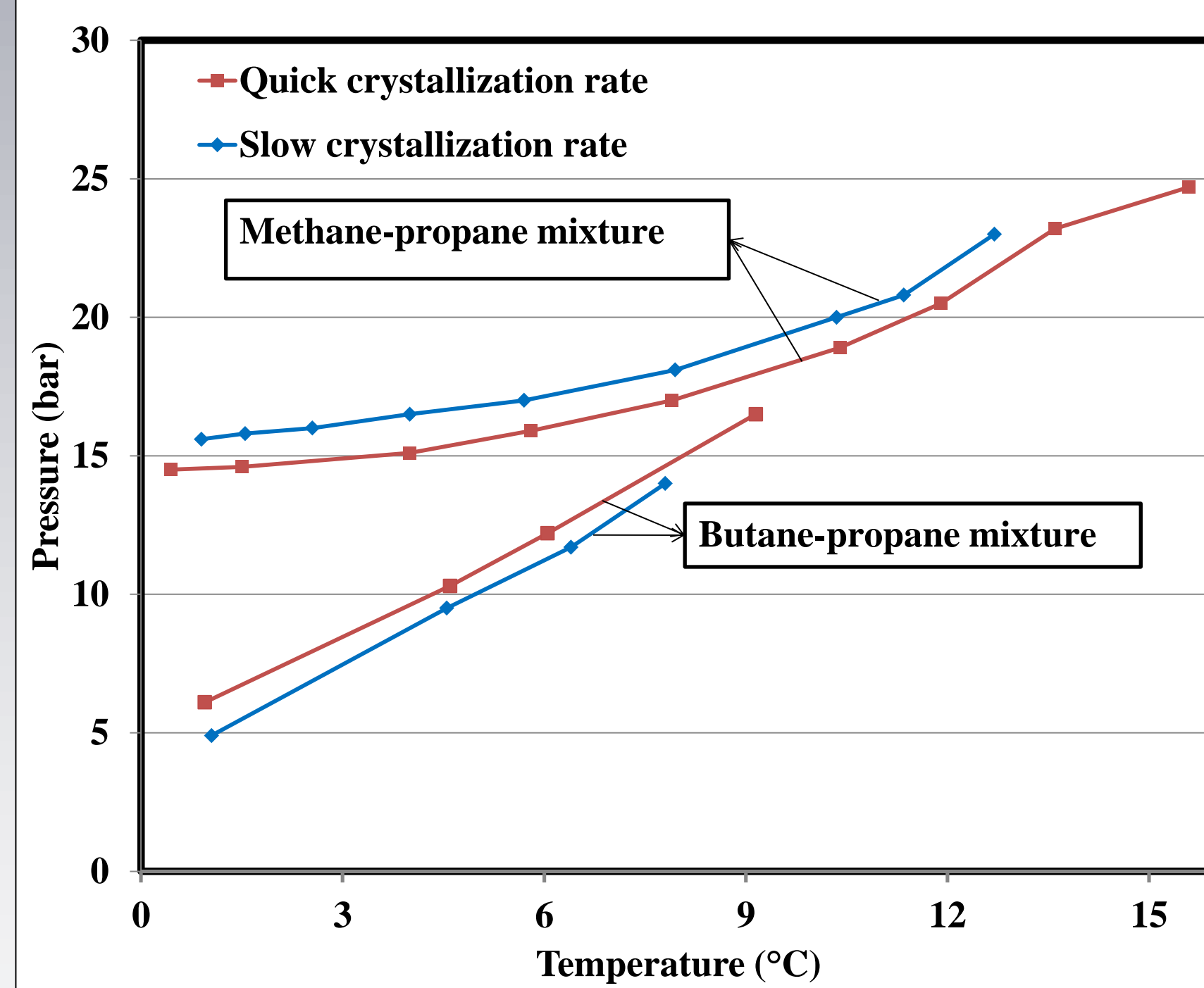
- Studying the thermodynamics of mixed clathrate hydrates
- Determination of the gas composition in hydrate phase
- Volume of clathrate hydrate and water conversion
- Developing a reliable thermodynamic model based on classical van der Waals and Platteuw model by implementing Kihara parameters.

Experimental section



Results

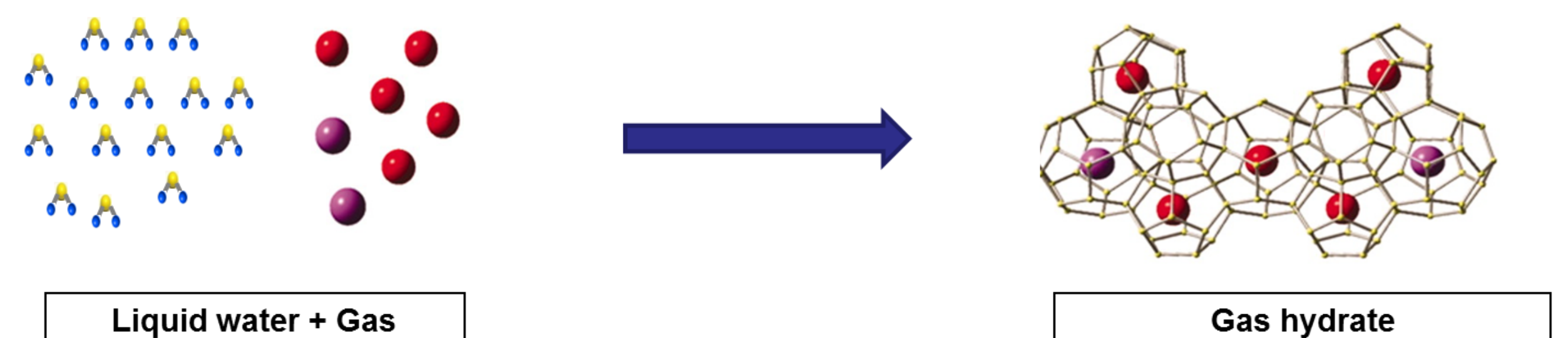
Two different gas mixture including propane were studied. For each mixture, we performed two different experiments based on two different crystallization rates at the same initial conditions.



The results of two different crystallization rates show that:

- The final pressure at final state is slightly different.
- Water conversion and hydrate volume at quick crystallization is more than slow crystallization.
- The composition of heavier hydrocarbon (here propane) at slow crystallization is more than quick crystallization

A thermodynamic model, implementing classic van der Waals and Platteuw model, was used. The Kihara parameters for methane were taken from the previous works of our team and the Kihara parameters for propane have been investigated in this work.



$$\mu_w^L$$

=

$$\mu_w^H$$

$$\Delta\mu_w^{\beta-L} = \mu_w^L - \mu_w^\beta$$

=

$$\Delta\mu_w^{\beta-H} = \mu_w^H - \mu_w^\beta$$

Classical thermodynamic

Statistical thermodynamic

Parameters from Handa and Tse

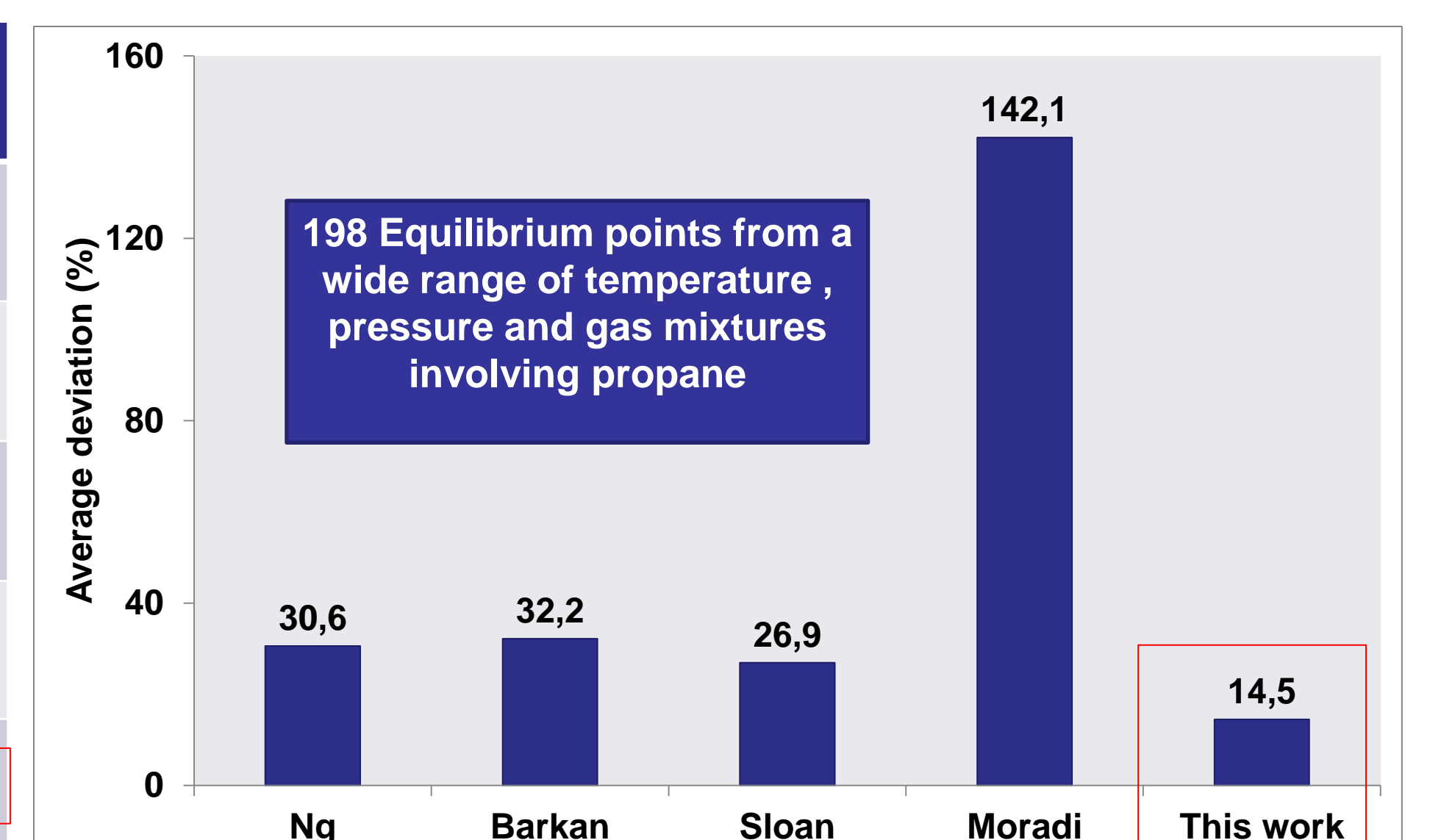
Kihara parameters

$$\Delta\mu_w^{\beta-L} = T \frac{\Delta\mu_w^{\beta-L}}{T} \Big|_{T^0, P^0} - T \int_{T^0}^T \frac{\Delta\mu_w^{\beta-L}}{T^2} dT + \int_{P^0}^P \Delta\mu_w^{\beta-L} dP - RT \ln a_w^L \Big|_{T, P}$$

$$w(r) = 2z\epsilon \left[\frac{\sigma^{12}}{R^{11}r} \left(\delta^{10} + \frac{a}{R} \delta^{11} \right) - \frac{\sigma^6}{R^5 r} \left(\delta^4 + \frac{a}{R} \delta^5 \right) \right]$$

Based on our results of slow crystallization and also some equilibrium data from literature, a new set of Kihara parameters for propane was obtained. The simulation based on the new Kihara parameters was compared with the other Kihara parameters in the open literature.

Reference	ϵ/K	σ	a
(Sloan, 1998)	203.31	3.3093	0.6502
(Ng and Robinson, 1977)	213.58	3.2296	0.6700
(Barkan and Sheinin, 1993)	194.55	3.3144	0.8340
(Moradi and Khosravani, 2013)	493.70	4.5190	0.6502
This work	195.00	3.3400	0.6502



Conclusions

- The equilibrium pressure for a given temperature is slightly different for different crystallization rates.
- The Volume of hydrate and also water conversion in quick crystallization process is larger than slow crystallization rate.
- Moreover, in a hydrocarbon mixture at slow crystallization rate, enclathration of heavier hydrocarbon is more important.
- A New set of Kihara parameters of propane was obtained and it has a good accordance to predict the hydrate equilibrium pressure for a wide range of temperature from literature.



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