

Fluid immiscibility

at metamorphic conditions

experimental evidence from fluid inclusions

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session

Advanced Structural and Geochemical Characterization of Geomaterials

“major earth science question”:

fluid immiscibility at crustal geological conditions

source of ore mineralizations (hydrothermal, magmatic)?

transport of elements in liquid or vapour phase?

fluid flow properties?

“cutting-edge analytical approach”:

entrapment of two unmixed fluids at experimental conditions in synthetic fluid inclusions

„Immiscibility in the fluid phase could be responsible for many spectacular petrological and geochemical phenomena“

Pichavant et al. (1982)

“There is no doubt that fluid immiscibility plays a significant petrologic role in metamorphic processes.
“The evidence obtained from the fluid inclusion record in rocks is overwhelming.”

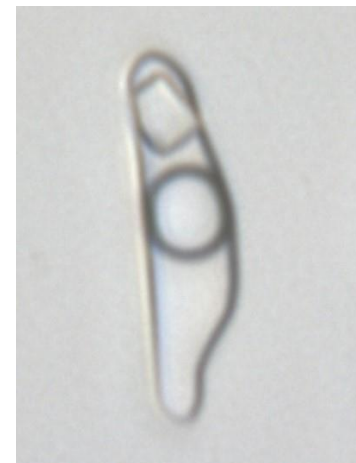
Heinrich (2007)

also known as:

- fluid immiscibility*
- chemical immiscibility*
- boiling*
- condensation*
- effervescence*
- stable mechanical mixture*



Fluid Inclusions in crystals are the only direct evidence of the presence and activity of fluids in rock in the geological past



can two distinct coexisting fluid phases be trapped and preserved in fluid inclusions?
in single crystals?

„Fluid immiscibility in natural processes: use and misuse of fluid inclusion data“

Pichavant et al. (1982)

Ramboz et al., (1982)

Chemical Geology, vol. 37

fluid immiscibility

state of the art

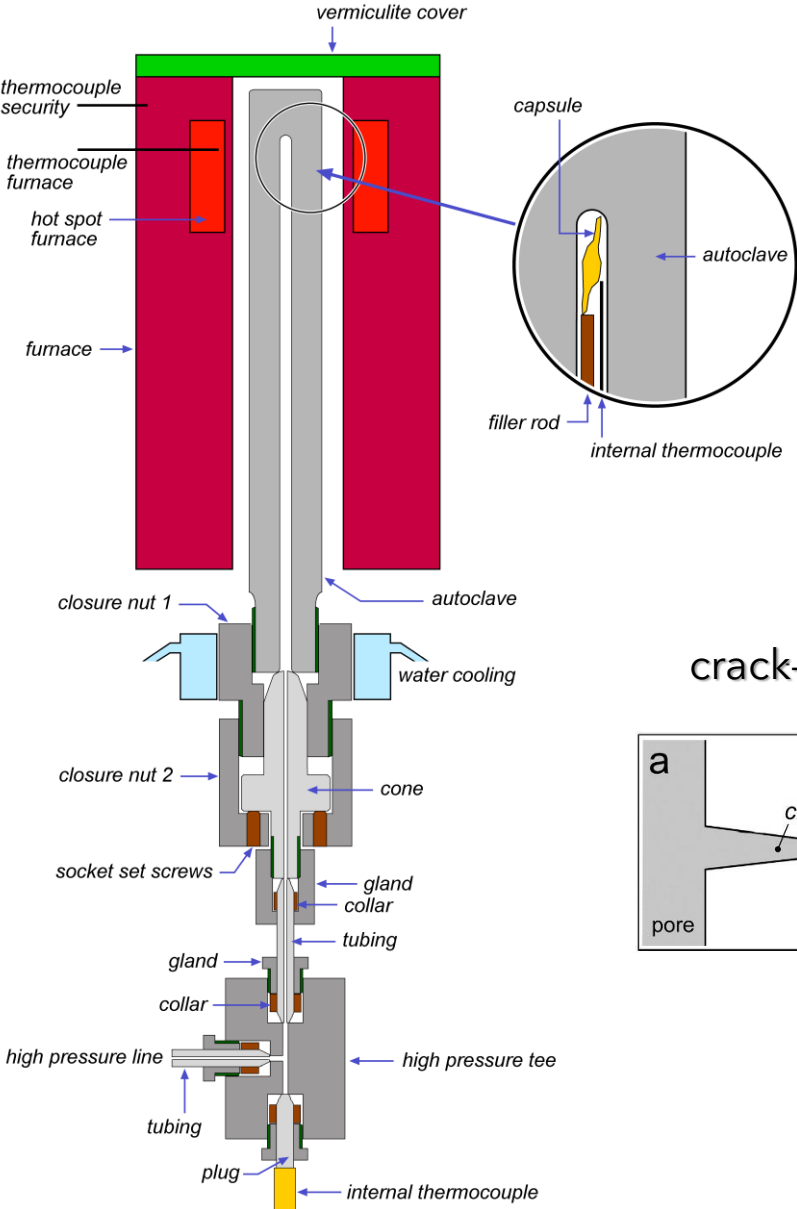
theory



data, model

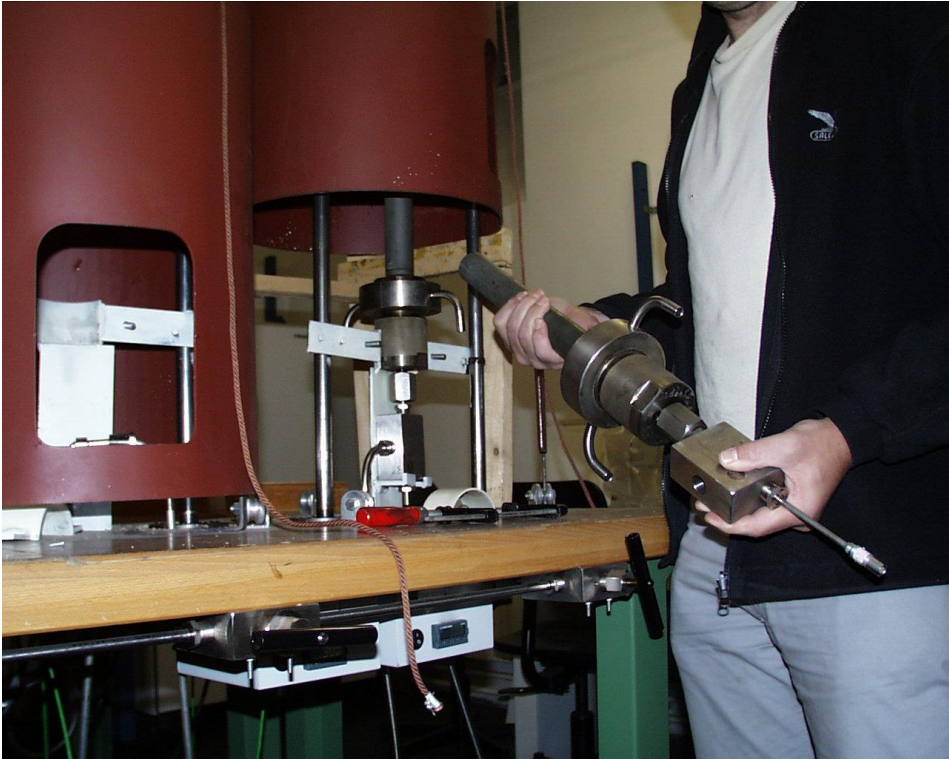
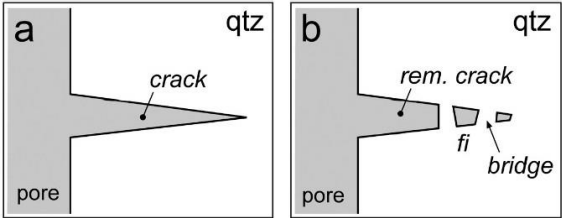


Experimental Method

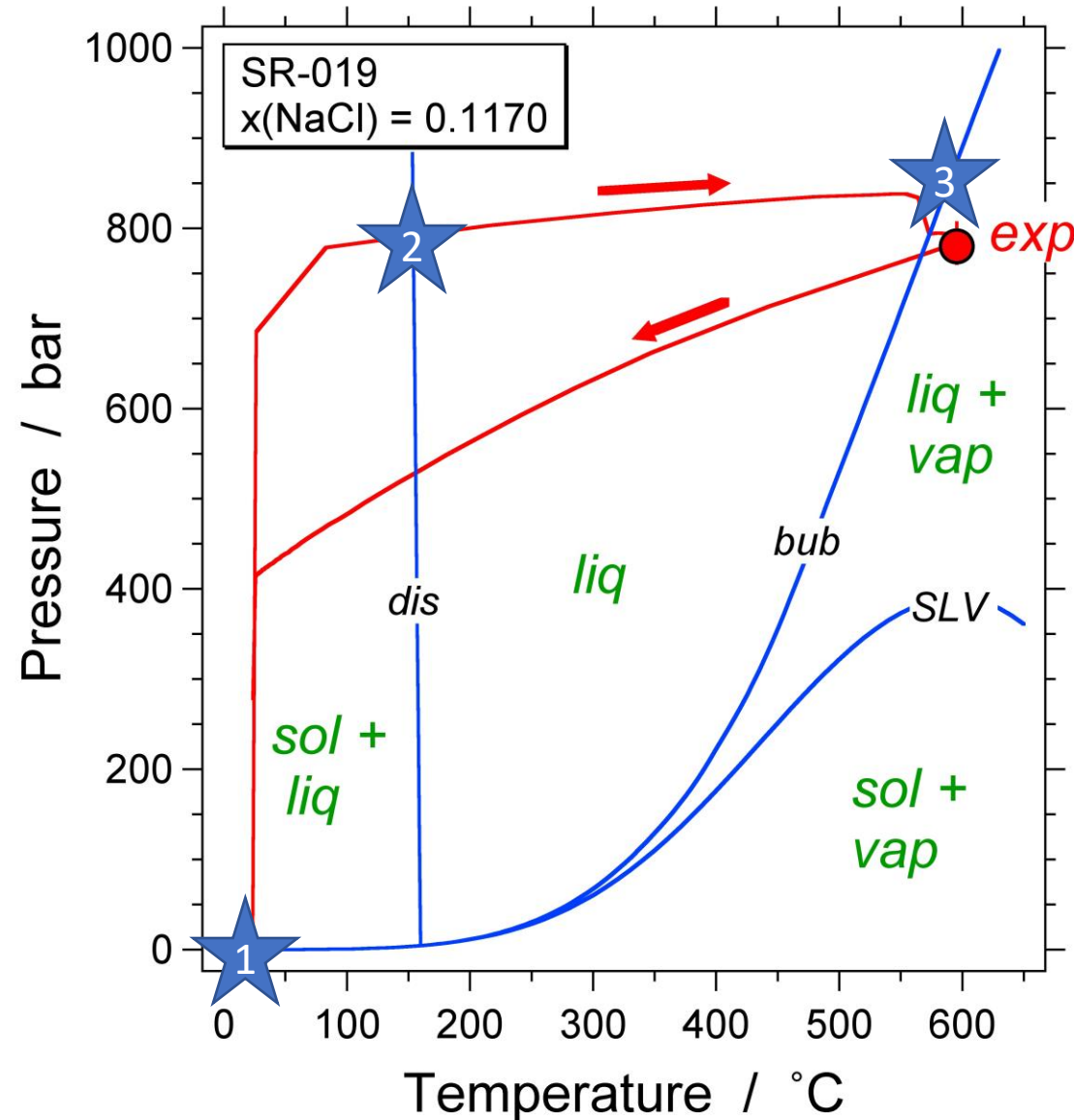


Synthetic Fluid Inclusions

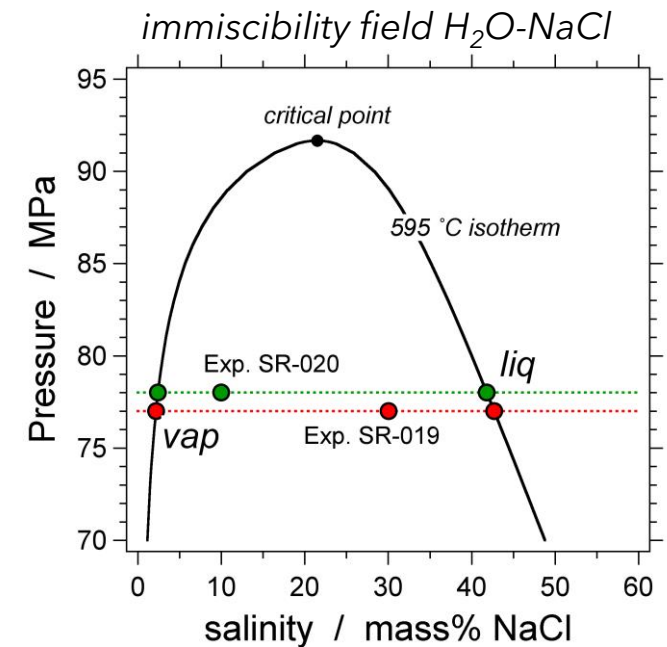
crack-healing in quartz



Immiscibility in the H_2O - NaCl fluid system



1. loading capsule at room temperature with pure H_2O and salt crystals
2. homogenization of salt- H_2O solution in capsule
3. fluid unmixing in capsule (liquid - vapour)



H₂O - NaCl

Synthetic fluid inclusions

Experimental conditions: 595.5 ± 0.3 °C

78.0 ± 0.2 MPa (SR-020)

77.0 ± 0.3 MPa (SR-019)

expected results:

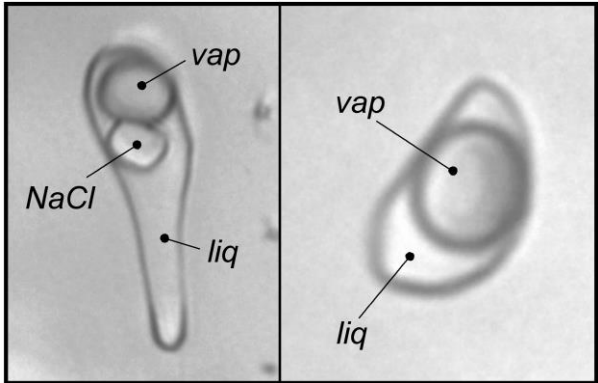
experiment	loaded fluid	immiscibility at experimental conditions					
		liquid-rich fluid			vapour-rich fluid		
	NaCl mass%	NaCl mass%	V _m cm ³ /mol	vol.%	NaCl mass%	V _m cm ³ /mol	vol.%
SR-019	30.07	41.40	29.34	47.0	2.41	58.51	53.0
SR-020	10.00	40.42	29.44	8.3	2.71	57.10	91.7

calculated with AqSo_NaCl (Bakker, 2019 and 2020)

thermodynamic model H₂O-NaCl after Driesner and Heinrich (2007) and Driesner (2007)

results optical microscopy: **two types of synthetic fluid inclusions per experiment**

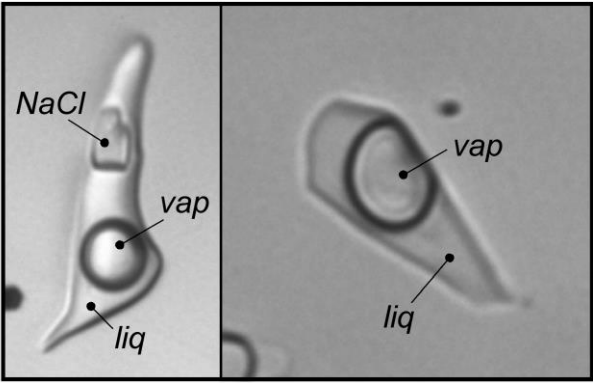
SR-019



abundant

rare

SR-020

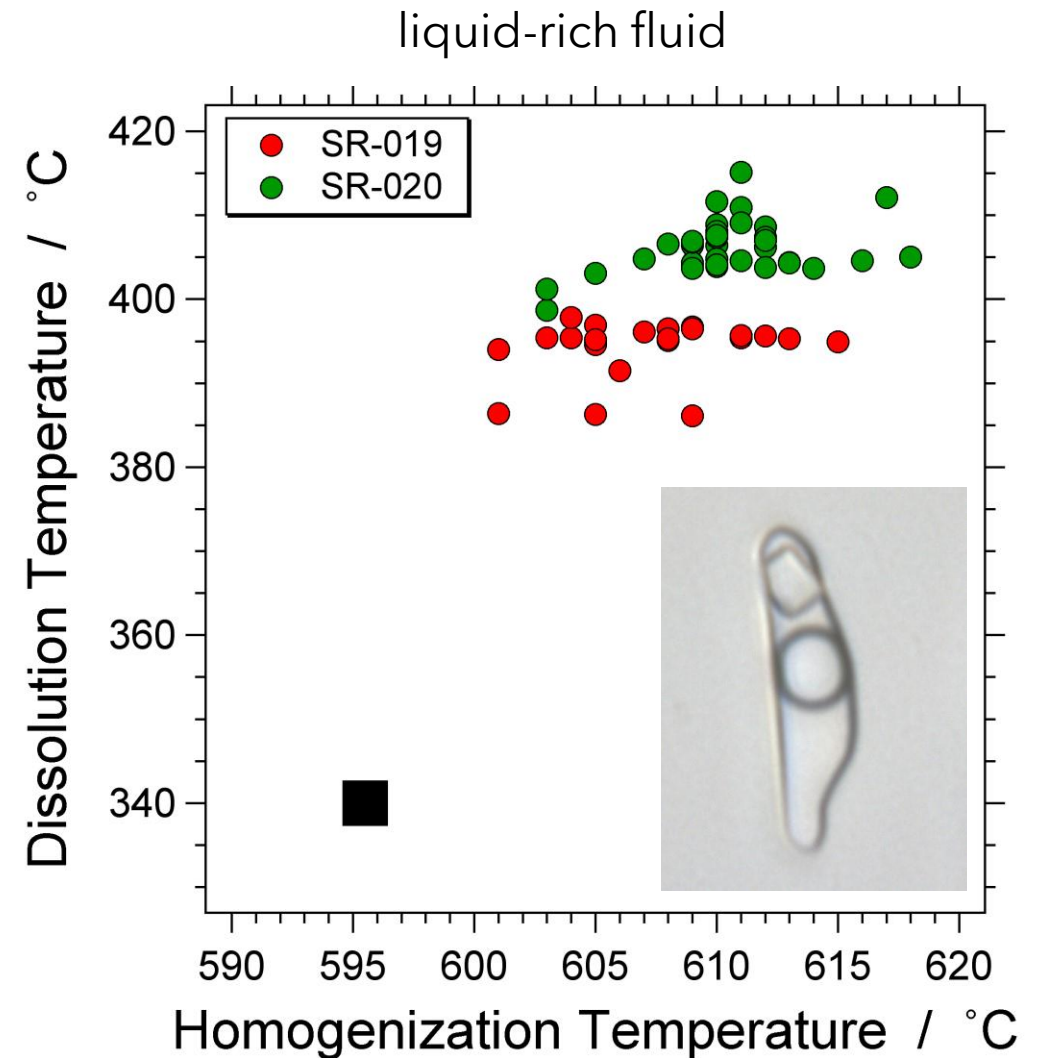
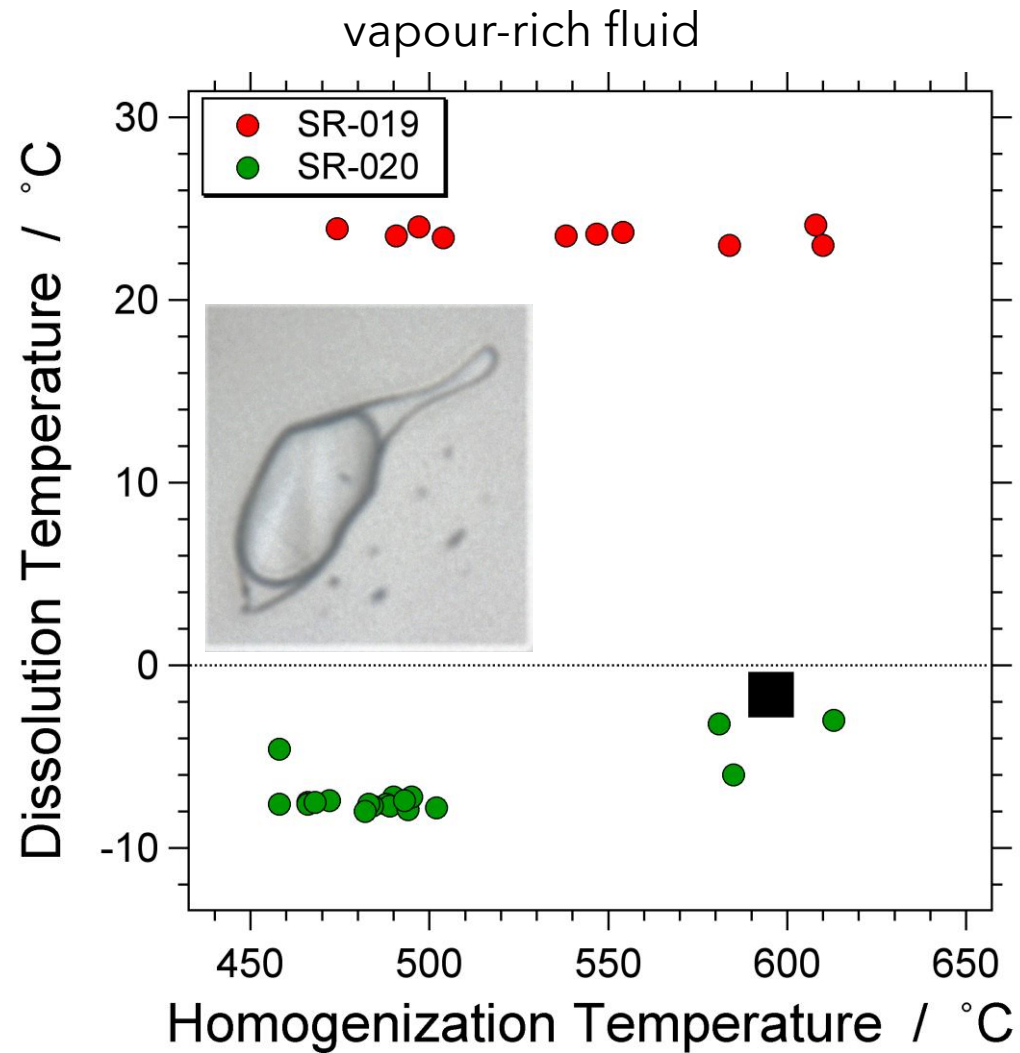


abundant

few

are these fluids the unmixed end-members?

results microthermometry:



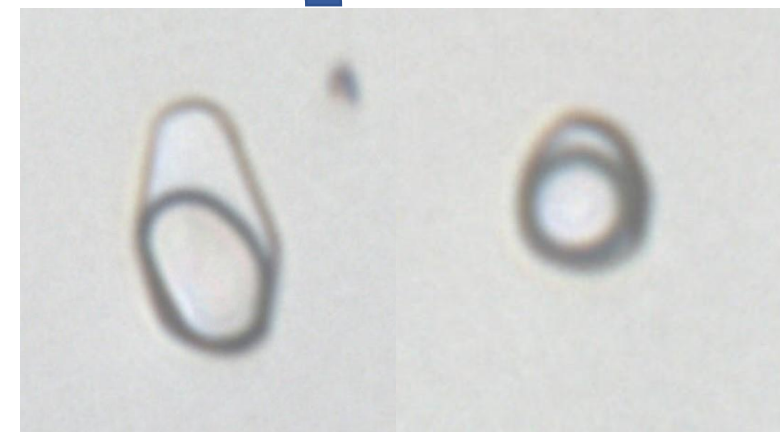
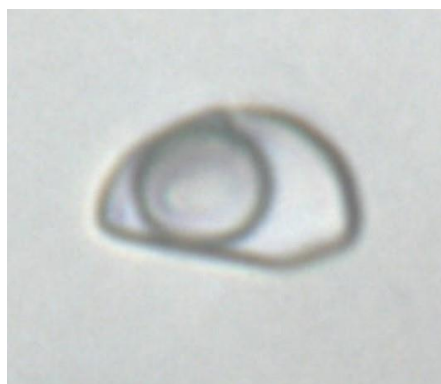
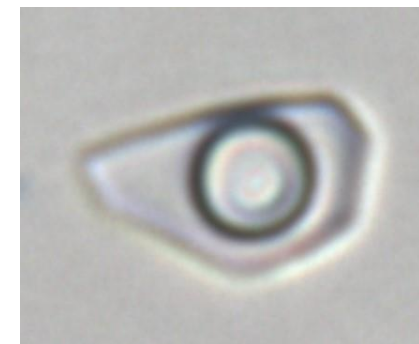
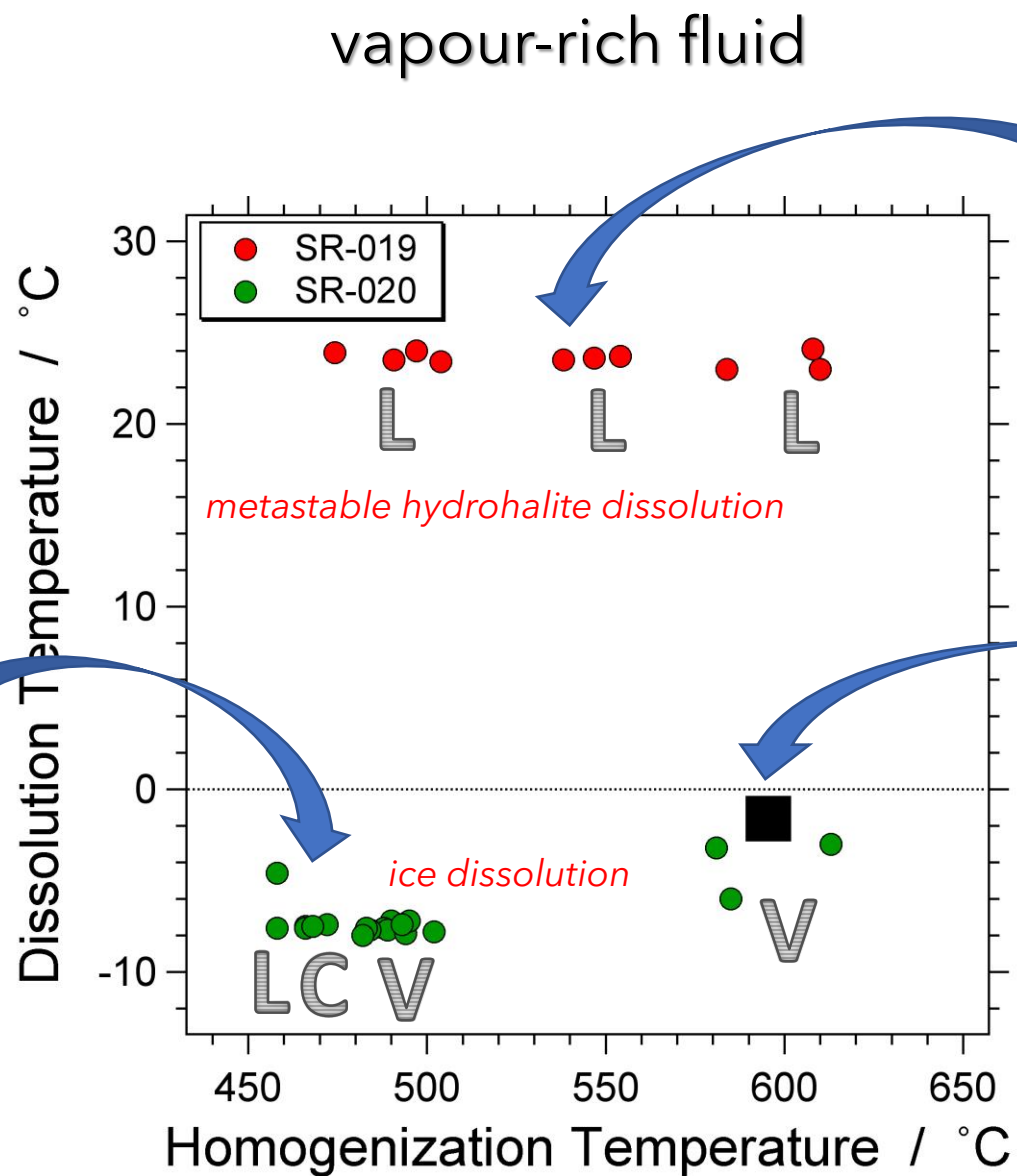
■ theory:

$$T_m(\text{ice}) = -1.4\text{ }^{\circ}\text{C}$$

$$T_h(\text{LV} \rightarrow \text{V}) = 595.4\text{ }^{\circ}\text{C}$$

33.4 vol.% liquid
66.6 vol.% vapour

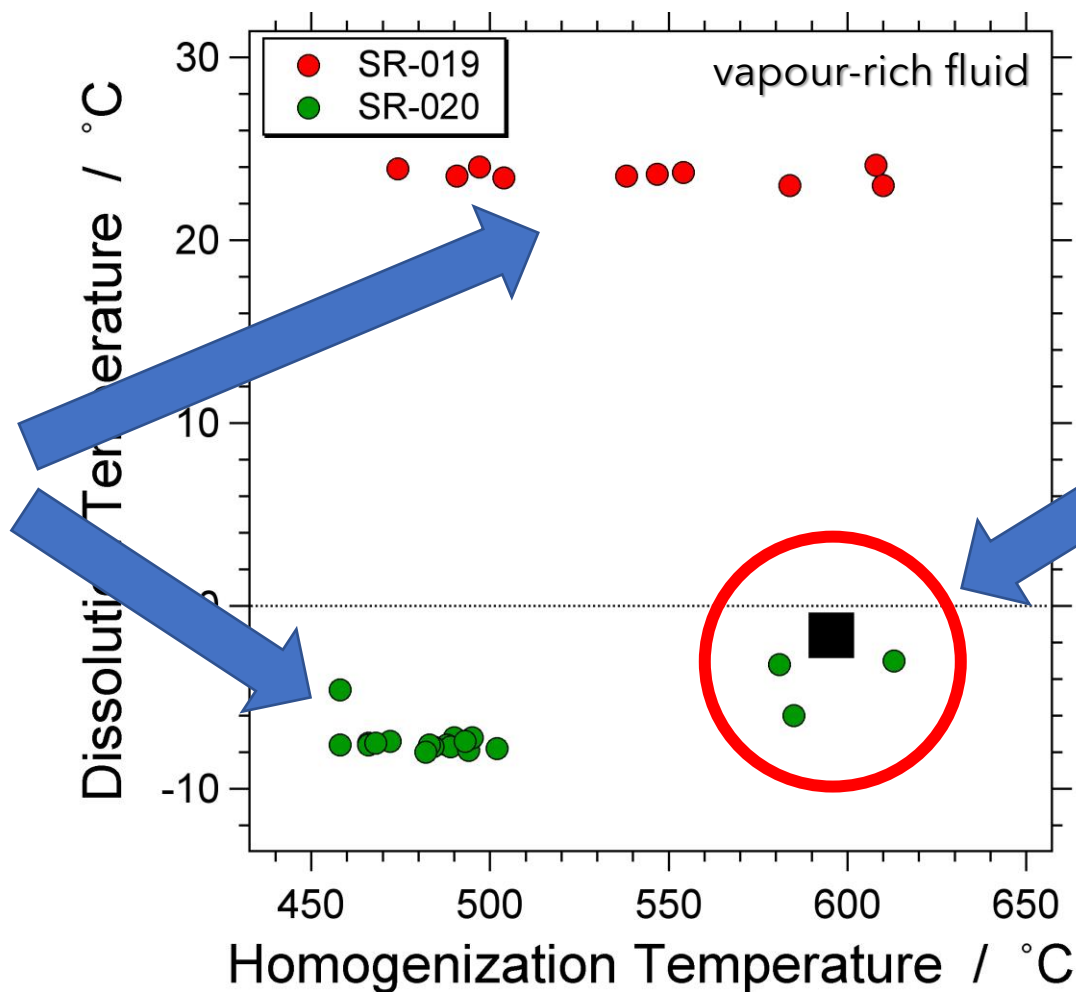
2.71 mass% NaCl
57.10 cm³/mol



interpretation

instantaneous entrapment of
original fluid before immiscibility
and experimental conditions are
reached

(within 30 minutes)



unmixed vapour-rich
fluid only from
original 10 mass%
NaCl solution

(slightly increased salinity)

liquid-rich fluid

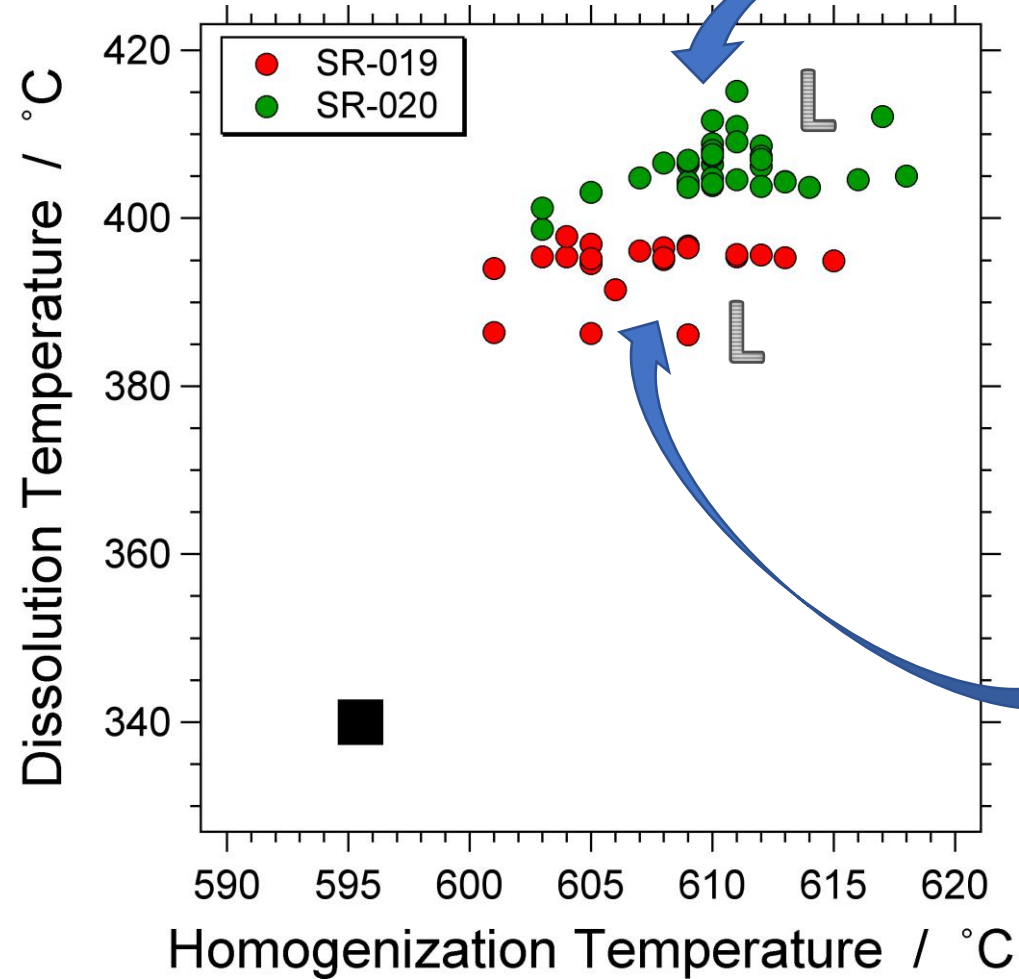
■ theory:

$$T_m(\text{NaCl}) = 349.2\text{ }^{\circ}\text{C}$$

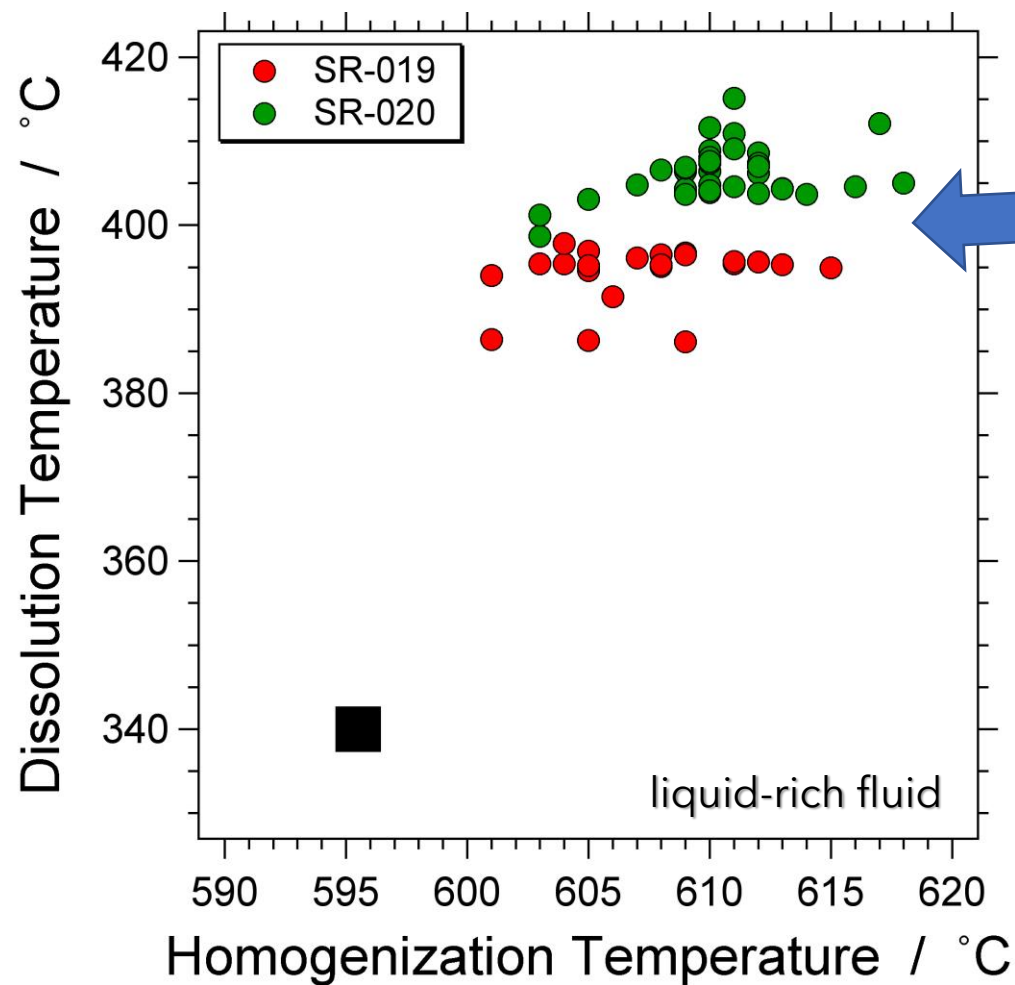
$$T_h(\text{LV} \rightarrow \text{L}) = 595.4\text{ }^{\circ}\text{C}$$

62.4 vol.% liquid
29.8 vol.% vapour
7.8 vol.% solid

40.42 mass% NaCl
29.44 cm³/mol



interpretation



unmixed liquid-rich fluid from
original 10 mass% NaCl solution
and 30 mass% NaCl solution

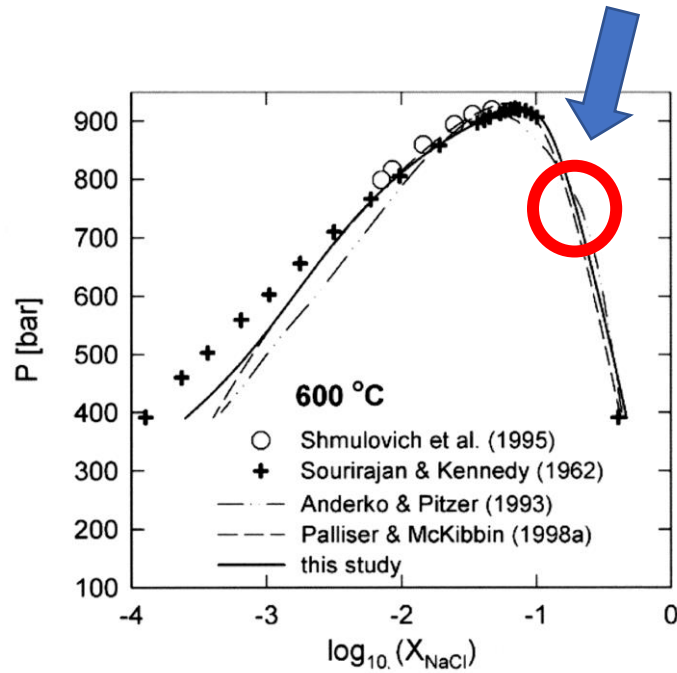
but:

salinity too high
(46.61 to 48.17 mass% NaCl) ?

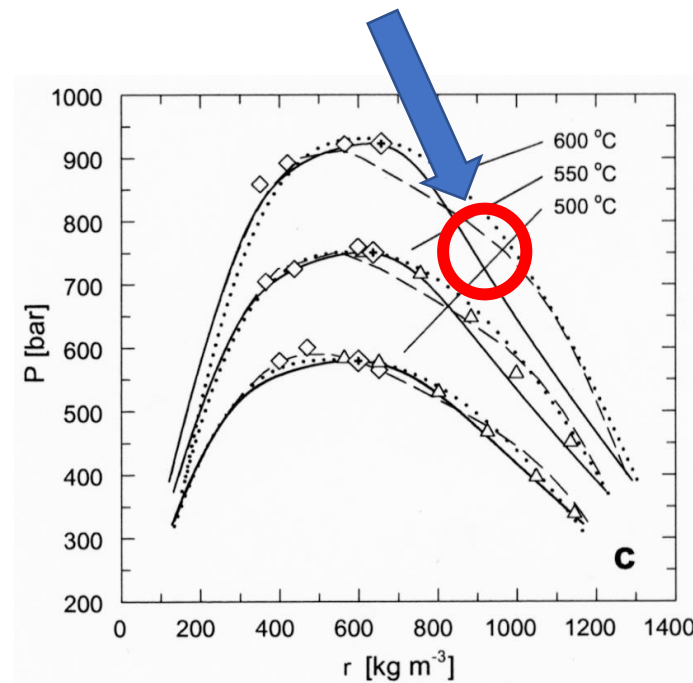
molar volume too low
(density too high) ?
(28.30 to 28.55 cm³/mol)

liquid-rich fluid

intrappolation of fluid model at conditions where data are not available



Driesner and Heinrich (2007)



Driesner (2007)

conclusions

a number of experimental curiosities

immiscibility can only be proven for an original low salinity fluid
where abundant vapour-like fluid is generated by unmixing

relative high salinity liquid-like unmixed fluid is preferentially trapped in fluid inclusions
due to favorable wetting properties in cracks and at crystal surfaces

whereas a low salinity vapour-like unmixed fluid is only seldomly trapped,
and often completely absent

heterogeneous trapping is not evidenced by the experiments,
and probably not a common phenomenon in nature

the present experiments illustrate the weakness of some thermodynamic models
and the need for additional data to improve well-established models