

IN-SITU X-RAY IMAGING OF PHASE SEPARATION IN THE NaCl-H₂O SYSTEM AT HIGH PRESSURE AND TEMPERATURE

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INTRODUCTION

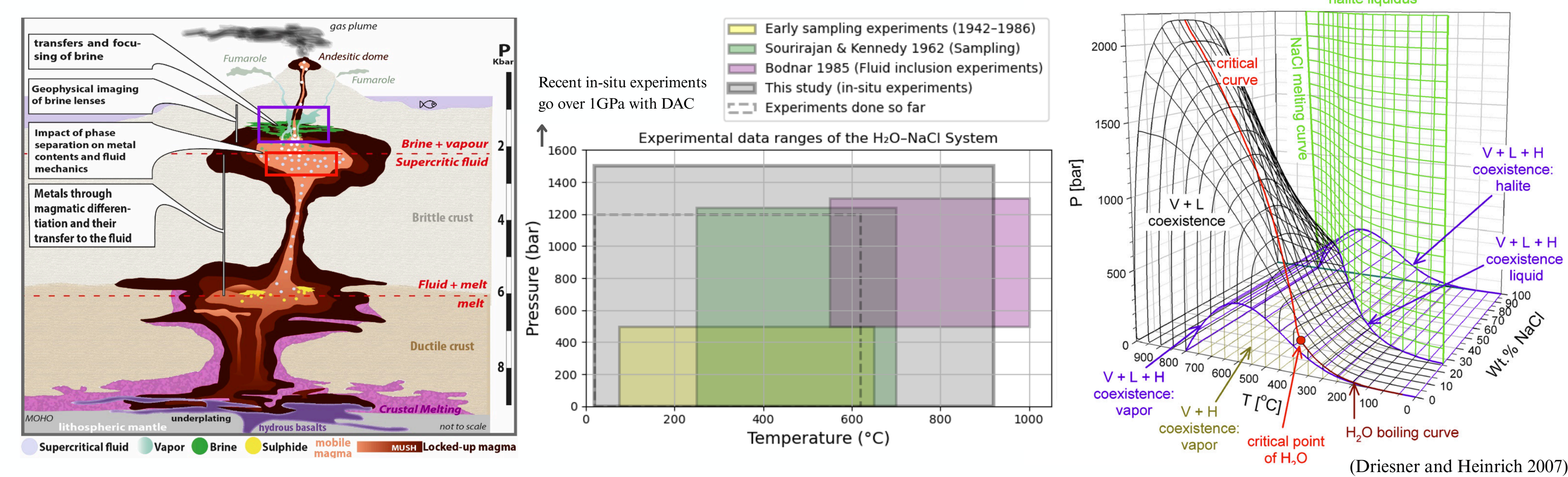
Chlorine rich magmatic fluids play an important role in metal transport and ore enrichment of critical metals like Cu, Au or Mo in hydrothermal systems. [1,2,3] The P-T conditions of the vapor-brine separation in these fluids impact the depth and geometries of metal enrichment. Therefore, understanding this vapor-liquid phase separation in the H₂O-NaCl system is essential for constraining the behaviour of magmatic-hydrothermal fluids. [4]

Previous experimental studies mainly rely on ex-situ methods leaving gaps in P-T coverage and showing discrepancies, particularly at higher P-T-X conditions. As a result, thermodynamic models based on existing experimental datasets lack validation and need to be re-evaluated. [5,6]

The objectives of this study are to:

- Constrain the P-T conditions at which V+L separation occurs in magmatic brines with varying compositions
- Determine the volumetric properties of the brine and vapor phases

The resulting experimental dataset will help improve equations of state and thermodynamic models for brines, with implications for magmatic degassing, hydrothermal circulation, and ore formation.

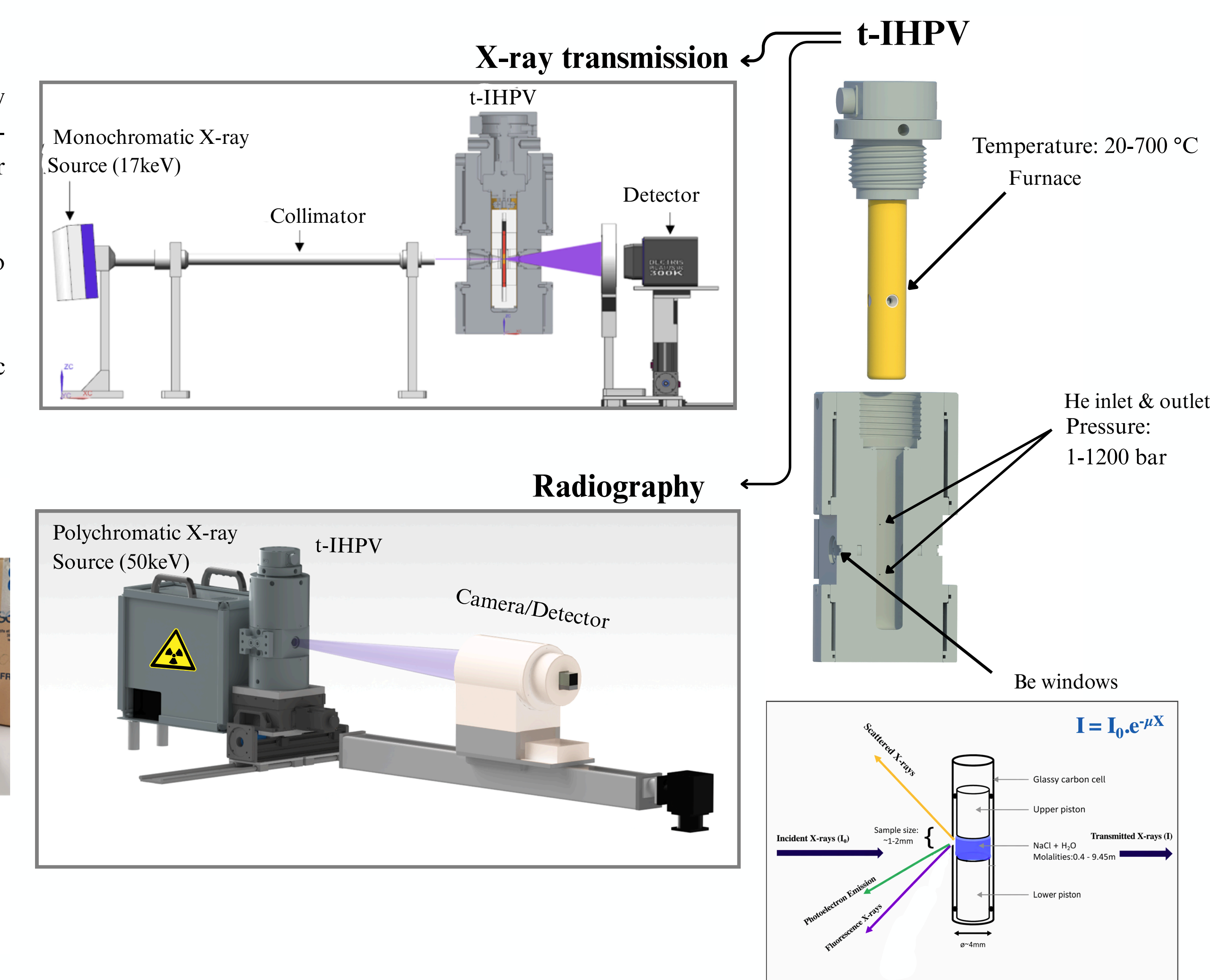
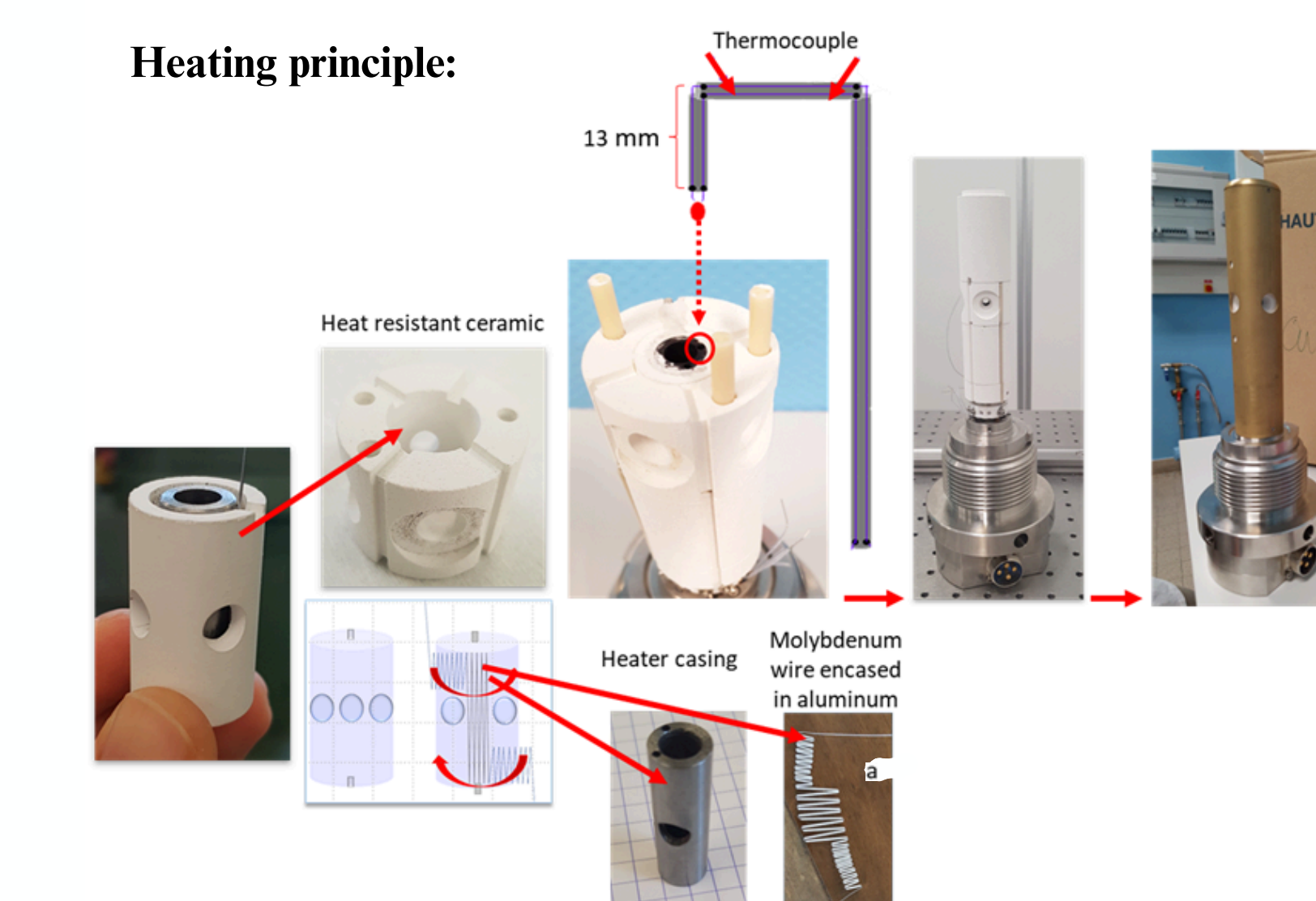


METHODOLOGY

Experiments were performed using a transparent internally heated high pressure vessel (t-IHPV) designed for in-situ X-ray (Radiography/XAS/WAXS) measurements up to 2000 bar and 1200 °C at ISTO and ESRF, Grenoble.

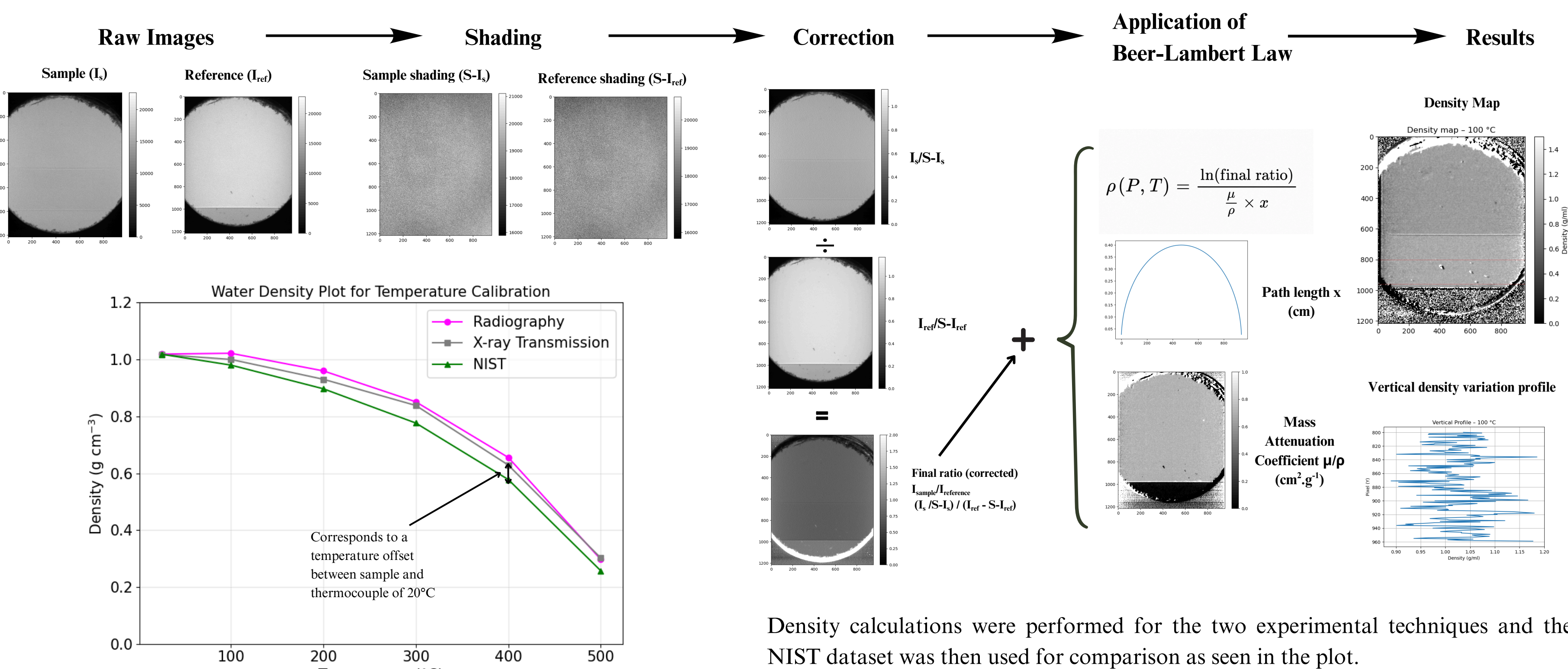
Experiments consisted in subjecting aqueous NaCl solutions to increasing temperature conditions under isobaric conditions.

The phase separation can be identified through radiographic imaging as a change in the X-ray attenuation contrasts.

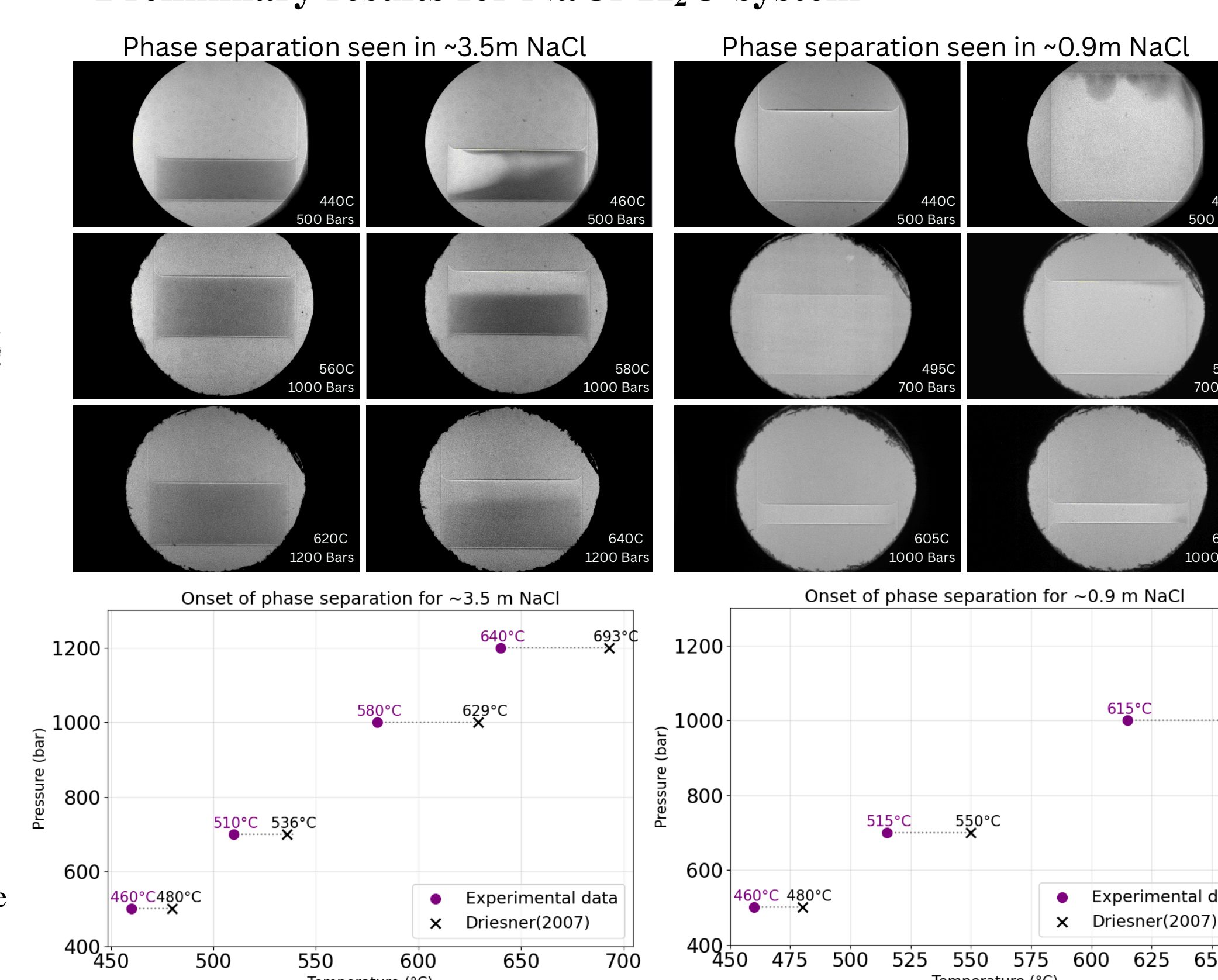


RESULTS AND OBSERVATIONS

Temperature Calibration



Preliminary results for NaCl-H₂O system



- The onset of vapor-liquid phase separation in H₂O-NaCl solutions is monitored in the radiographic images as a function of NaCl concentrations.
- More concentrated solutions (e.g. ~3.5m) exhibit well-defined phase separation with large distinct brine layers. In contrast, less concentrated solutions (e.g. ~0.9m) produce significantly smaller volumes of brine, resulting in small bubbles concentrated near the cell's pistons.
- Experimental observations show deviations from thermodynamic predictions, particularly at higher P-T conditions compared with the Driesner and Heinrich (2007) [6].
- The observed variability in brine distribution suggests that sampling-based [9] and synthetic fluid inclusion based [5] experimental approaches may not always capture representative phase compositions.

*The true temperatures may be slightly lower by ~20°C; calibration is underway.

KEY CHALLENGES

- Temperature calibration is difficult due to very small sample sizes, possible heating gradients, and the varied efficiency of heaters that are often replaced.
- Thermocouple readings = precise but not always accurate
- Vapor and brine phases can be unstable and unevenly distributed, affecting X-ray transmission and density results.

CONCLUSIONS & PERSPECTIVES

Take-home message

- Direct in-situ radiographic observations of fluid phase separation provides new constraints on saline fluid behaviour at crustal conditions
- The results shift the domain of V+L phase separation to lower temperatures compared to the current models (e.g. Driesner 2007)

Moving further, we plan to:

- Extend experiments to higher P-T conditions (upto 1500 bar and 900 °C) and obtain PVT properties (e.g. density).
- Investigate additional systems including H₂O-KCl, H₂O-CaCl₂, and H₂O-FeCl₂, which are important components of natural hydrothermal brines and will help improve experimental and thermodynamic models of magmatic-hydrothermal brines.

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