

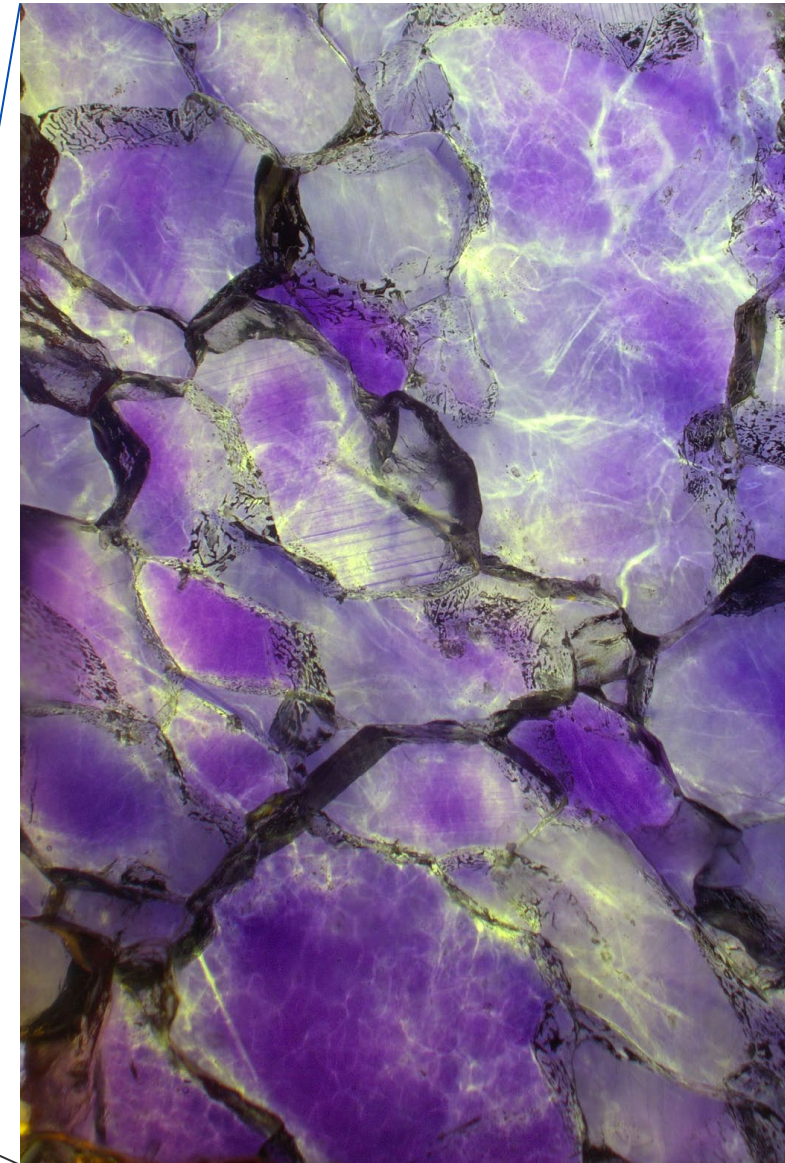
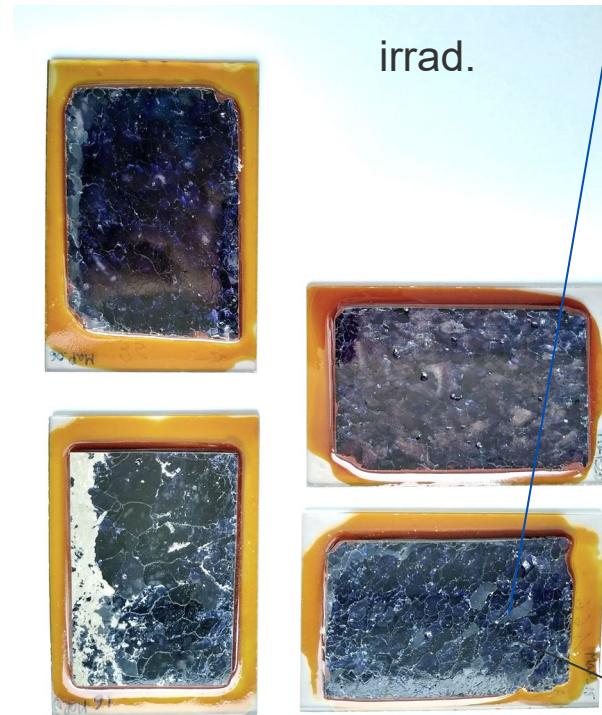
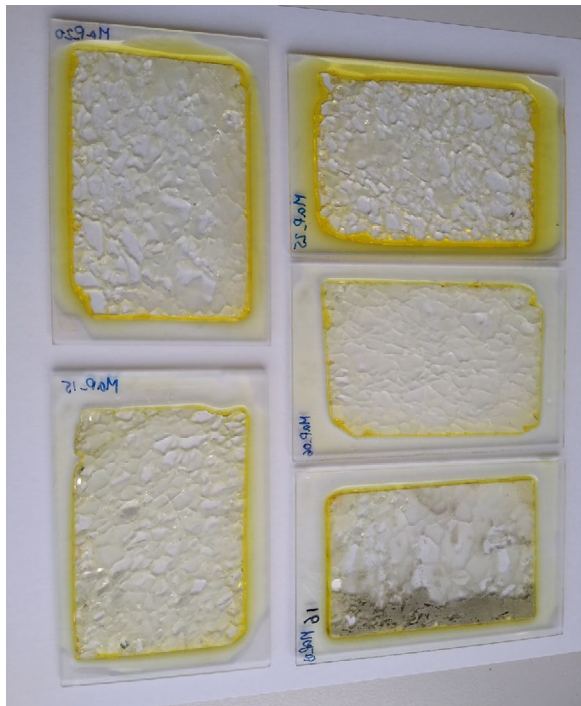
Gamma decoration at FRM II: recent optimizations and parametric studies for microstructural investigations on halite

Vladimir Hutanu¹, Xiaosong Li¹ and Joyce Schmatz²

- 1) FRM II, TUM, Garching, Germany
- 2) MaP GmbH, Aachen, Germany

USP Direct and volumetric visualization of hidden features like:

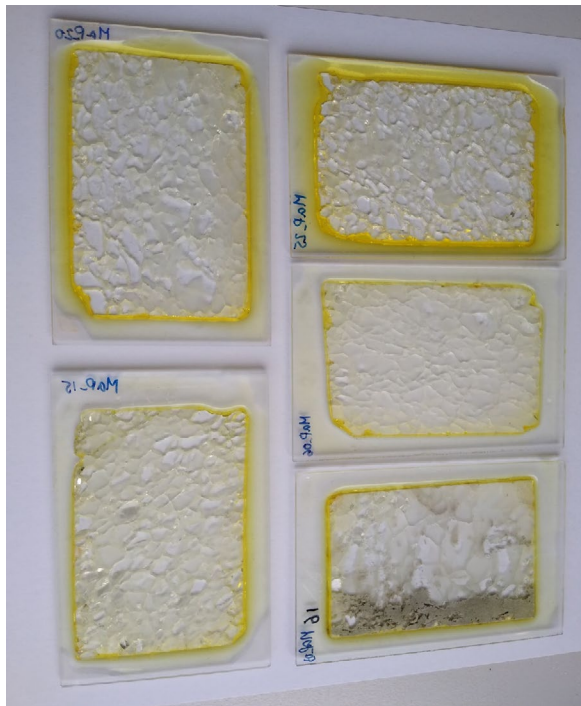
- Grain boundaries, subgrain boundaries, growth bands, zonation, impurity content within and between grains, pores, fluids on the grain borders, grain rotation and deformation history.
- Slipping bands, newly formed, strain-free grains can be distinguished from older, deformed grains, providing insights into recrystallization mechanisms, paleostress estimation (piezometry).



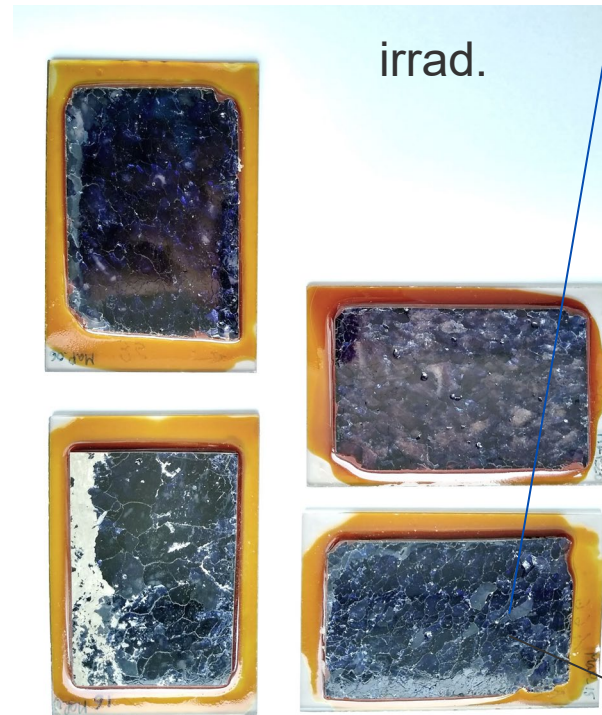
pristine non-irrad.

Direct and volumetric visualization of hidden features like:

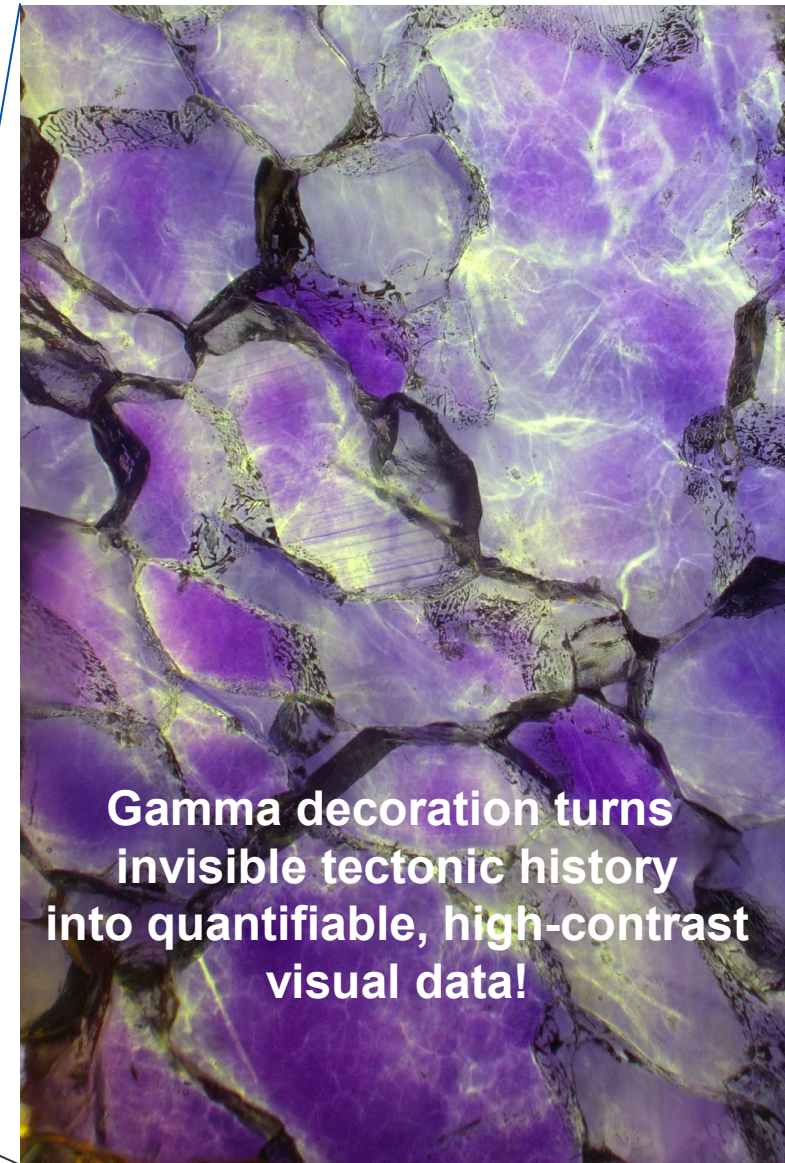
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pristine non-irrad.



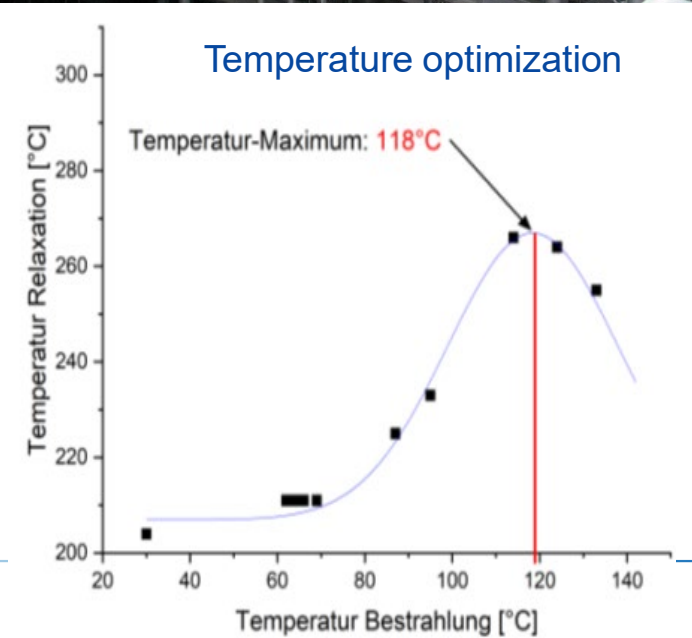
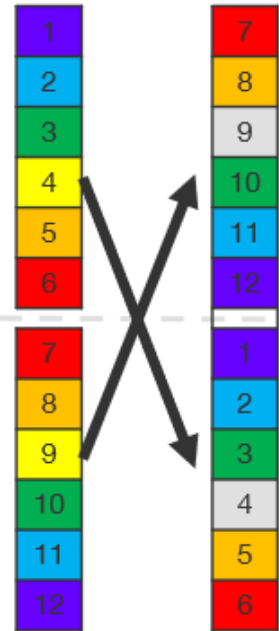
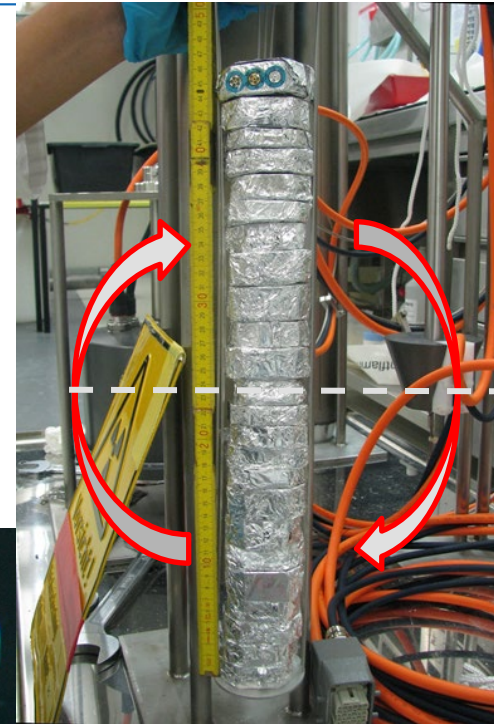
irrad.



Gamma decoration turns invisible tectonic history into quantifiable, high-contrast visual data!



Large samples
homogeneous
irradiation



Irradiation position and
time (dose) optimization

ISO 9001:2015 certified
process!

Modified Jain-Lidiard theoretical model of radiation damage in single crystals

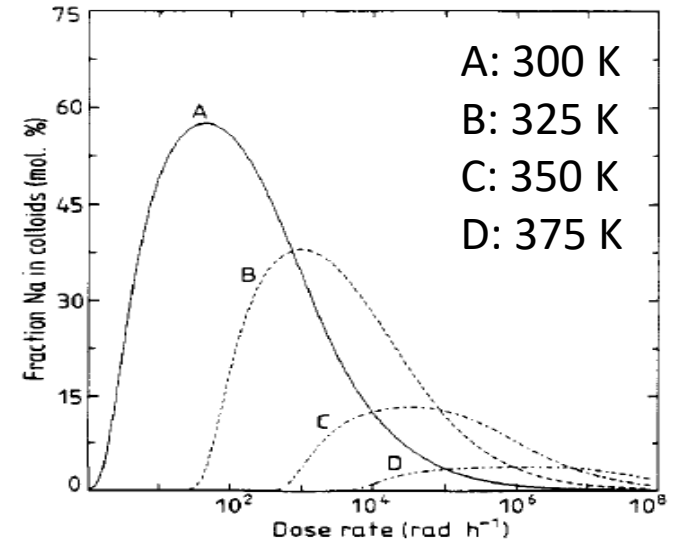
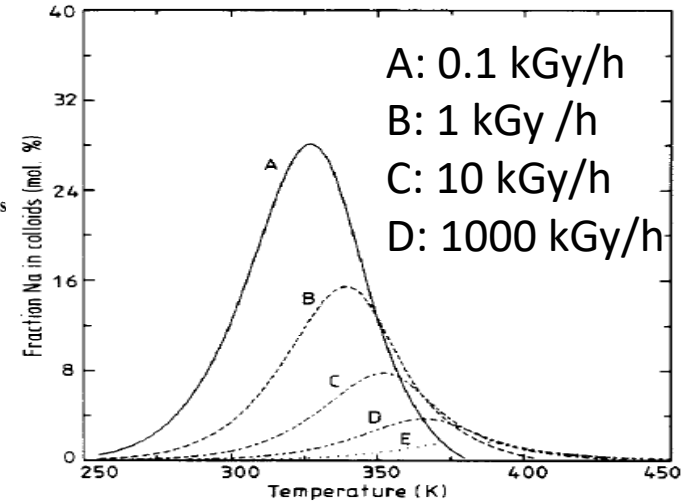
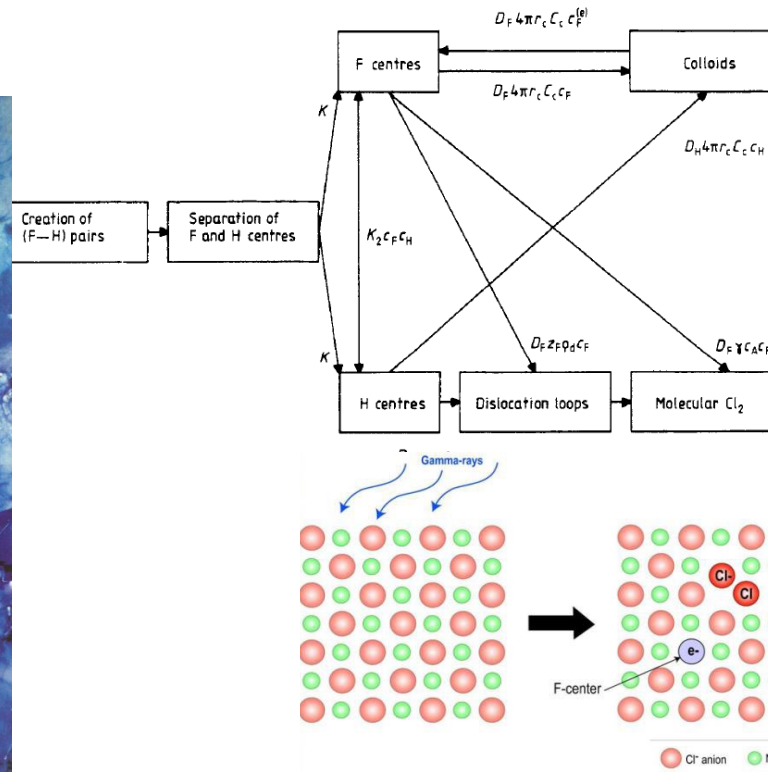
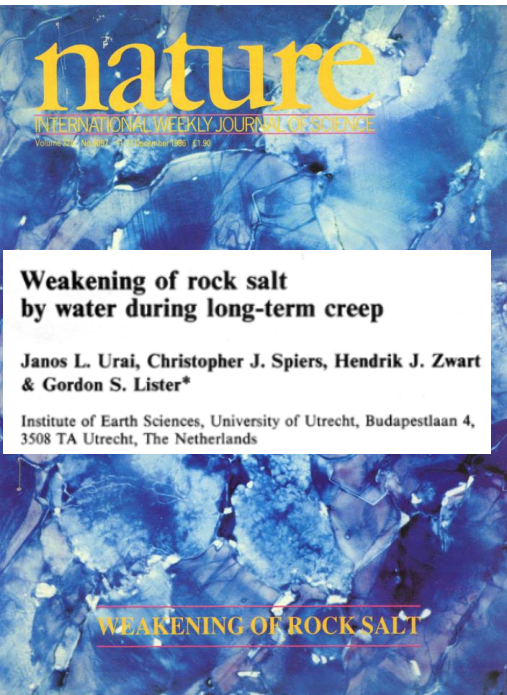
J. Phys. C: Solid State Phys., 18 (1985) 257–268. Printed in Great Britain

Radiation damage of NaCl: dose rate effects†

G van Opbroek and H W den Hartog
Solid State Physics Laboratory, 1 Melkweg, 9718 EP Groningen, The Netherlands

- only one application irradiation protocol:

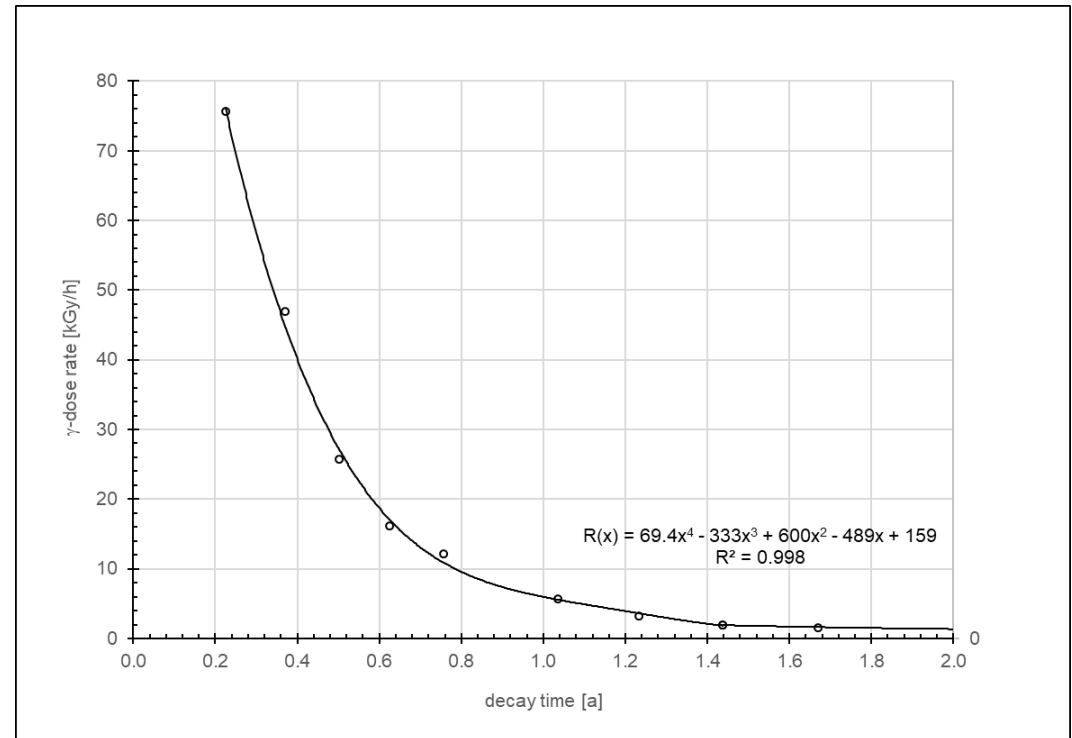
- 4 MGy total dose at 100°C found by chance in the late 1980-th with 10-20 kGy/h



Time- decay of dose-rate in spent FE

- Fast time decay within first 1.5 a after cycle (short living isotopes): (from 75 kGy/h (100 d) to 2 kGy/h (500 d))
- Mixed decay region 1.5 - 3.5 a
- Slow quasi-monoenergetic (Cs-137) decay afterwards ($T_{1/2} = 30$ a)

Stable analytical function fit in three time regions with predictive power.



X. Li, OPA 00569: Gamma-Dosisleistungsmessung in abgebrannten BE (2025)

X. Li, OPA 00328: Gammabestrahlungsanlage in abgebrannten BE JBE 10 (2011)

Master's Thesis

**Experimental study on coloration efficiency
in halite by gamma irradiation in spent fuel
elements at FRM II**

Department of Earth and Environmental Sciences
Ludwig-Maximilians-Universität München

Naritai

Munich, Oct. 15th, 2023



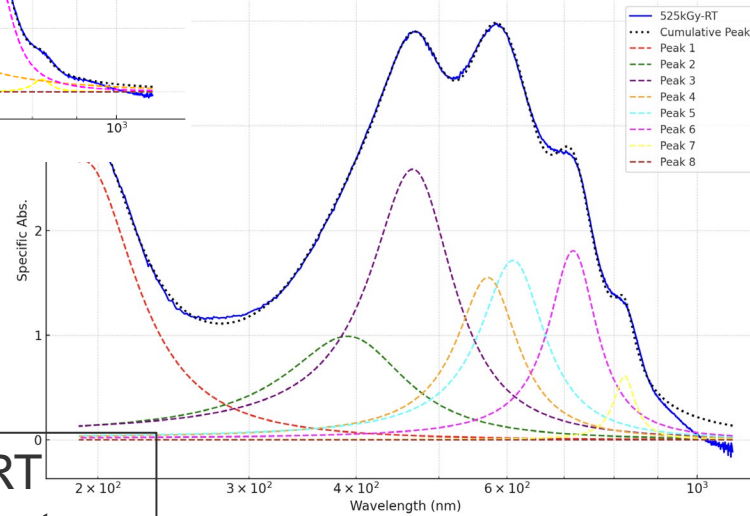
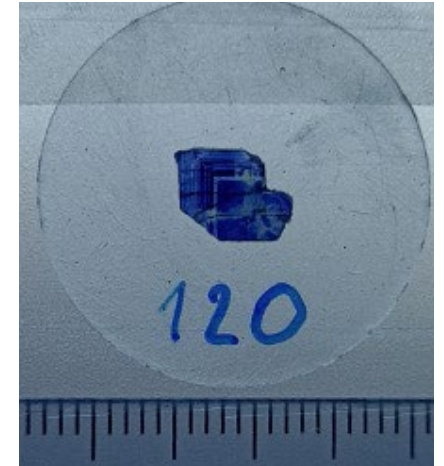
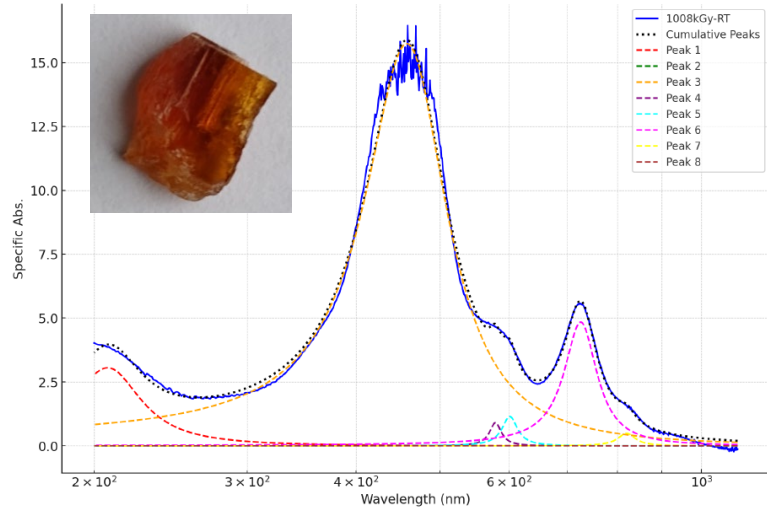
Submitted in partial fulfillment of the requirements for the degree of M. Sc.
Supervisor: Prof. Dr. SoHyun Park
Co-Supervisor: Dr. Vladimir Hutanu

- Natural rock salt from Heilbronn Germany has been irradiated to various total dose at different temperatures in GAB at FRM II;
- Samples have been investigated by UV-vis-NIR optical spectroscopy;
- X-Ray single crystal diffraction;
- Raman spectroscopy;
- X-Ray fluorescence for impurity determination;
- Optical microscopy

Results compared to previous literature



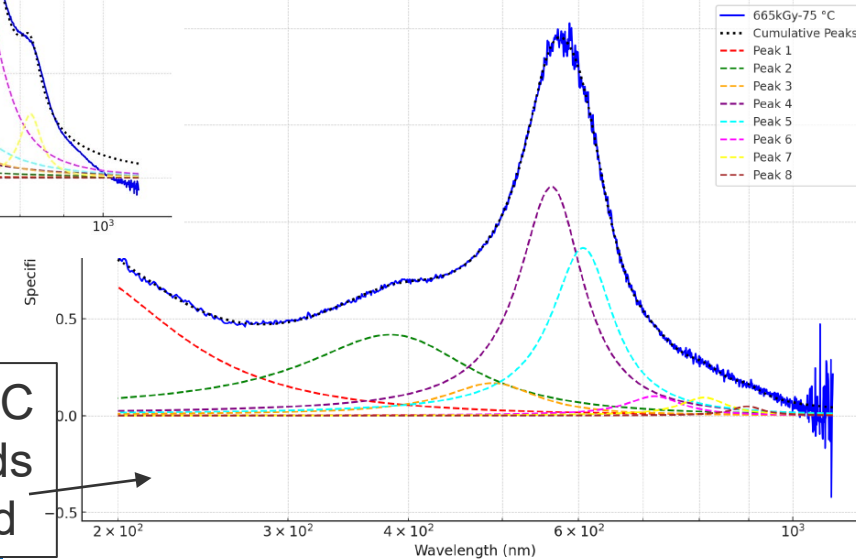
UV-Vis-NIR optical spectroscopy



550 kGy@RT
470 nm F- centers

1100 kGy@RT
470 nm F- centers
+ 560 nm colloids

670 kGy@75°C
560 nm colloids
dominant band

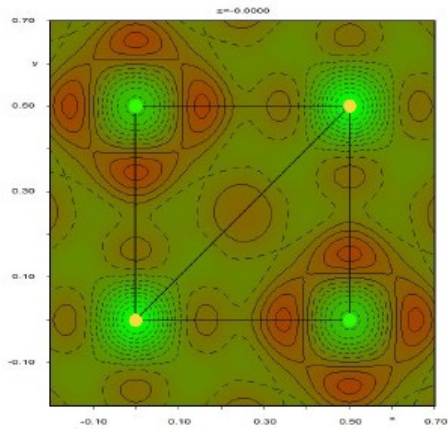


The number of the defects is estimated from the peaks-fit by Smakula formula

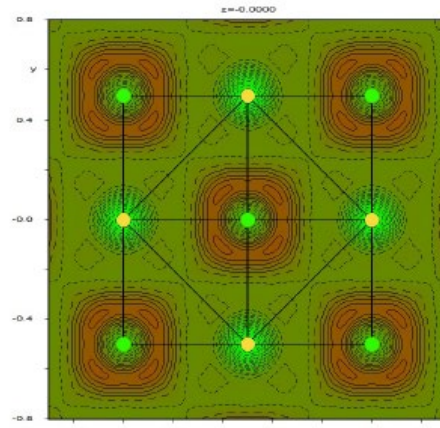
XRD at LMU

Significant lattice distortions depending on irradiation dose and temperature.

- 1 MGy total dose at RT irradiation leads to less distortions than e.g. 0.55 MGy at 75°C. Strong temperature effect in colloidal formation;
- Clear shift from the high symmetry positions is observed at highly irradiated samples (lattice shrinks)

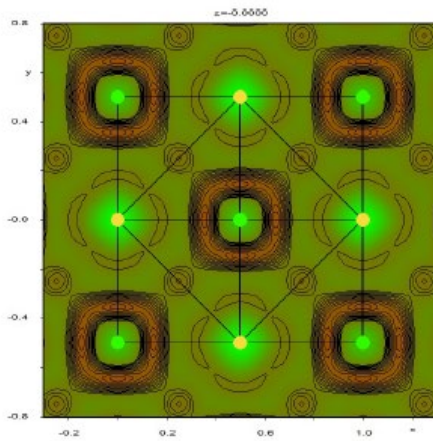


(a) Difference Fourier Map of non-irradiated halite single crystal at $Z = 0$

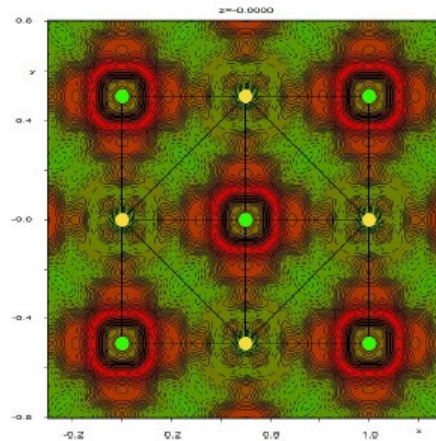


(b) Difference Fourier Map of 525 kGy-RT halite single crystal at $Z = 0$

Figure 22: Difference Fourier Map



(a) Difference Fourier Map of 1008 kGy-RT-halite single crystal at $Z = 0$



(b) Difference Fourier Map of 665 kGy 75°C halite single crystal at $Z = 0$



Bachelorarbeit

im gemeinsamen Studiengang
Bachelor of Science in Geowissenschaften
der Ludwig-Maximilians-Universität München
und der
Technischen Universität München

Farbeffizienz der Gammabestrahlung und nachfolgender Wärmebehandlung zur Herstellung von blauem Halit

Color efficiency of gamma irradiation and subsequent
heat treatment to produce blue halite

eingereicht am Münchner GeoZentrum

Julia Bachl

Matrikelnummer: 12401655 (LMU), 03756885 (TUM)

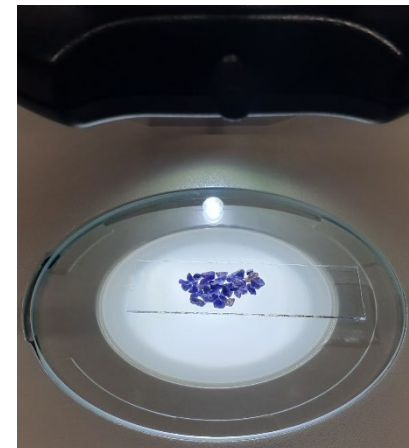
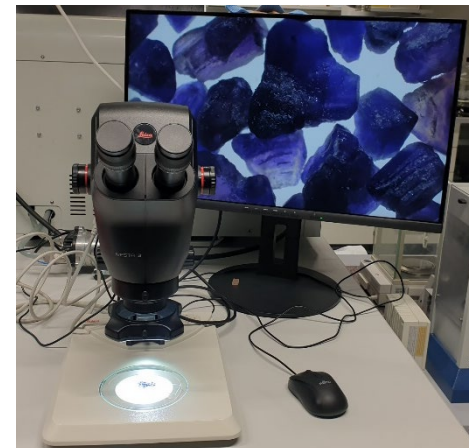
Datum der Abgabe: 07.04.2025

Betreuerin: Prof. Dr. SoHyun Park (Lehrstuhl für Kristallographie, LMU)

Co-Betreuer: Dr. Vladimir Hutanu (FRM II, TUM)

- Natural rock- salt and sea- salt from various origins (Italy, Germany, Netherlands, Pakistan, Iran, Spain, Portugal, Norway, Russia) has been irradiated to intermediate total dose at different more systematic temperatures in GAB at FRM II;
- Samples have been investigated by Digital Image colorimetry
- Chemical colorimetry for measuring free Cl_2 content
- Micro-Raman spectroscopy
- The irradiated samples have been subject of subsequent thermal annealing in order to study their relaxation behaviour in dependence on dose and irradiation temperature.

The comparison between different sample types and irradiation conditions have been performed as well as compared to previous results both original and from the literature.



Chemical Colorimetry. First time direct quantitative evidence of free Cl in irradiated salt.

Chemical analysis with the 'Pocket Colorimeter II' from Hach:

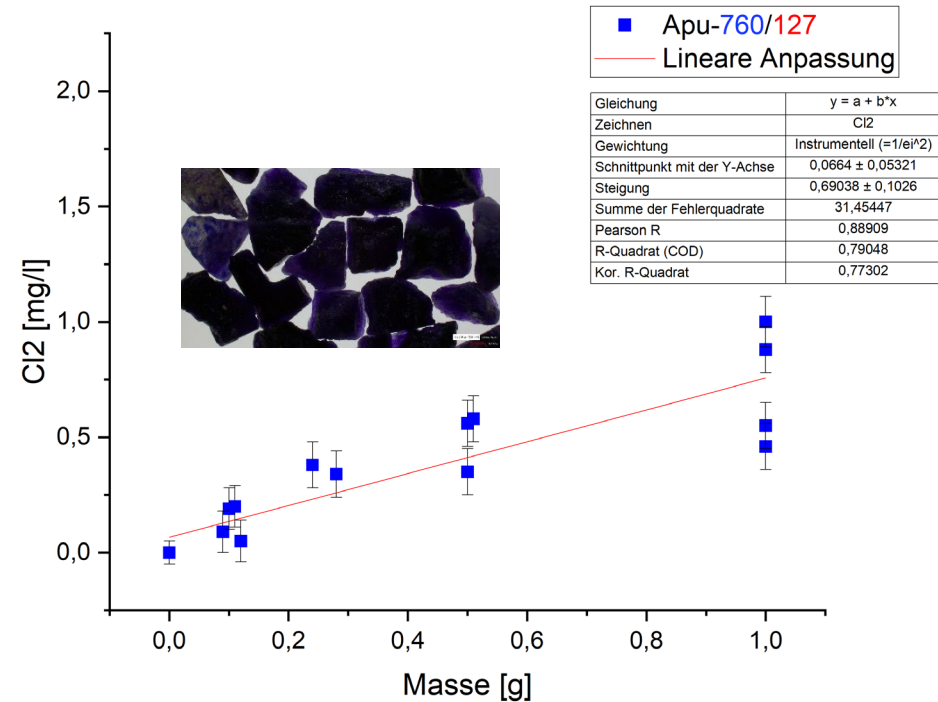
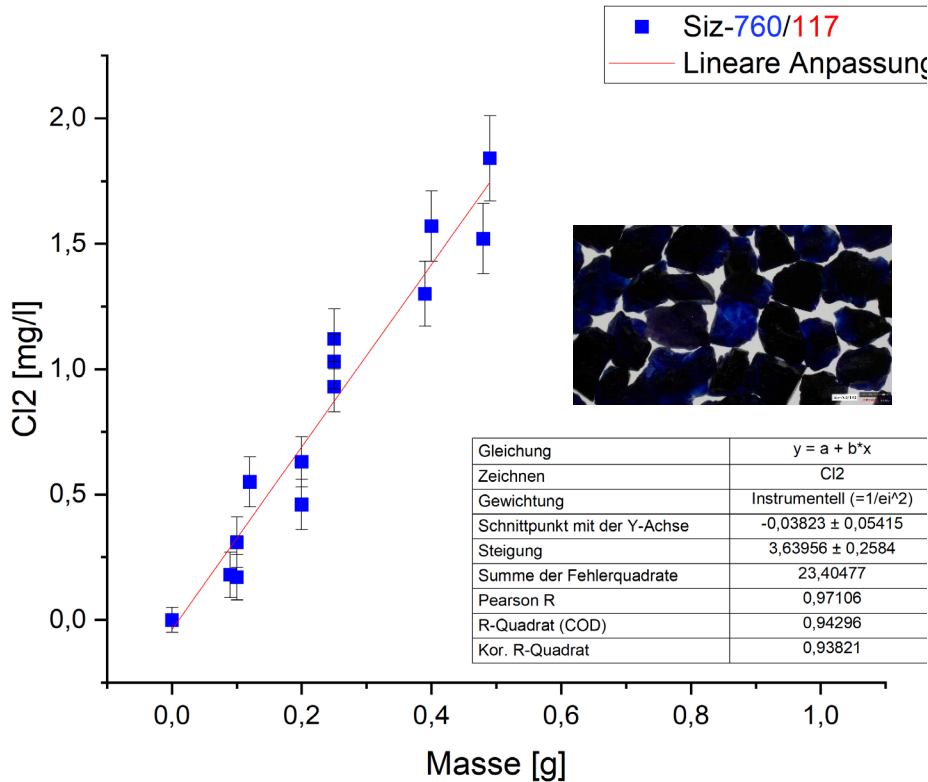
- According to the standardized procedure of the USEPA DPD Method 8021 with powder pillows
- Portable measuring device for water analysis
- Based on photometric measurement methods: the device measures the light absorption of the sample and calculates the concentration of free chlorine

Measurement procedure:

- Dissolving a specific amount of salt in 10 mg of distilled water
- Adding a specific reagent and shaking the sample for 20 seconds
- The sample changes color depending on the chlorine content:
high Cl content → intense magenta
low Cl content → weak magenta
- Unirradiated salt: $\text{Cl}_2 = 0$ [mg/l]



Chemical Colorimetry - Measurements

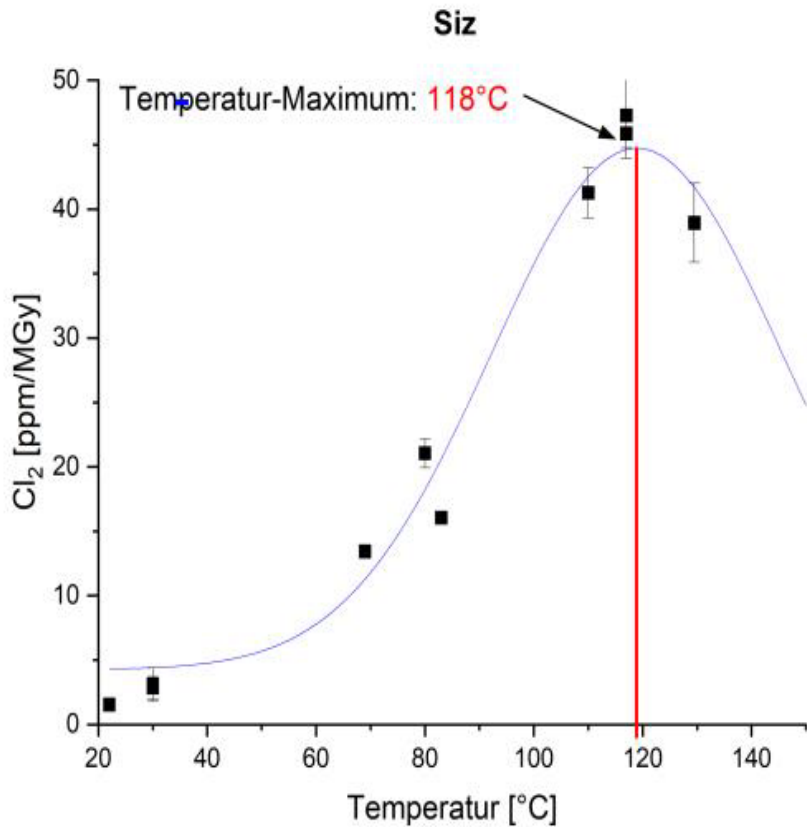


- Blue color due to Na-colloids correlates with the chloride concentration in the lattice
- Differences in the various types of salt are noticeable

Max depends on salt type, deep, impurities, but not on dose rate.

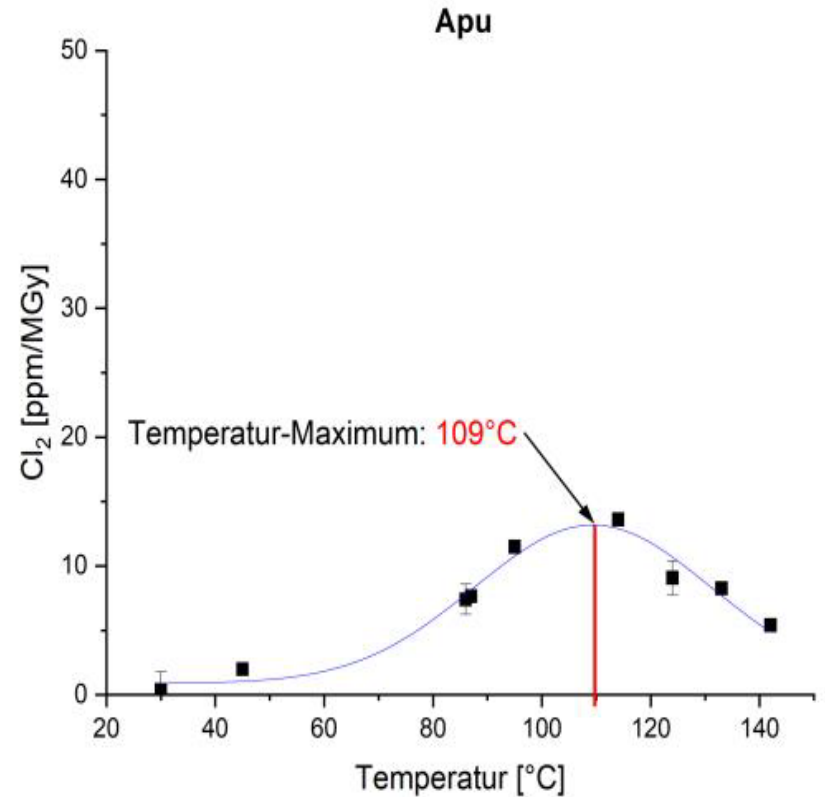
Jain-Lidiard theoretical model is incomplete!!! No one from tetsed salts shows $T_{\max.} < 105^{\circ}\text{C}$ and literature reported value for pure lab grade NaCl without stress.

a

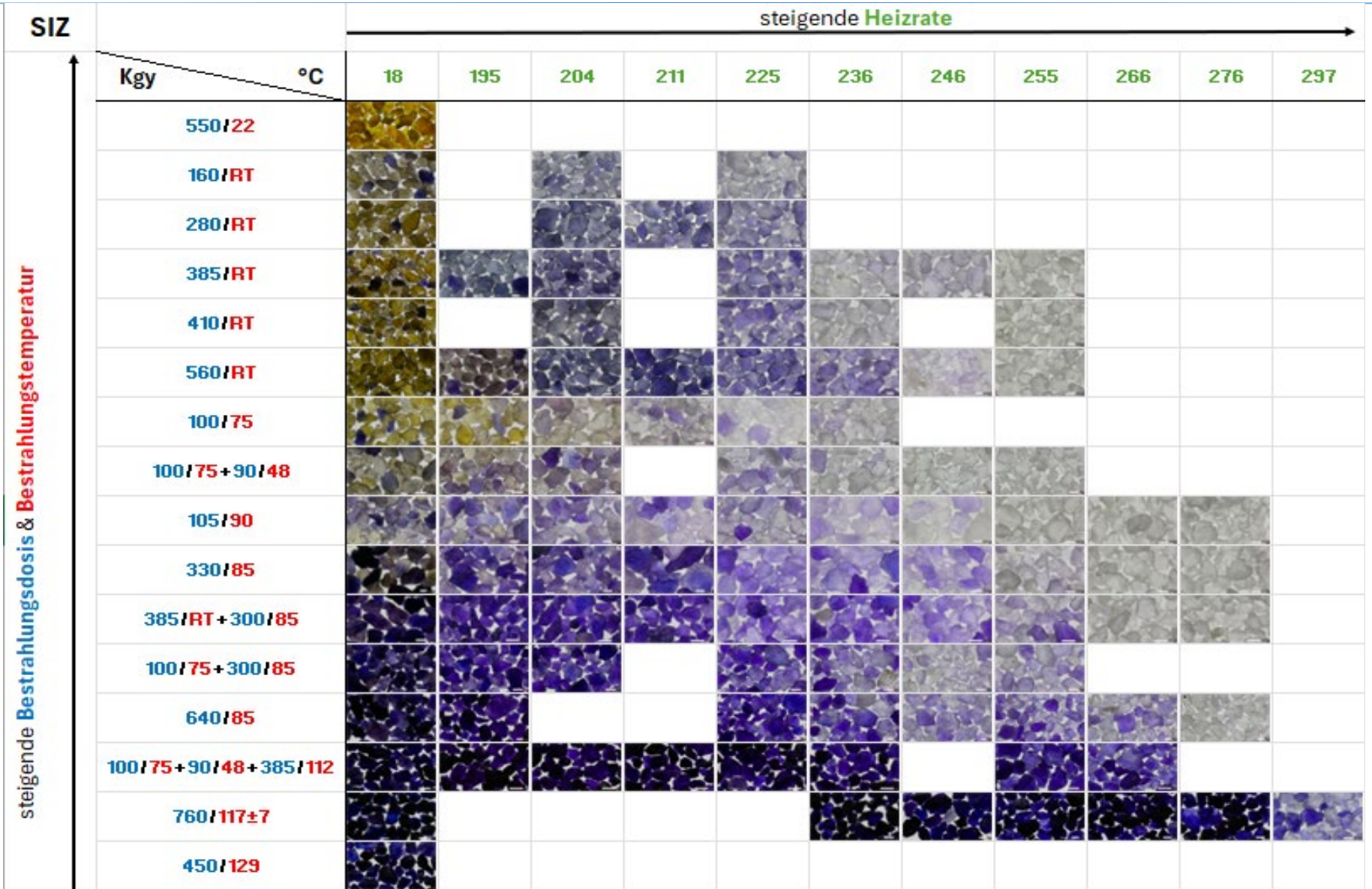


Rock salt 500-600 m, 7 Mio. a

b

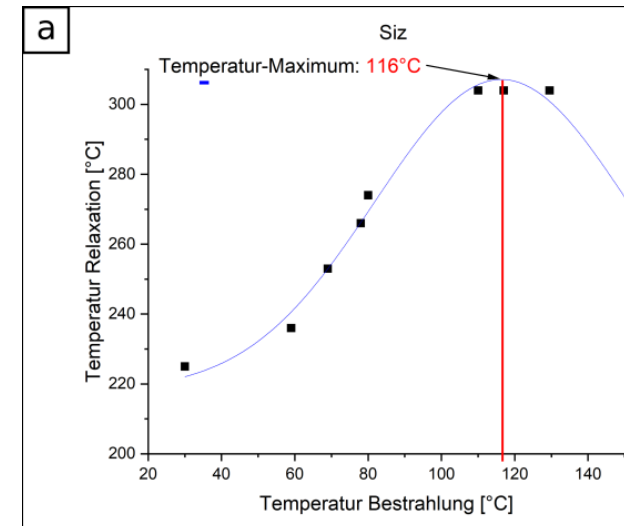
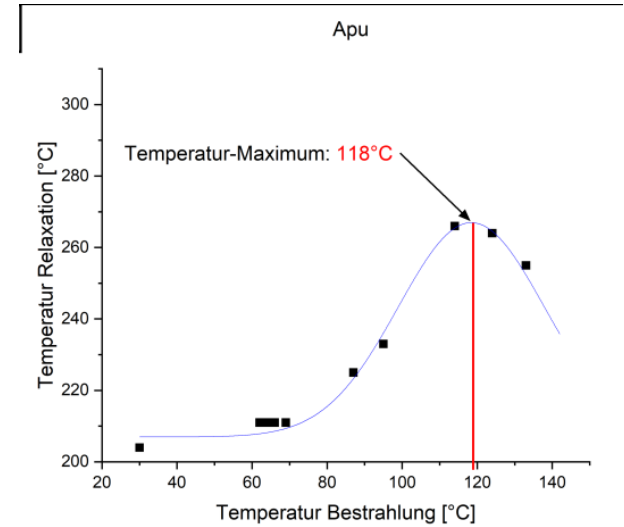
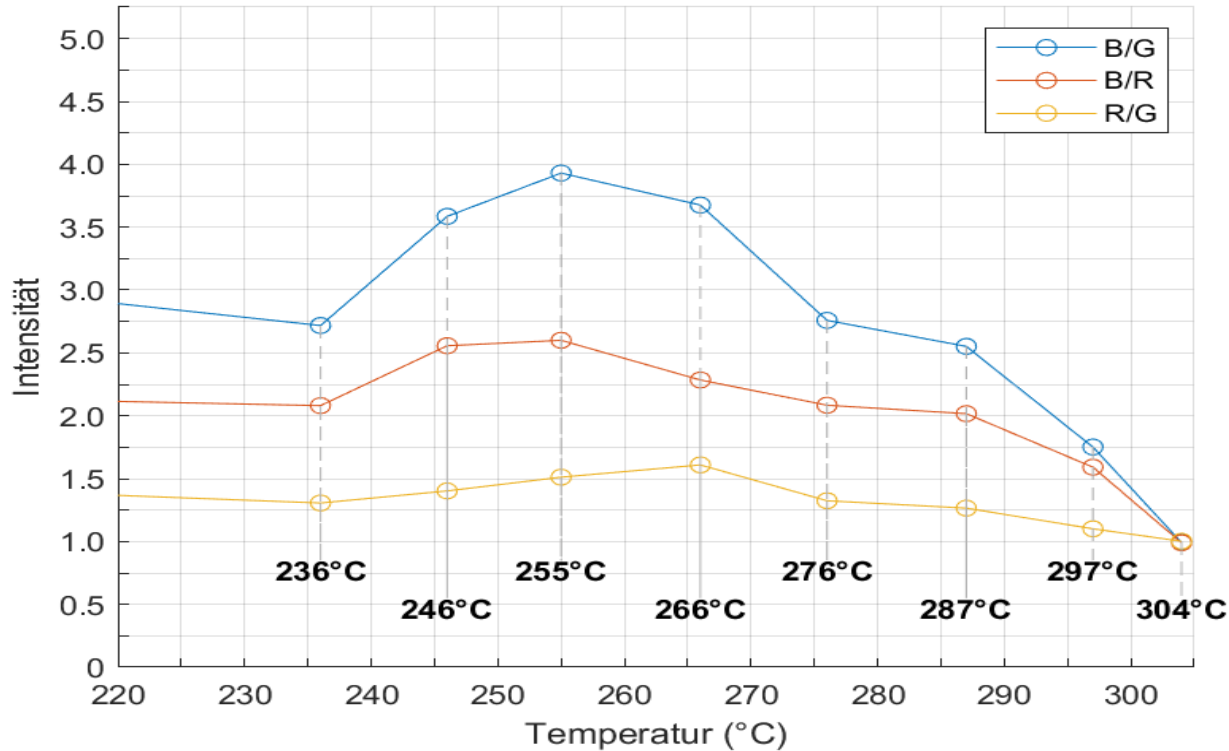


See salt, 0 m, 2 a



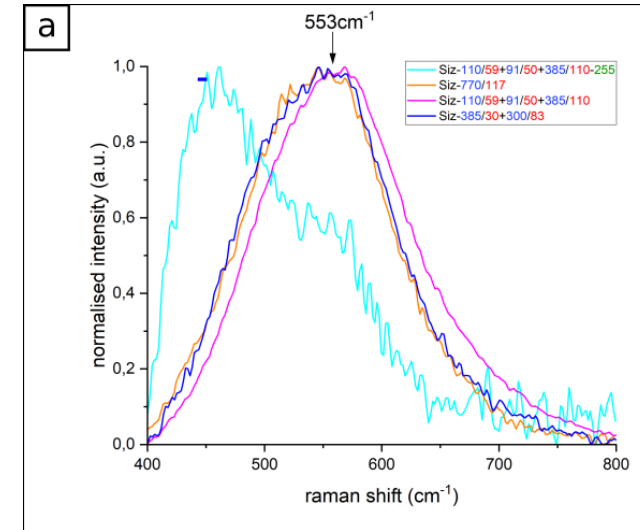
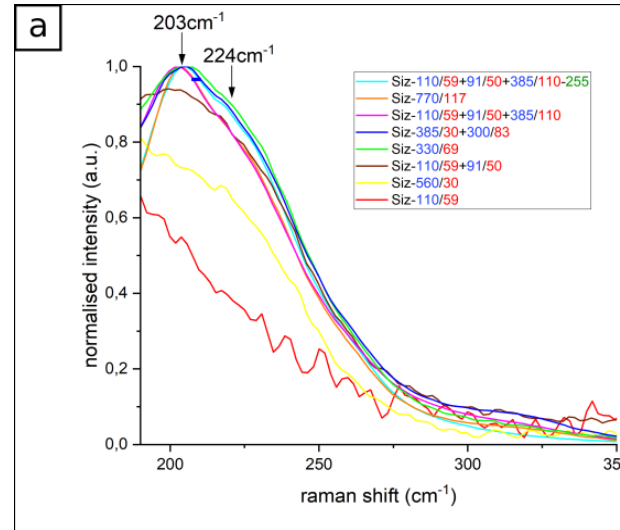
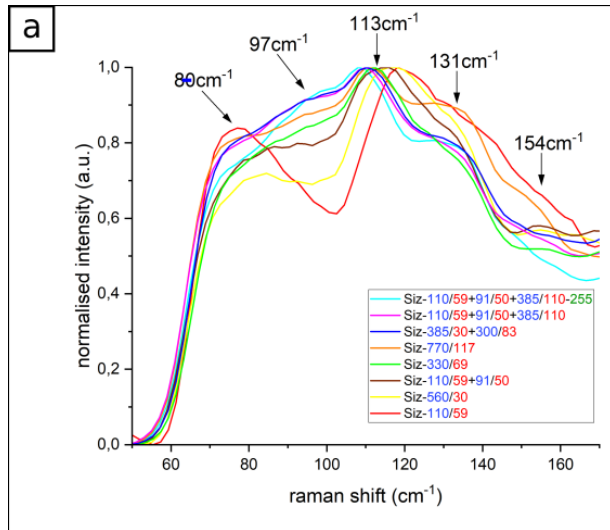
Digital color analysis of the pictures after subtracting bgr. mask

**Verhältnisse der RGB-Werte
Siz-770/117**



Very good correlation between color analysis and free Cl₂ measurements !

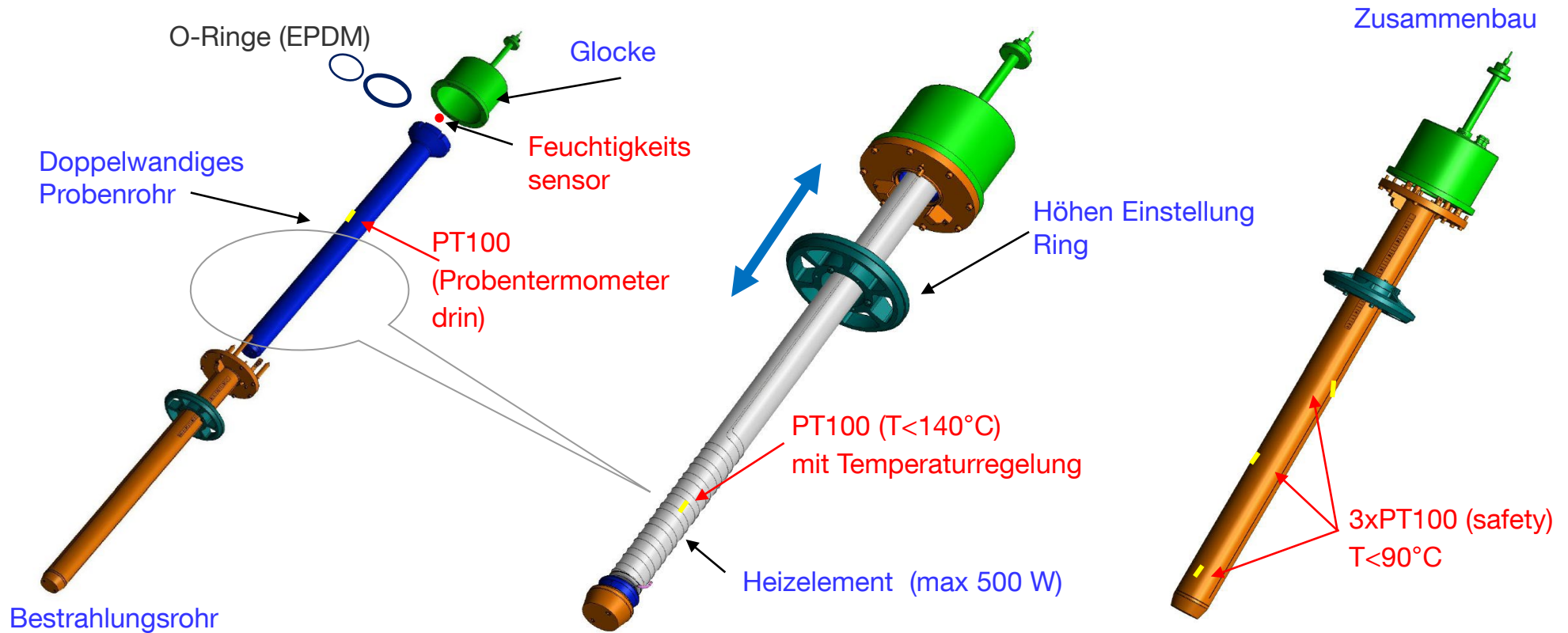
More in-detail Raman analysis shows complex band structure related to the presence of nanoparticles directly



Main findings: - contrary to widely postulated dose-rate dependence of the radiolysis created free colloidal products in NaCl, our result do not support this approach.

- Temperature plays dominant role facilitating creation of the secondary collective defects starting from the primary point defects (F-centers). This process starts immediately after creation of first point defects by irradiation and not as typically proposed only at very high irradiation dose.
- Creation of nanostructured material with controlled size and amount of Na metallic nanoparticles or Cl₂ nanobubbles in NaCl matrix is feasible and reliable.

Aufbau GAB



Control

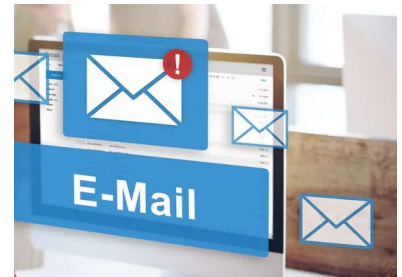
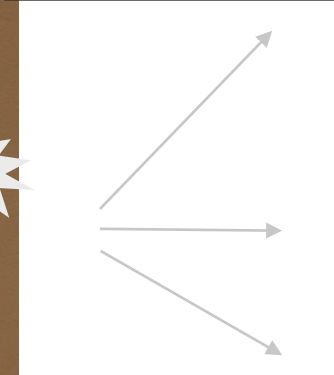
- JUMO LogoScreen nt, PCC, PCA3000



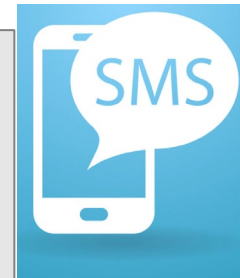
Temperatur Kontroller
preset nur manuell



Fehlermeldungen
über RPA
Sammelleitung
in die Warte



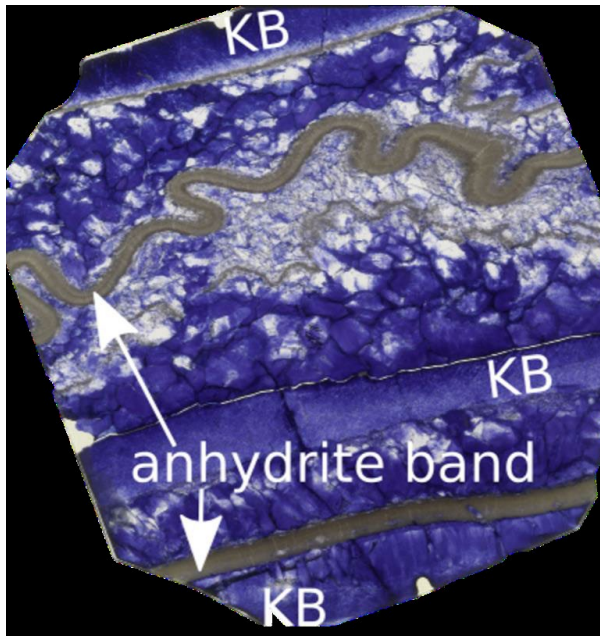
z.B. kein Kontakt,
Überschreitung
Temp., Leckage,
(Feuchtigkeit >
75%), etc.



Case Study 1: The Mechanics of Mixture Rheologies

2.4 MGy @ 70°C

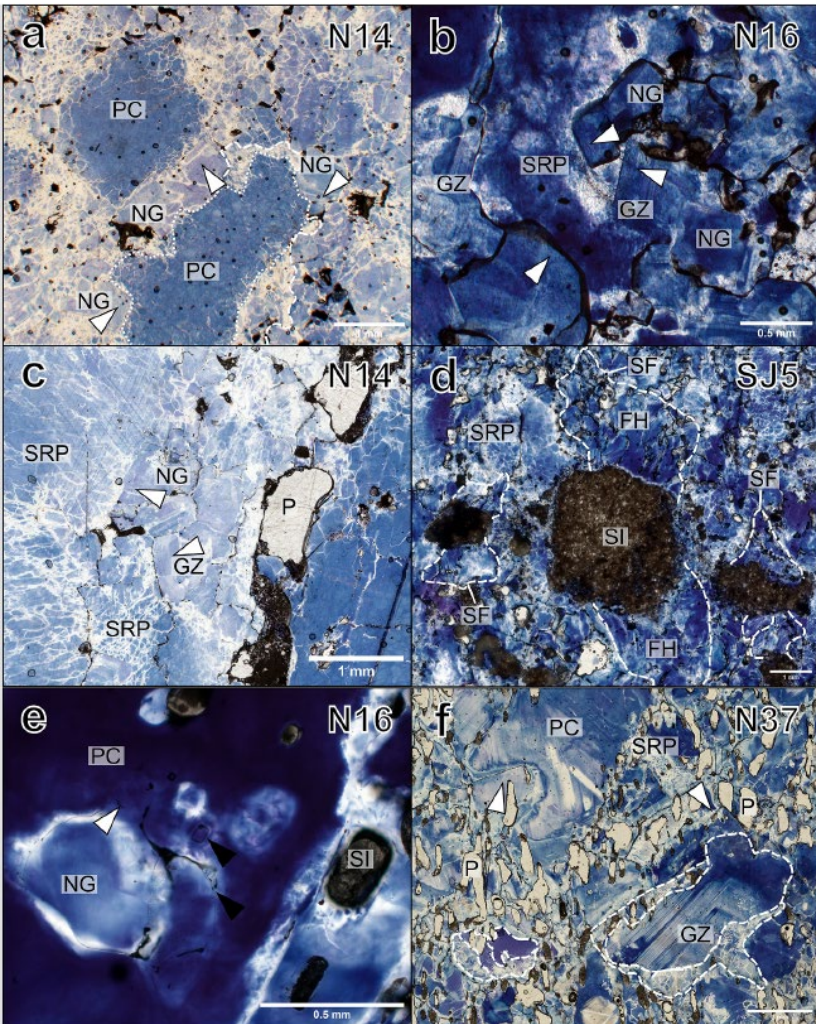
J. Barabasch et al.
 Solid Earth, 14, 271–291, 2023
<https://doi.org/10.5194/se-14-271-2023>



- ❑ **The Reality of Mixture Rheologies:** Natural rock salt is rarely homogeneous; Zechstein 2 *Kristallbrockensalz* exhibits a severe structural dichotomy, consisting of massive megacrystals embedded within a weak, fine-grained matrix.
- ❑ **Megacrystal Deformation (Dislocation Creep):** Gamma decoration proves the large megacrystals deform internally via dislocation creep, forming dense subgrain networks under remarkably low differential stresses of 0.5 to 2 MPa.
- ❑ **Matrix Deformation (Pressure Solution):** In extreme contrast, the fine-grained matrix accommodates almost all tectonic strain through fluid-assisted pressure solution, remaining entirely "substructure-free".
- ❑ **The Dual Mechanical System:** The salt mass operates as a two-part framework where strong porphyroclasts bear the plastic load, while the weak, fine-grained matrix flows rapidly around them.
- ❑ **The Flaw in Current Engineering:** Predictive models for nuclear or toxic waste repositories that simply extrapolate high-stress, laboratory power-law data are fundamentally flawed unless they incorporate these exact grain-size-dependent rheological contrasts.

Case Study 2: The Influence of Fluids and Impurities

2.4 MGy @ 63°C



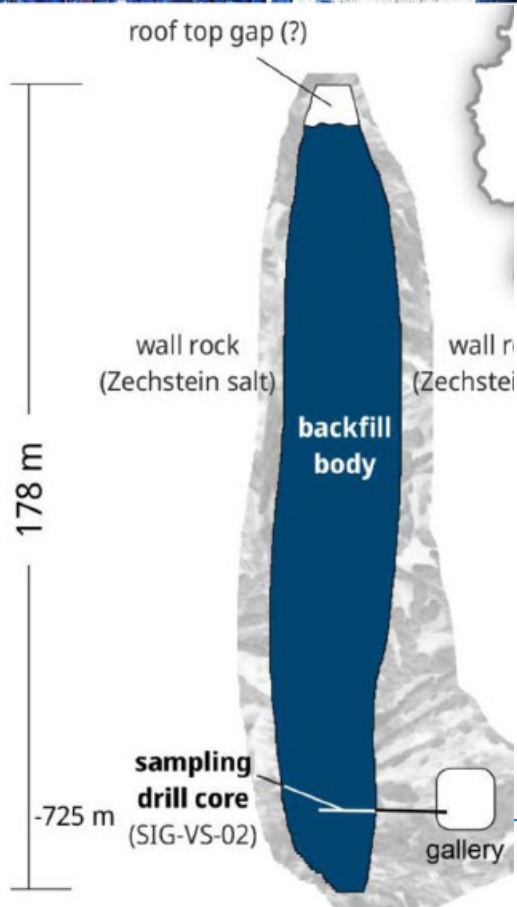
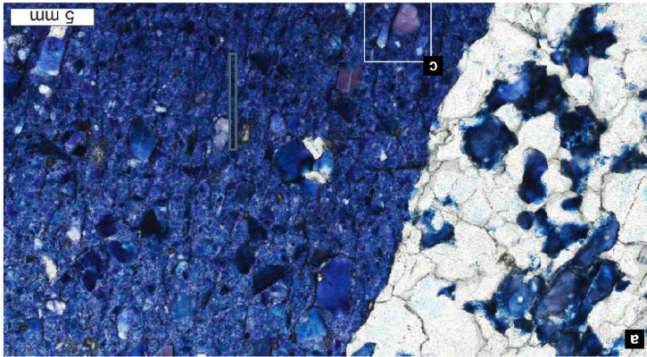
J. Schmitz et al. Journal of Structural Geology 198 (2025) 105441

- **The Extrusion Transition:** Analysis of the Kuh-e-Namak salt glacier tracks the rheological evolution from the confined diapiric stem to the subaerial extrusion, differentiating between highly translucent "clean" salt and impurity-rich "dirty" salt.

- **Domal Dislocation Creep:** Confined domal salt deforms primarily via dynamic recrystallization and dislocation creep, leaving a microstructural record of large, subgrain-rich porphyroclasts that indicate high differential stresses of 1.9 to 10.2 MPa.

- **The Fluid-Driven Mechanism Switch:** As the salt extrudes, the influx of meteoric rainwater and the pinning effect of solid impurities trigger a massive, fundamental shift toward solution-precipitation (SP) creep and grain boundary sliding.

- **Low-Stress Glacial Flow:** The highly stressed porphyroclasts are consumed and replaced by substructure-free fibrous halite, drastically lowering the rock's viscosity and enabling rapid glacial flow (10^{-10} to 10^{-8} s⁻¹) under shear stresses below 1 MPa [1].



Case Study 3: The In-Situ vs. Laboratory Discrepancy

K. Svensson et al. *J. Structural Geology* 197 (2025) 105429

2.4 MGy @ 63°C

- **The In-Situ Benchmark:** 40-year-old salt backfill from the Sigmundshall mine successfully compacted to ~1% porosity under actual, low-stress repository conditions (~2.5 MPa).
- **The Laboratory Illusion:** Standard short-term laboratory tests require artificially high stresses to force rapid compaction, which erroneously induces dislocation creep and dense subgrain networks [1].
- **The "Substructure-Free" Proof:** Gamma decoration of the in-situ backfill definitively proves the grains are completely "substructure-free," confirming that dislocation creep plays zero role in actual long-term compaction.
- **The True Mechanism & Implication:** Natural densification is driven entirely by pressure solution (evidenced by tight, interpenetrating grain boundaries); therefore, repository safety models relying on high-stress lab extrapolations are fundamentally flawed.

Validating Rheological Models through Microstructural Visibility

- **The Geomechanical Imperative:** Accurate, long-term safety forecasting for underground repositories and caverns requires precise identification of the active creep mechanisms operating under low, in-situ differential stresses.
- **The Analytical Bottleneck:** The inherent optical transparency of halite historically complicates the crucial differentiation between dislocation-driven deformation and fluid-assisted pressure solution.
- **The Visualization Solution:** Parametrically optimized gamma decoration provides stable, high-contrast, volumetric visualization of internal defect architectures and substructure-free grains.
- **The FRM II Capability:** Utilizing precise thermodynamic control and accommodating large sample volumes, we offer a robust method to empirically validate numerical models, bridging the gap between laboratory testing and geological reality.



Gamma decoration in halite - artistic power !

Using gamma decoration do not create the new, but reveal the naturally existing, within the salt mineral, usually hidden or otherwise invisible microstructures. Watching them in the microscope reveals sometimes amazing, powerful, bizarre and unexpected motifs and shapes. I tried to catch some, in my opinion most impressive, of the observations as digital image just for esthetic reasons. For this collage, salt from Germany, the Netherlands, and Italy (Sicily) was used.

