

**NEUP Final Report: An Integrated Multiscale
Experimental-Numerical Analysis on Reconsolidation of
Salt-Clay Mixture for Disposal of Heat-Generating Waste**

FINAL TECHNICAL REPORT

WaiChing Sun

March 12, 2020

DEPARTMENT OF ENERGY NUCLEAR UNIVERSITY PROGRAM

CONTRACT NO. 16-10058

Contents

1	Overview of objectives	11
1.1	Task 1: Characterization of multiscale structural and compositional hierarchy of salt-clay mixtures	11
1.2	Task 2: Multiscale discrete-to-continuum model for crystalline salt with clay additive	12
1.3	Task 3: Macroscopic Experiments	13
1.4	Task 4: Macroscopic Benchmark Comparison and Forward Predictions . .	14
1.5	List of co-PI, students, collaborators and corresponding contributions . . .	14
2	Summary of achieved accomplishments	16
2.1	Training and professional development	17
2.2	Awards and honors	17
2.3	List of publications through the performance of the award	19
3	Experimental Effort	22
3.1	One-dimensional oedometer consolidation tests	22
3.1.1	Introduction	22
3.1.2	Experimental results	22
3.2	Triaxial compression tests for salt-clay mixture	23
3.3	Long-term creep experiment	25
4	Micro-mechanics of single-crystal salt	31
4.1	Introduction	31
4.2	Governing equations	34
4.2.1	Strain energy equivalence for coupling phase field and plasticity model	35
4.2.2	Kinematics for crystal deformation of the damaged halite	35
4.2.3	Multi-phase-field approximation for anisotropic fracture	37
4.2.4	Balance of linear momentum and microforce	38
4.2.5	Energy balance equation and dissipation inequality	40
4.2.6	A specific free energy functional	42
4.3	Constitutive law	44
4.3.1	Elasticity of single-crystal halite	45
4.3.2	Single crystal plasticity	45
4.4	Variational formulation	49
4.4.1	Galerkin form	49
4.5	Operator-split solution strategies	50
4.5.1	Linearization for the staggered algorithm	51

4.6	Numerical Examples	52
4.6.1	Effect of crystal orientations	53
4.6.2	Thermal effect on anisotropic creeping	55
4.6.3	Loading rate effect	57
4.6.4	Crack propagation in a bi-crystal halite	60
4.7	Conclusions	62
.1	Stress-point simulations for model calibration	63
.2	Benchmark: multiple phase-field method for anisotropic crack propagation	65
5	Modeling size effect of clay in salt formation	68
5.1	Introduction	68
5.2	Micromorphic anisotropic MCC model	70
5.2.1	Anisotropic elasticity	71
5.2.2	Anisotropic local plasticity	73
5.2.3	Anisotropic plastic regularization for non-coaxial plastic flow	75
5.2.4	Incremental stored plastic work	76
5.2.5	Variational constitutive update with anisotropic micromorphic regularization	77
5.3	Balance and evolution equations	80
5.4	Local-global constitutive updates	81
5.4.1	Local system	81
5.4.2	Consistent tangent operator (CTO)	83
5.4.3	Spatial discretization	84
5.5	Plastic flow near and at the anisotropic critical state	84
6	Modeling crystallization of rock salt	86
6.1	Introduction	86
6.2	Conservation laws	90
6.2.1	Continuum representation	90
6.2.2	Balance of mass	91
6.2.3	Balance of linear momentum	92
6.2.4	Balance of energy	92
6.3	Effective stress and constitutive framework	93
6.3.1	Effective stress	93
6.3.2	Constitutive framework	95
6.4	Phase-field formulation for fracture	99
6.4.1	Phase-field approximation of fracture surfaces	99
6.4.2	Balance law derivation of phase-field evolution	100
6.4.3	Stored energy function	103
6.4.4	Thermodynamic restriction and crack irreversibility	104
6.4.5	Permeability evolution by phase-field fracture	104
6.4.6	Estimation of phase-field modeling parameters for geomaterials	105
6.5	Discrete formulation	106
6.5.1	Governing equations	107
6.5.2	Time discretization	108
6.5.3	Space discretization	108
6.5.4	Fully discrete form and staggered scheme for phase field	111
6.5.5	Block-partitioned preconditioner	112
6.6	Numerical examples	115

6.6.1	Drainage of freshwater in a porous column	116
6.6.2	Capillary infiltration of salt water and crystallization-induced crack- ing	117
6.7	Closure	122
7	Modeling polycrystalline rock salt	124
7.1	Introduction	124
7.1.1	Fracture and healing in rock salt: experimental evidence	125
7.1.2	Fracture and healing in rock salt: material modeling	126
7.1.3	Outlines, major contributions and notations	127
7.2	Balance laws	128
7.2.1	Definition of interface region and indicator function	128
7.2.2	Balance of mass	129
7.2.3	Balance of linear momentum and microforce	130
7.2.4	Balance of energy and dissipation inequality	131
7.3	Constitutive relations	134
7.3.1	Small strain crystal plasticity	134
7.3.2	Constitutive relation for interfacial diffusion	135
7.3.3	Constitutive relations for anisotropic phase field and crack healing	137
7.4	Numerical aspect on FFT-based method	138
7.5	Examples	140
7.5.1	Simulation setup and material model calibrations	140
7.5.2	Long-term creeping due to solution precipitation	142
7.5.3	Anisotropic cracking	144
7.5.4	Chemical-diffusion-controlled crack healing	145
7.6	Conclusions	148
8	Three-dimensional benchmark simulations	149
8.1	Numerical treatment	150
8.2	Numerical Benchmarks	150
9	Final Quad Chart	154

List of Figures

3.1	(a) Consistency of consolidation testing of crushed Avery Island salt-water-bentonite clay mixtures. All cases use a mixture of 3% water, 5% clay, and 92% salt by weight. (b) Consolidation testing of three different compositions.	24
3.2	Effects of (a) clay content, (b) water content, and (c) consolidation rate on the consolidation of crushed Avery Island salt with different percentages of water and bentonite clay. For testing, the reference case is 3% water and 5% clay content (by weight).	24
3.3	Consolidated AI salt sample with water (3% wt) and clay (5% wt) contents.	25
3.4	MicroCT horizontal slice images of four different samples taken around the central region. (a) before consolidation testing and (b-d) after consolidation testing with (b) salt only, (c) 3% water content and no clay, and (d) 3% water and 5% clay contents.	26
3.5	MicroCT Vertical images of three different samples taken around the central region after consolidation testing. (a) salt only, (b) 3% water content and no clay, and (c) 3% water and 5% clay contents.	27
3.6	Experimental setup for triaxial strength tests of the cored salt samples using two Isco pumps and a thermal unit.	28
3.7	Triaxial strength test results for salt samples at 20°C at confining pressures of 0.5 MPa, 5 MPa, and 10 MPa at a strain rate $\dot{\epsilon} = 10^{-5} \text{s}^{-1}$.	28
3.8	Salt samples before (top) and after testing (bottom) at 20°C for confining pressures of 0.5 MPa (a, b), 5 MPa (c, d), and 10 MPa (e, f).	29
3.9	Experimental setup for (a) a small sample (1 inch diameter) using two Isco pumps and thermal unit and (b) a large sample (≈ 4 inches diameter) with a lead jacket with thermal controlling unit.	29
3.10	MicroCT images of Avery Island salt sample with 3% water and 5% clay addition after creep testing. (a) 3D view of the sample and (b-c) vertical and horizontal views along the center of the sample.	30
3.11	Strain evolution for the creep testing of an 1-inch diameter salt sample (water 3% - clay 5%) at 60°C. The sample was initially under isotropic pressure of 20 MPa and was then subjected to differential stress of 10 MPa.	30
4.1	Disposal operations for Transuranic (TRU) waste at the Waste Isolation Pilot Plant (WIPP). (Reproduced from Hansen and Leigh [2011] .)	31
4.2	The description of the slip-system of a single-crystal halite, (a) an aggregate of many atoms for the face centered cubic crystal structure (reproduced from Callister Jr and Rethwisch [2012] – adapted from the original Moffat et al. [1964] , (b) a unit cell for the sodium chloride (NaCl) crystal structure (reproduced from Callister Jr and Rethwisch [2012]), and (c) the slip-system $\{110\}\langle 1\bar{1}0 \rangle$.	36

4.3	Analysis of material anisotropy by changing the orientation of the slip system, (a) the set-up for plane strain compression test, (b) the definition of Euler angle (ϕ , θ) in the three-dimensional plane (reproduced from Borja [2013]), including the slip system of a single crystal halite (see each slip system in Fig. 4.2).	54
4.4	Stress-strain curve of a single-crystal halite with different orientation of the slip-system (the Euler angle θ is fixed 0° while changing $\phi = 0^\circ$, 10° , and 30° , respectively).	55
4.5	Phase-field values (top) and plastic slip results patterns (bottom) with different orientations of the slip-system to investigate the material anisotropy under plain strain compression test condition. The patterns are captured at the last numerical step of each case. While fixing the Euler angle $\theta = 0^\circ$, ϕ varies 0 , 10 , and 30°	56
4.6	Analysis of temperature effect on mechanical responses, (a) the set-up for plane strain compression test, (b) stress-strain curves for each temperature condition ($T = 25^\circ\text{C}$, 50°C , and 90°C , respectively).	57
4.7	Phase-field values (top) and plastic slip results (bottom) patterns with different initial temperature conditions at the same vertical strain, 0.23% . The constant displacement loading is applied at the top, and the Euler angle of each temperature condition is fixed $\theta = 0^\circ$ and $\phi = 70^\circ$	58
4.8	The conceptual boundary value problem set-up for evaluating thermal impact on a single crystal halite, (a) a photo of room B (captured from Reedlunn [2016] , Munson et al. [1990]), (b) the sections of rooms in WIPP (reproduced from Reedlunn [2016]), and (c) the problem set-up. The dimension of a room section (L_v and L_h) varies on each room in the WIPP. The top, bottom, and right surfaces are insulated, and the initial temperature T_0 is set to 25°C . The whole domain temperature is increased to 100°C by applying constant heat flux q . The Euler angle is set to	59
4.9	The resultant patterns of (a) phase field for damages, (b) the plastic lip, and (c) the x direction displacement (scaled by $\times 10$) are illustrated when the overall temperature domain is increased up to 100°C . The ranges of each result are selected for visualization purpose.	59
4.10	Analysis of the loading rate effect under two-dimensional tension test, (a) the set-up for boundary value problem, (b) stress-strain curve with different loading rates ($\dot{\epsilon} = 1.0\text{e}^{-7}/\text{sec.}$). The initial temperature T_0 is set to 25°C , and the Euler angle is set to $\theta = 0^\circ$ and $\phi = 0^\circ$	60
4.11	Phase-field values (top) and plastic slip results (bottom) patterns with different loading rates are illustrated. The results are captured at the last numerical step of each simulation. The loading rate $\dot{\epsilon} = 1.0\text{e}^{-7}/\text{sec.}$	61
4.12	The numerical set-up for a bicrystal is depicted to analyze the impact of the grain boundary fracture energy on crack propagations (reproduced from Oshima et al. [2014]). The Euler angles ϕ are set to -15° and 15° , respectively to left and right side grains. The single cleavage plane is assumed to the grain boundary with the direction of 45° along the diagonal. The initial temperature is set to 25°C , and the fracture energies of the grain boundary is adopted 86% , 75% , and 50% of the grain fracture energy, respectively. . .	62

4.13	Phase-field values (left) and plastic slip results (right) patterns with different fracture energies of the grain boundary are illustrated. Case 1 (a) and (b) assumes the fracture energy of the grain boundary with 86% of grain fracture energy; Case 2 (c) and (d) with 75%; Case 3 (e) and (f) with 50% of the grain fracture energy, respectively.	63
14	Comparison of stress-strain curves from the experiment [Carter and Heard, 1970] and current study under different temperature (25°C, 100°, 200°, and 300°) and strain rate conditions, (a) with the strain rate 1.0×10^{-4} /sec, (b) the strain rate 1.0×10^{-7} /sec.	65
15	Comparison of stress-strain curves from the experiment [Carter and Heard, 1970] and current study under different temperature (25°C and 100°) and strain rate conditions, (a) with the strain rate 1.0×10^{-4} /sec, (b) the strain rate 1.0×10^{-7} /sec.	66
16	The concept of multiple phase-field method for anisotropic crack propagation, (a) test set-up for guided crack propagation (the top and bottom parts of the domain outside the guided line have higher fracture toughness not to initiate cracks, (b) the combined crack phase field using the equation suggested by Nguyen et al. [2017a,b], (c) the crack propagation of phase-field 1, (d) the crack propagation of phase-field 2.	67
5.1	Rock material heterogeneity at the micrometer scale, showing two different materials of significant engineering interest, both characterized by size-dependence of the anisotropy.	69
5.2	Schematic of mesoscale and microscale-homogenized material effective media, where l is the microstructural direction whereas $l_{\tilde{\alpha}}$ is the direction characterizing the anisotropic micromorphic regularization of a plastic internal variable α , via the projected internal variable $\tilde{\alpha}$, governed by energy functionals as detailed in Section 5.2.3, with inhomogeneous microscale identification and constituent categorization after Bennett et al. [2015]; q/f/p means inorganic quartz, feldspar, or pyrite, and o/c matrix indicates organic and clay matrix.	72
5.3	Geometric interpretation of the mapped two-invariant yield criterion, Eq. (5.30): (a) introducing M as the slope of the critical state line (CSL) in the starred scalar space, after Ortiz and Pandolfi [2004], and with softening and hardening trends after Borja [2013]; and, (b) unmapped two-invariant yield criterion parameterized by the mapping coefficient, varying $\beta^{\text{dev}} = \beta^{\text{vol}}$ at fixed $\alpha^{\text{dev}} = \alpha^{\text{vol}} = \gamma^{\text{dev}} = \gamma^{\text{vol}} = 1$, after Semnani et al. [2016].	80
6.1	Example of damage in building stones by crystallization of salts. (Photograph by Suzanne MacLeod, distributed under a CC-BY 2.0 license.) . . .	87
6.2	Schematic illustration of a crystal inside a pore. Note that the crystal is confined within the liquid solution, not being in contact with the solid matrix. Similar illustrations have been presented in Scherer [2004], Steiger [2005a], Derluyn et al. [2014].	88
6.3	Elementary volume representation of a four-phase mixture composed of the solid matrix, liquid solution, gas, and crystals. The liquid solution is also a sub-mixture of water and dissolved minerals.	91
6.4	An example mesh illustrating the locations of (linear) finite element and finite volume degrees of freedom.	109

6.5	Schematic illustration of the column drainage example that simulates Liakopoulos' experiment Liakopoulos [1964] . Initially, the domain is fully saturated with zero pore pressure ($p_0 = 0$). Once the simulation begins, the pore fluid is drained through the bottom outlet filter by gravitational force.	117
6.6	Drainage simulation results of the current code using finite elements for the solid deformation problem and finite volume for the fluid flow problem (FEM + FVM), and an unsaturated poromechanics code using finite elements for both the solid deformation and fluid flow problems (FEM + FEM). The numerical results are compared with experimental data of Liakopoulos Liakopoulos [1964] . (a) Pore pressure variation along the height (numbers inside the figure denote elapsed time in minutes). (b) Fluid velocity at the bottom outlet filter.	118
6.7	Schematic illustration of the capillary rise example. The domain is initially filled with freshwater and air such that they manifest a capillary pressure of 20 kPa. The simulation begins by prescribing the pressure and mass fraction at the bottom boundary as 1 kPa and c_{eq} , respectively. The lower half of the boundaries are no flux boundaries, whereas the upper half domain is subject to a constant liquid flux of -2×10^{-5} kg/m ² /s. Mineral flux $\hat{j}$ is zero throughout the boundaries.	119
6.8	Evolution of the supersaturation ratio U until 2760 minutes.	120
6.9	Evolution of the crystal saturation S^c from 2760 minutes to 4092 minutes. Color bar in log scale.	120
6.10	Evolution of the crystallization pressure p_{cr} from 2760 minutes to 4092 minutes. Color bar in MPa.	121
6.11	Evolution of the phase-field variable d from 4092.0 minutes to 4092.3 minutes.	121
7.1	Microstructure of rock salt with pores and intergranular brine: (a) SEM image [Desbois et al., 2012c] ; (b) schematic illustration.	126
7.2	A schematic representation of (a) A solid body Ω with crack discontinuity Γ_c and grain boundary discontinuity Γ_{GB} ; (b) A regularized grain boundary region Ω_{GB} by grain boundary indicator d_{GB} ; (c) A regularized crack region Ω_c by fracture phase field d_c ; (d) A regularized interfacial region $\Omega_i = \Omega_{GB} \cup \Omega_c$ by interfacial indicator d_i	129
7.3	Effectiveness of using equation (7.38) to detect the crack tip region. The crack region detection criteria takes the value $l_0^3 \ (\nabla \otimes \nabla d_c - \nabla^2 d_c \mathbf{I}) \cdot \nabla d_c\ $. (a) Initial phase field distribution of curved crack using high-order phase-field model; (b) Crack tip determination criteria of curved crack; (c) Initial phase field distribution of intersecting crack using high-order phase-field model; (d) Crack tip determination criteria of intersecting crack.	137
7.4	Confined compression stress-strain response with different strain rates at room temperature [Carter and Heard, 1970]	141
7.5	Analysis of creep loading effect on solution-precipitation creep rate: (a) The numerical set-up for the creep simulation; (b) Creep rate evolution for each creep loading condition.	143

7.6	Brine concentration and strain distribution at different creep stages for the $\sigma = 1$ MPa simulation in Figure 7.5: (a-c) Evolution of grain boundary brine concentration [unit: mol m^{-3}]; (d-f) Evolution of strain in the loading direction ε_{xx} ; (g-i) Evolution of strain in the transverse direction ε_{yy}	144
7.7	Analysis of crack initiation and propagation in polycrystalline rock salt: (a) Initial configuration of polycrystal RVE and grain boundary. The RVE edge length L is 1 mm, and the grain boundary thickness is $l = 0.014L$, the initial orientation is random with $[001]$ axis perpendicular to the xy plane. (b) Homogenized stress-strain curve.	145
7.8	Phase field and Von Mises stress distribution within the polycrystal specimen at different load stages: (a-c) Phase field; (d-f) Von Mises stress ([unit: MPa]).	146
7.9	Strain path and stress strain curve of the crack healing simulation. (a) Strain path of the loading-unloading-reloading process. (b) Uniaxial stress-strain curve during the loading-unloading-reloading process. The RVE holds for a while with no external loading between the unloading process and the reloading process to provide enough time for crack healing.	147
7.10	Phase field evolution during the loading-unloading-reloading process. (a) Initial phase-field representing the initial circular flaw with 0.1 mm radius; (b) Fracture phase-field distribution after the loading and unloading process; (c) Crack healing after the stress-free holding process; (d) Crack propagation continues to penerate the specimen after the reloading process.	147
7.11	Chemical concentration distribution during the loading-unloading-reloading process. (a) Homogeneous initial chemical concentration equals to the equilibrium concentration c_{eq} at stress free status; (b) Chemical concentration after the loading and unloading process corresponding to Figure 7.10 (b); (c) Chemical concentration distribution after the stress-free holding process corresponding to Figure 7.10 (c); (d) Chemical concentration after the reloading process corresponding to Figure 7.10 (d).	148
8.1	Planar view of the 3-D computational domain.	150
8.2	Temperature history at different observation points: P1 = (0, 1.17, 1.4)m; P2 = (0, 0.59, 0.57)m; P3 = (0, 10, 1.42)m.	151
8.3	Temperature field in near-alcove region after 0.125, 2, 25 and 50 years.	152
8.4	Temperature field across the domain after 0.125, 2, 25 and 50 years.	153
9.1	Final Quad Chart.	154

Abstract

The possibility of permanent disposal of nuclear waste in salt has been investigated for decades. This interest is primarily due to the availability of the stable salt formations and the desirable thermal-hydro-mechanical behaviors of salt (e.g. high thermal conductivity, low permeability, self-healing). To ensure the long-term safety of the disposal, sealing is one of the primary concerns [Hansen and Leigh, 2011]. One promising approach is to reconsolidating the crushed salt readily available in the repository with clay (Bentonite) additive [Hansen et al., 2015]. This treatment is favorable, because clay exhibits low permeability and sorbs radionuclides and water during the early stage of the repository [Hansen and Leigh, 2011]. While reconsolidated salt with clay additive may act as an excellent flow barrier for the sealing system, loss of tightness of salt may occur due to the creation of connected pathways of the grain boundaries of salt and the deviatoric-stress-induced crack growth across and along grain boundaries of salt. This microscale deformation mechanisms play a significant role in the integrity of the repository and the sealing system in rock salt formations, while the grain boundary healing process could prevent active deformation. However, deformation mechanisms at micro-scale and their linkage to the bulk behaviors of salt with additives (i.e., clay and moisture) are still not fully understood. The difficulty of predicting the influence of moisture content and clay additives is particularly due to the lack of the essential physical underpinnings to replicate the clay-salt interaction at the grain scale. Furthermore, a lack of insight on the distribution of fluids and its role in salt-clay consolidation processes makes it difficult to optimize clay and moisture additives for enhancing reconsolidation under various operational conditions and how to incorporate the microscopic interaction of salt and clay into macroscopic predictions.

Reconsolidation of granular (e.g., crushed) salt is one of the most critical processes crucial to plugging, sealing, testing, and modeling for sealing a repository [Hansen et al., 2014a]. Granular salt is often used as buffer or backfill material where void space created by excavation is reduced over time. The combination of high thermal conductivity and low permeability, the self-healing properties, as well as the fact that crushed salt is readily available in a repository, all make the re-consolidation of crushed salt an attractive option for the salt repository environment. While it is now well-known that elevating temperature may accelerate reconsolidation of the crushed salt, recent experimental work (e.g. Hansen et al. [2014a], Broome et al. [2014]) has pointed to a new promising approach in which a small amount of moisture and clay additives may be sufficient to alter the coupled TMHC mechanism to meet the performance criteria for backfills. Hence, understanding the reconsolidation process of salt with moisture and clay additives is instrumental for designing high-performance backfilling or sealing nuclear waste repositories in salt. This work provides comprehensive experimental data under a variety of conditions (e.g., temperature, stress, moisture contents) expected for waste disposal sites, which allows us to improve the viability of disposal concepts with a salt-clay

mixture.

The overall purpose of this research is to improve understanding of THMC coupling effect on the reconsolidation of granular (or crushed) salt-clay mixture used for seal systems of shafts and drifts in salt repositories. This proposed work is partially motivated by the recent work on the Waste Isolation Pilot Plant (WIPP) that shows the promising sealing capability of clay-salt mixture compared to crushed salt (e.g., [Hansen et al. \[2015\]](#)). In particular, primary emphasis is to develop a fully integrated multiscale experiment-numerical study to determine and explain what leads to the superior sealing ability of the clay-salt mixture. These research activities are designed to seek further understanding of (1) why clay additives may enhance the fluid trapping and (2) whether this flow barrier effect may prevail under different combinations of temperature, confining pressure, deviatoric stress and other foreseeable environmental factors. If successful, this enhanced flow trapping ability of the seal provides significant improvement to the seal and repository performance and therefore make the repository safer in the long-term. The experiment component includes microstructural investigation and macroscopic tests on a reconsolidated salt-clay mixture. In the microstructural study, the goal is to (1) characterize microscopic distributions of distinct phases (e.g., clay, salt crystal boundaries, trapped brine, and pore) to examine the connectivity of the pore network inside the salt-clay mixture with different amounts of clay additive and moisture content and (2) analyze multiscale imaging data to reconstruct the polycrystalline microstructures for numerical simulations. Meanwhile, macroscopic tests are performed to analyze how clay alters the failure/creep mechanisms in the salt-clay mixture. Microscopic and macroscopic experimental observations will both be used to calibrate and validate a multiscale model that explicitly simulates the capillary and multiphase flow in the connected pores and the deformation due to the presence of intra-crystalline brine at the pore scale via a new polyhedral discrete element–lattice Boltzmann method (DEM-LBM) coupling model. The pore-scale simulations are homogenized via an upscaling procedure that converts pore-scale information (e.g. force exerted on grain boundary, sliding, pressure-solution) to continuum measures (e.g. Cauchy stress, Darcy’s flow) at each integration point in the macroscopic multiphase TMHC model. This multiscale scheme will allow coupling between high-fidelity simulations of brine-salt-clay interaction and the macroscopic TMHC model. The multiscale model helps the understanding of how the trapped brine inclusion affects the pressure-solution mechanism with the presence of clay and moisture. This work brings new insight into the sealing capacity of salt-clay mixture under elevated temperature over a long period of time — a key to evaluating the potential of salt-clay mixture usage for salt repositories.

Chapter 1

Overview of objectives

The objective of this research project is to further the understanding how the micromechanics of salt-clay-fluid interaction affects the macroscopic responses of salt. As such, the research plan integrated advanced modeling work with laboratory tests and imaging technologies undertaken by Columbia University and Sandia National Laboratories. This project consists of four major tasks, i.e.,

1. Characterization of multiscale structural and compositional hierarchy of salt-clay mixtures
2. Multiscale discrete-to-continuum model for crystalline salt with clay additive
3. Macroscopic Experiments
4. Macroscopic benchmark studies

1.1 Task 1: Characterization of multiscale structural and compositional hierarchy of salt-clay mixtures

We used crushed salt samples (Salado Formation) collected from the Waste Isolation Pilot Plant (WIPP) site. These samples are selected because of the ease of in-situ sample collection and relatively well-characterized properties (e.g., Caporuscio et al., 2013; Broome et al., 2014). We tested two bentonites including sodium-bentonite as clay sources and two different moistures including distilled water and salt solution. All experimental conditions were based on the previous works at WIPP (e.g., Daeman and Ran, 1996; Caporuscio et al., 2013). To select suitable material mixtures for installation properties (e.g., density and saturation), hydrological and swelling pressure were measured for various mixtures of salt, clay, and moisture content. A set of salt-clay mixtures with different moisture contents were selected based on preliminary measurements for reconsolidation experimental work with cores of 1"-2" diameter. For a few representative mixtures, the mineral composition were measured with crushed samples using an X-ray diffraction (XRD) method and porosity and pore size distribution were determined with a mercury intrusion approach. A series of thin sections (2-3 cm scale) impregnated with fluorochromes were examined using techniques of epi-fluorescence and laser scanning confocal microscopy (LSCM) to delineate compositional distributions with multiple filters (e.g., UV, fluorescence filters) as shown in Figure 2. Quantitative analysis of fluorescent and confocal microscopy at 1 micron resolution is compared with XRD analysis for bulk mineral composition and

energy-dispersive spectroscopy (EDS)/QEMSCAN analysis of mineralogical distribution (typically, 2x3 mm² area at 5 micron resolution). High magnification SEM images (500x to 30,000x) provided qualitative or semi-quantitative measures of chemical compositions (using EDS), crystalline structure, and crystal orientations using electron backscatter diffraction (EBSD). MicroCT images were used to investigate deformation mechanisms and constituent distribution in 3D in both the unconsolidated state and after the mine run salt had been consolidated. Based on thin section and micro-CT analysis, FIB-SEM images were used to characterize phase distribution over 10 to 100 micron scales in 3D at 10 to 100nm resolutions, respectively. For 3D digital reconstruction from nano- to core scales, advanced multiscale image analysis were applied to identify several key patterns of pore structure and compositional distribution. This multiscale imaging of microstructures and deformation experiments (e.g., Desbois et al. 2012a,b) provide the microscopic evidence to analyze how clay additive affects the pore network and reduces the effective porosity and templates for the micro-mechanical crystal models.

1.2 Task 2: Multiscale discrete-to-continuum model for crystalline salt with clay additive

The creeping response of salt rock and reconsolidated salt are consequences of dislocation (short-term) and fluid-assisted diffusional transfer deformation (long-term) occurring at grain boundaries [Olivella and Gens, 2002]. Conventionally, these creeping behaviors are taken into account indirectly by coupling plasticity models with additional creeping laws suitable for a crystallographic cubic system [Spiers et al., 1990]. For instance, Hampel [2012] takes a different approach by treating the rock salt as a composite material consisting of hard (representing the grain boundaries) and soft regions (representing subgrain interiors) and propose constitutive laws based on composite theory. This phenomenological approach is powerful when one can sufficiently interpret the physical mechanism and converts them into proper evolution laws governed by internal variables. However, such an approach requires very detailed interpretations whenever new factors, such as pressure solution and moisture content and clay additive, are taken into account. A more direct and intuitive way that can potentially overcome this obstacle to model the dislocation, grain boundary diffusion and solution precipitation is at the grain-scale level. This approach is particularly useful for capturing the poroelastic responses due to the presence of brine in the intergranular pore space and the influence of humidity on creep rate (Joffe effect). The microstructural simulations can be either standalone or they can be linked, homogenized or up-scaled to the field-scale simulations as a replacement of the classical constitutive law. In the latter case, the interplay between the microscopic responses of representative elementary volume and macroscopic field-scale simulations is particular useful to understand how microstructural attributes changes in a large-scale operation.

The multiscale coupling is established by replacing the constitutive laws with grain-scale THMC simulations conducted on representative elementary volumes of salt-clay mixture. At the pore-scale level, sub-scale simulation capacities have been established based on the existing code developed by the PI. The polycrystalline structures are explicitly captured via a FFT solver where the grain boundary interaction and sub-grain fractures and damage are replicated numerically [Na and Sun, 2018, Ma and Sun, 2020]. Meanwhile, the dissolution/precipitation/migration of salt taking place through the liquid phase when brine is presented in the pore space is captured by a multi-permeability model. To

simplify the formulation, we assume that the salt behaves as a single ion such that the couples of Na^+ and Cl^- dissolve and precipitate jointly. Mineral reactions among the ions are treated kinetically through boundary conditions along the grain boundaries in the lattice Boltzmann step [Yoon et al., 2012]. If necessary, more sophisticated geochemical code (e.g., Phreeqc) can be easily implemented (e.g., Yoon et al. [2015]). The coupling of the DEM and LBM solvers has been established via the macroscopic finite element. This treatment is not only beneficial for analyzing the creeping mechanism, but also provides the essential physical underpinning to model the salt healing processes that are due to closure of micro-fractures and bonding of fractured surface due to crystal plasticity and pressure solution.

This dual-scale micropolar discrete-continuum coupling model are formulated to explicitly simulate the interactions among trapped brine, salt crystal and grain boundaries and connect these pore-scale mechanisms into the macroscopic finite element model at finite strain. The model will capture the brittle-ductile transition of salt-clay additive under different confining pressures and temperatures. 3D numerical simulations will provide key insight on how the presence of clay affects the pressure solution and brittle responses. Notice that recent research has shown that naïve hierarchical multiscale coupling schemes may lead to the loss of physical length scale and create mesh-biased solutions [Wang and Sun, 2019]. To resolve this problem, the research team will extend a higher-order discrete-to-continuum multiscale solver for multiphysical problems. The modeling efforts in return will serve as the key for designing the specimen-level triaxial creep tests. One key upshot of the proposed multiscale coupling method does not require any macroscopic phenomenological constitutive law [Liu et al., 2016a, Wang and Sun, 2016a, Bryant and Sun, 2019]. Instead, the simulated macroscopic responses of the macroscopic finite element are due to the collective motion of the halite crystal, moisture content and clay additive. As a result, the multiscale simulations open doors to testing different hypotheses that explain the acceleration of consolidation of backfill materials and explain how thermal conductivity changes due to porosity. This valuable information helps reduce the number of laboratory tests necessary to assess the performance of the backfill materials.

1.3 Task 3: Macroscopic Experiments

In order to provide an experimental basis for numerical validation and verification a suite of experimental tests have been employed. First, a series of preliminary oedometric compression tests are performed with samples consisting of a wide variety of salt-clay-moisture contents as described in Task 1. Tests were performed at temperatures ranging from room temperature to temperatures above in-situ conditions. Observations of consolidation, permeability changes, and static recrystallization (i.e., annealing times from minutes to year) [Kröhn et al., 2012, Desbois et al., 2012a, Bauer et al., 2015] combined with auxiliary meso-scale simulations conducted in Task 2 were used to down select salt-clay-moisture mixtures for full triaxial and creep testings. This is necessary because while it is well known that moisture and clay content can accelerate salt consolidation, the mechanism, and rate of compaction at very low porosity is not fully understood [Hansen et al., 2015]. Full triaxial testing consisted of multiple stress paths, including hydrostatic, traditional triaxial compression, constant mean stress, and if necessary extension. These stress paths are necessary to not only fully characterize the material mechanically but also to investigate how intra-granular fluid inclusions affect the pressure solution in

the presence of clay under different states of stress. Fluid inclusions were investigated through microstructural observations as described in Task 1. Creep testings were used to both determine basic creep parameters for the salt-clay-moisture mixtures, as well as characterize the fluid assisted diffusional trans creep and the dislocation creep commonly observed in crushed salt over time. Damage induced permeability and thermal conductivity changes predicted by the numerical model were compared against this data to examine the threshold for percolation flow along the grain boundaries.

1.4 Task 4: Macroscopic Benchmark Comparison and Forward Predictions

The proposed multiscale models have been calibrated using a optimization code Dakota using a combination of experimental data sets and published results. The multiscale analysis focuses on the early period after hot waste is placed in the salt. This stage is critical for the seal system as the steep thermal gradient will cause the migrating in grain boundaries and micro-fractures move down the stress gradient toward the repository ventilation. Hence moisture will be driven away from the salt [Hansen and Leigh, 2011]. The multiscale model provides insights on how clay affects this thermos-hydro-mechanical coupling mechanism and the help assessing the safety of the repository with the salt-clay backfill when brine intrusion occurs. While the rate dependence due to dislocation creep can be captured in experiments during the lifespan of the physical tests, the long-term effects, such as the decay of the waste or the fluid-assisted creeps, may take in decade- and century-scales.

1.5 List of co-PI, students, collaborators and corresponding contributions

The complete list of the co-PI, students, collaborators, and corresponding contributions are listed below:

- PhD students
 - SeonHong Na, duration = 36 months, major contributions: completing the first coupled multi-phase-field damage/crystal plasticity model for salt.
 - Kun Wang, duration = 24 months, major contributions: establishing multiscale LBM-DEM-FEM model using machine learning approach.
 - Eric Bryant, duration = 24 months, major contributions: 1) establishing mixed-mode model for modeling anisotropic fracture and a micromorphic regularized Cam-clay plasticity to model clay inclusion inside salt.
 - Nikolas Vlassis, duration = 36 months, major contributions: Analyzing microstructures of salt from micro-CT imaging, conducting all experiments at Sandia National Laboratories and provide documentations of all the experimental results.
 - Hyoungh Suk Suh, duration = 3 months, major contributions: Introducing micropolar effect on the phase field fracture model for salt and other anisotropic materials.

- Mils Melissa (University of New Mexico), contributions: helping Hongkyu Yoon analyzing data and conducting literature reviews.
- Associate research scientist and postdoctoral research scholars
 - Jinhyun Choo, duration = 12 months, major contributions: modeling brittle-ductile transition of rock salt and the crystal growth and recrystallization induced damage in crystalline rocks.
 - Yousef Heider, duration = 24 months, major contributions: modeling moisture effect in rock salt and the drying and wetting induced fractures in geological materials.
 - Ran Ma, duration = 15 months, major contributions: modeling polycrystal using FFT- solvers and introducing healing models for single- and polycrystal salts.
 - Chuanqi Liu, duration 12 months, major contributions: introducing of material point models to enable direct image-to-simulation workflow.
- Co-PIs
 - WaiChing Sun (Columbia), contributions: mentoring students, postdoc and visiting scholars, deriving and validating theoretical models and documenting results in published papers, and quarterly reports.
 - Hongkyu Yoon (Sandia National Laboratories), contributions: mentoring Nikolaos Vlassis during the summer, analyzing microstructures and images, supervising summer research students.
 - Mathew Ingraham (Sandia National Laboratories), contributions: providing high-level advice during sample preparation in Year 2.

Chapter 2

Summary of achieved accomplishments

We have introduced a new meso-scale model that can explicitly capture the precipitation creeping, heat transfer, crack and healing via phase field. This is an important milestone, because the previous salt models either did not incorporate such a comprehensive features, or have to capture these effects through phenomenology. Nevertheless, the advancement of higher-order phase field modeling provides us a new way to explicitly model these coupling effects occurred in the bulk and ground boundaries. This treatment is more robust than the phenomenological approach, as the multi-physical coupling effect may make it easy to have over-fitting that is difficult to detect. To overcome the computational barrier, we have introduced a way to use deep neural network to upscale meso-scale models to the macroscopic scale. Since the surrogate model is trained using the meso-scale simulation data, the resultant multiscale model does not require resolving RVE responses at each incremental step. This upshot is important for simulate geomechanics problems at field scale where the computational efficiency is important.

From the experimental side, we have conducted various one-dimensional consolidation tests, triaxial compression tests and creep tests performed on specimen with different amount of clay content. Our work also encompasses strain localization in rocks and soils due to plastic deformation. A non-regularized plasticity model can exhibit instability upon the formation of strain localization, due to the ill-posedness of the boundary value problem. Evolved shear-band thicknesses may erroneously depend on the finite element size. Embedding a strong discontinuity upon onset of shear band can resolve the ill-posedness issue. However, this approach is not feasible to predict heterogeneous materials, where multiple initial shear band patterns may evolve into one persistent shear band. Furthermore, the spatial heterogeneity of many geological materials are often of a multiscale nature. While it is well known that this multiscale nature leads to different shear strengths at different length scales, the anisotropy of the shear strength may also vary according to the specimen size. To resolve these three issues, we have formulated a non-coaxial version of Cam-clay plasticity model anisotropically regularized by Helmholtz equations. While classical gradient plasticity model introduce gradient dependence on the plastic flow to regularize constitutive laws isotropically, our approach introduce an anisotropic regularization. The plastic flow is not only gradient-dependent but can be non-coaxial to the normal direction of the yield function. This remedy proposed is not only helpful for simulating shear band but has an important implication to the plasticity theory.

2.1 Training and professional development

This project provided the essential training opportunities for 8 young talented researchers to develop their research skills. Through the support, graduate students are developing new technical skills while refining their presentation and social skills in a close-knit research group. The PI met the students and postdoc on a daily basis and provided mentorships to them. During the summer, selected students are chosen to collaborate with research scientists at Sandia National Laboratories to broaden their research experience. GRA Eric Bryant has visited Lawrence Livermore National Laboratory during the summer; GRA Nikolas Vlassis has worked at Sandia National Laboratories for 3 years for this project and former PhD student Kun Wang has joined Los Alamos National Laboratory as postdoc (in the T-3 group). In summary, the support of the DOE NEUP project contributed to the thesis writing of the following current and former students:

- SeonHong Na. Multiscale thermo-hydro-mechanical-chemical coupling effects for fluid-infiltrating crystalline solids and geomaterials: theory, implementation, and validation. Fall 2014-Fall 2018. Now assistant professor of Civil and Environmental Engineering.
- Kun Wang. From multiscale modeling to meta-modeling of poromechanics problems. Spring 2015 to Spring 2019. Now postdoctoral research scientist, Theoretical Division of Los Alamos National Laboratory since September 15th, 2019.
- Eric Bryant. Capturing evolving size-dependent anisotropy from brittle fracture to plasticity for geological materials, Fall 2016-Spring 2020 (proposal defended on 1/22/2020; will join Theoretical Division of Los Alamos National Laboratory as postdoctoral research scientist in summer 2020).
- Nikolas Vlassis, Geometric deep learning from multi-graphs and manifolds for computational poromechanics, pre-qualifying exam.

Meanwhile, the postdoctoral research scientists and associated research scientists supported by the NEUP program are in high-demand and have found tenure-track and research scientist positions as listed below:

- Yousef Heider, PhD (Institute of Continuum Mechanics, University of Stuttgart, Germany), High-strain-rate responses of geomaterials, Fall 2018-Spring 2019. Now lecturer and research group leader at RWTH Aachen University (Germany).
- Jinhyun Choo, PhD (Stanford University), joined McMaster University as an assistant professor in January 2018) Phase field modeling of crystal growth induced damage in porous media and salt, Fall 2016-Spring 2018. Now assistant professor at Hong Kong University.
- Ran Ma, PhD (University of Tennessee), Fast Fourier Transform Solver for polycrystal plasticity and fractures of reconsolidated salt, Spring 2019-current. (One on-site interview at the University of Mexico).

2.2 Awards and honors

- Selected individual awards received by the PI

- **UPS Foundation Visiting Professor of Civil and Environmental Engineering**, Stanford University, 2020. The support will provide the opportunity of the PI to visit the Department of Civil and Environmental Engineering from Fall 2020 to Spring 2021.
- **John Argyris Award for Young Scientists**, the International Association for Computational Mechanics, 2020. The IACM recognizes outstanding accomplishments, particularly outstanding published papers, by researchers 40 or younger. Eligibility requires that the nominee not turn 41 in the year the award is presented. The IACM John Argyris Award for Young Scientists is sponsored by Elsevier to honor Professor John Argyris' significant contributions in the field.
- **NSF CAREER Award**, National Science Foundation (Mechanics of Materials and Structures Program, Civil, Mechanics and Manufacturing Innovation Division), 2019. The NSF's most prestigious award in support of junior faculty who exemplify the role of teacher-scholar through outstanding research and excellent education, 2019.
- **EMI Leonardo Da Vinci Award**, the Engineering Mechanics Institute of American Society of Civil Engineers, 2018. The purpose of the award is to recognize outstanding young investigators early in their careers for promising ground-breaking developments in the field of Engineering Mechanics and Mechanical Sciences as relevant to Civil Engineering, understood in the broadest sense. The award is given annually to a young investigator, generally under 35 years of age or have worked no more than 7 years since receiving their doctoral degree, and whose contributions have the promise to define new directions in theory and application of Engineering Mechanics, in the vein of Leonardo da Vinci (1452-1519), a man of unquenchable curiosity and feverishly inventive imagination. The EMI of ASCE selected the PI *"for his fundamental contributions to computational multiscale poromechanics"*.
- **Zienkiewicz Numerical Methods in Engineering Prize**, Institution of Civil Engineers (ICE) and John Wiley & Sons, 2017. Instituted following a donation by John Wiley & Sons Ltd to commemorate the work of Professor Olgierd Cecil Zienkiewicz CBE. DSc FRS FREng of the Institute for Numerical Methods in Engineering, University of Wales, Swansea. The medal is awarded biennially by the Institution of Civil Engineers (ICE) to a researcher under 40 for the paper which contributes most to research in numerical methods in engineering, among 8 prime peer-reviewed journals published by ICE or Wiley, i.e., *Géotechnique*, *Géotechnique Letters*, *International Journal for Numerical Methods in Engineering*, *International Journal for Numerical Methods in Biomedical Engineering*, *International Journal for Numerical Methods in Fluids*, *International Journal for Numerical and Analytical Methods in Geomechanics*, *International Journal of Numerical Modelling: Electronic Networks, Devices and Fields*, and *ICE Proceedings*.
- **AFOSR Young Investigator Program Award**, Air Force Office of Scientific Research, US Air Force, 2017. The Air Force's Young Investigator Program (YIP) award is one of the most prestigious honors bestowed by the US Air Force to outstanding scientists beginning their independent careers. The program is designed to identify and support talented scientists and engineers who show

exceptional promise for doing creative research in order to encourage their teaching and research careers.

- **Awards received by PI's students and group Members**

- **Mindlin award** (SeonHong Na), Department of Civil Engineering and Engineering Mechanics, Columbia University, 2018.
- **Travel Scholarship** (Kun Wang and Chuanqi Liu), Workshop on Meshfree and Particle Methods: Application and Theory, Santa Fe, 2018.
- **Travel Scholarship** (Eric Bryant), 3rd Biennial CO₂ for EOR as CCUS conference, Petroleum Research School of Norway, 2017.
- **Dongju Lee Memorial Award** (SeonHong Na), Columbia University, 2017.
- **Travel Scholarship** (SeonHong Na), US National Congress of Computational Mechanics, Montreal, Canada, 2017.
- **Teaching Assistant Award** (SeonHong Na), Columbia University (Soil Mechanics), 2017.
- **2nd Place in Best Paper Student Competition** (SeonHong Na), Engineering Mechanics Institute, Modeling Inelasticity and Multiscale Behavior Committee, EMI 2016 & PMC 2016, Vanderbilt University, Nashville, TN, 2016.

2.3 List of publications through the performance of the award

The following journal articles have been published under the sponsorship of this award.

1. S. Na*, **W.C. Sun**, H. Yoon, M. Ingraham, Effects of elastic heterogeneity on the fracture pattern and macroscopic effective toughness of Mancos Shale in Brazilian tests, *Journal of Geophysical Research: Solid Earth*, 2017. [\[URL\]](#)
2. O.I. Ulven[†], **W.C. Sun**, Capturing the two-way hydro-mechanical coupling effect on fluid-driven fracture in a dual-graph lattice beam model, *International Journal for Numerical and Analytical Methods in Geomechanics*, 42(5):736-767, 2017. [\[URL\]](#)
3. K. Wang*, **W.C. Sun**, A multiscale multi-permeability poroplasticity model linked by recursive homogenizations and deep learning, *Computer Methods in Applied Mechanics and Engineering*, 334(1):337-379, 2018. [\[URL\]](#)
4. S. Na*, **W.C. Sun**, Computational thermomechanics of crystalline rock salt Part I: a combined phase field/crystal plasticity approach for single grain simulations, *Computer Methods in Applied Mechanics and Engineering*, 338:657-691, 2018. [\[URL\]](#)
5. J. Choo*, **W.C. Sun**, Coupled phase-field and plasticity modeling of geological materials: from brittle fracture to ductile flow, *Computer Methods in Applied Mechanics and Engineering*, 330:1-32, 2018. [\[URL\]](#)
6. J. Choo*, **W.C. Sun**, Cracking and damage from crystallization in pores: Coupled chemo-poro-mechanics and phase-field modeling, *Computer Methods in Applied Mechanics and Engineering*, 335:347-379, 2018. [\[URL\]](#)

7. **W.C. Sun**, T-F. Wong, Prediction of permeability and formation factors of sandstone with multiscale lattice Boltzmann/finite element simulation on microtomographic images, *International Journal of Rock Mechanics and Mining Sciences*, 106:269-277, 2018. [\[URL\]](#)
8. R. Gupta, S. Salager, **W.C. Sun**, K. Wang*, Open-source support toward validating and falsifying discrete mechanics models using synthetic granular materials Part I: Experimental tests with particles manufactured by a 3D printer, *Acta Geotechnica*, 2018. [\[URL\]](#)
9. G. Liu[†], **W.C. Sun**, S. M. Lowinger, Z. Zheng, M. Huang, J. Peng, Coupled flow network and discrete element modeling of injection-induced crack propagation and coalescence in brittle rock, *Acta Geotechnica*, 2018. [\[URL\]](#)
10. E. Bryant*, **W.C. Sun**, Mixed-mode phase field fracture for secondary cracks in anisotropic brittle rocks with consistent kinematics, *Computer Methods in Applied Mechanics and Engineering*, 342:561-584, 2018. [\[URL\]](#)
11. X. Zhong[†], **W.C. Sun**, An adaptive reduced-dimensional discrete element model for dynamic responses of granular materials with high-frequency noises, *International Journal of Multiscale Computational Engineering*, 16(4):345-366, 2018. [\[URL\]](#)
12. K. Wang*, **W.C. Sun**, An updated Lagrangian LBM-DEM-FEM coupling model for dual-permeability porous media with embedded discontinuities, *Computer Methods in Applied Mechanics and Engineering*, 334:276-305, 2019. [\[URL\]](#)
13. K. Wang*, **W.C. Sun**, Meta-modeling game for deriving theory-consistent, micro-structure-based traction-separation laws via deep reinforcement learning, *Computer Methods in Applied Mechanics and Engineering*, 346:216-241, 2019. [\[URL\]](#)
14. A. Qinami[†], E. Bryant*, **W.C. Sun**, M. Kaliske, Circumventing mesh bias by r- and h-adaptive techniques for variational eigen-fracture, *International Journal of Fracture*, 220:129-142, 2019.
15. C. Liu*, **W.C. Sun**, Shift domain material point method for solids in the finite deformation range, *Computational Particle Mechanics*, 2019.
16. K. Kang*, **W.C. Sun**, Q. Du, A cooperative two-player game for automated generations of elastoplasticity theories and models with AI-guided experimentation, *Computational Mechanics*, 64(2):467-499, 2019. [\[URL\]](#)
17. E. Bryant*, **W.C. Sun**, A micromorphic-regularized anisotropic Cam-clay for capturing size-dependent anisotropy of geomaterials, *Computer Methods in Applied Mechanics and Engineering*, 354:56-95, 2019. [\[URL\]](#)
18. Y. Heider*, **W.C. Sun**, Phase-field fracture in unsaturated porous media: application to drying-induced cracking, *Computer Methods in Applied Mechanics and Engineering*, accepted, 2019.
19. S. Na*, E.C. Bryant*, **W.C. Sun**, A configurational force for adaptive re-meshing of gradient-enhanced poromechanics problems with history-dependent variables, *Computer Methods in Applied Mechanics and Engineering*, 357, 2019. [\[URL\]](#)

20. Y. Heider*, K. Wang*, **W.C. Sun**, SO(3)-invariance of graph-based deep neural network for anisotropic elastoplastic materials, *Computer Methods in Applied Mechanics and Engineering*, accepted, 2019.
21. R. Ma*, **W.C. Sun**, FFT-based higher-order vs. multi-phase-field approaches for simulating strongly anisotropic brittle fracture, *Computer Methods in Applied Mechanics and Engineering*, accepted, 2019.

Chapter 3

Experimental Effort

3.1 One-dimensional oedometer consolidation tests

3.1.1 Introduction

A series of one-dimensional (1D) consolidation tests were designed and conducted using Avery Island (AI) salt samples to investigate the impact of clay and water contents on salt consolidation. The experimental tests were conducted with salt samples of 1.1 inch in height and 1.1 inch in diameter in the 1D oedometer. The salt used was crushed AI salt rock that was sieved between sieve #18 and #70. A test of salt only was performed as a reference and salt samples with varying water content (up to 7 % by weight) and clay content (up to 10%) were tested. The clay used in these tests was Ca-bentonite. The weights of the three components (salt, water, and clay) were calculated for the designated initial cylindrical sample volume to have an initial porosity of $\approx 40\%$. The sample mixture was placed in the oedometer cylinder and was consolidated at a constant consolidation rate of 10^{-5}s^{-1} by default. Three other consolidation rates were also evaluated. To obtain the experimental results at an intermediate step during the consolidation, two different maximum loading systems (2000 lbf and 5000 lbf or 12MPa and 30MPa in this work) were used. During testing, the sample was only allowed to deform axially, thus by collecting axial strain data, the change in porosity was estimated. More than 20 tests were conducted at ambient temperature ($\approx 20^\circ\text{C}$). For selected samples (>10), 3D microCT images were taken to evaluate the consolidated pore structure under different compositional and consolidation conditions.

3.1.2 Experimental results

We first evaluated experimental results performed under different maximum loading levels and different sample preparation procedures (i.e., order of water and clay addition to prepare a mixture of sample). Figure 3.1 (a) shows the consistency under three different conditions to confirm that experimental results ended at different consolidation levels can be interpreted together under the same compositional and testing conditions. In addition, we were able to achieve a low porosity ($<3\%$ porosity) with the higher loading frame. Figure 3.1 (b) shows how water and clay additives impact the consolidation process, compared to AI salt only. Since the same displacement rate (i.e., consolidation rate) was used, the addition of water and clay into the salt grains reduces the degree of stress or resistance, indicating that salt mixtures with additives clearly will be consolidated faster. The addition of 3% water slightly increases the consolidation rate compared to the dry salt

sample, while addition of both 5% clay and 3% water greatly increases the consolidation rate. To evaluate how each additive quantitatively impacts the consolidation rate, we evaluated the impact of each additive by varying the content. Figures 3.2 (a) & (b) show the impact of varying clay (with a constant 3% water content) and water (with a constant 5% clay content) contents on the consolidation respectively. Here, the case with 3% water and 5% clay contents was taken as the reference case. The results clearly show that the addition of 5% or higher clay content tends to increase the consolidation rate, while the addition of water content tends to slightly impact the consolidation rate. Although we did not evaluate the optimal water content at every clay content, with 5% clay content, a range of 1% to 7% water contents apparently has a similar impact on the consolidation rate. Overall, these experimental results were qualitatively in good agreement with previous results of consolidation on crushed salt and bentonite mixtures for shaft sealing purposes [BGR, Stührenberg 2007].

The main microstructure mechanisms that take place during consolidation of granular salt have been previously studied and described [Hansen, 2015]. At higher porosity ranges, the mechanisms that are expected to mainly drive the consolidation process are translational sliding and crushing of the grains, as well as mechanical and dynamic compaction – especially for a well-graded salt sample with wide range of particle sizes. At higher strains and lower porosity regimes, plasticity begins to participate in the consolidation process. The salt grains demonstrate plastic and ductile deformation, occupying the available pore space. Figure 3.3 shows a photo of consolidated sample (3% water and 5% clay). Figures 3.4 & 3.5 show the microCT images of horizontal and vertical profiles of consolidated samples for salt only, 3% water addition, and 3% water and 5% clay addition. The microCT image of salt sample before consolidation testing is shown in Figure 3.4 (a). Conducted sample imaging before and after consolidation shows that, after consolidation, few grains retain their initial boundary-pore interface as most of the pore space is eliminated (Figure 3.4). At this state possible grain pulverization might be possible, but this needs to be examined with thin-section samples. The presence of water in the sample apparently affects the consolidation procedure as shown in Figures 3.4 (b) - (d) where consolidated samples with 3% water content (Figures 3.4 (c) - (d)) have much less connected pores and pore pressure solution might smooth out the grain contact boundary. It is estimated that, at a microstructural level of grain contacts, the water trapped between grains – with no passage to diffuse at lower porosity states – furthers the plastic deformations locally. The addition of clay will accelerate the plastic deformations as shown in Figures 3.5 (c) where pore space is much less connected or distinctive. MicroCT images of consolidated salt samples will be further analyzed to mechanistically account for consolidation process in the presence of varying water and clay contents and thin section analysis will be performed to account for local consolidation processes.

3.2 Triaxial compression tests for salt-clay mixture

A series of triaxial compression strength tests were conducted on WIPP salt sample cores with clay inclusions. The experiments were performed for a combination of 0.5 Mpa, 5 MPa, and 10 MPa of confining pressure at 20°C and 60°C. The aim of these experiments was to investigate the strength and transition of ductile to brittle failure of the samples under different confining pressure and temperature conditions in the presence of clay inclusions. The tests were strain-controlled with a strain rate of $\dot{\epsilon} = 10^{-5} \text{s}^{-1}$. The samples

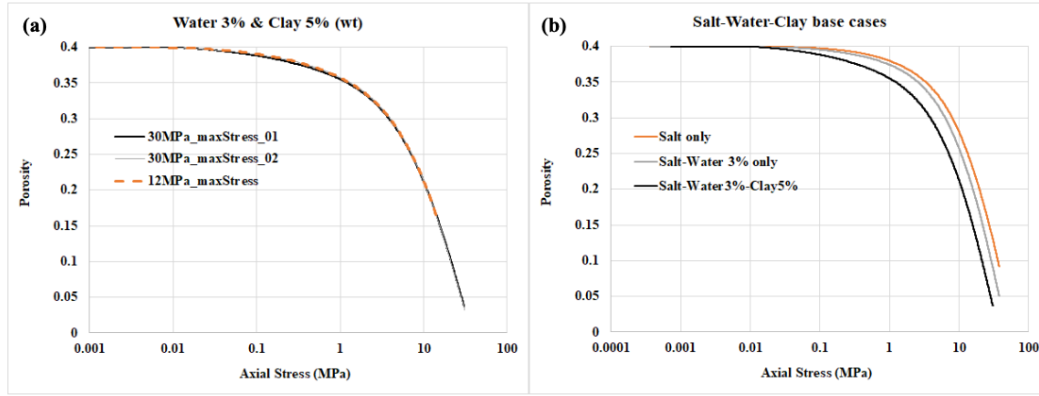


Fig. 3.1. (a) Consistency of consolidation testing of crushed Avery Island salt-water-bentonite clay mixtures. All cases use a mixture of 3% water, 5% clay, and 92% salt by weight. (b) Consolidation testing of three different compositions.

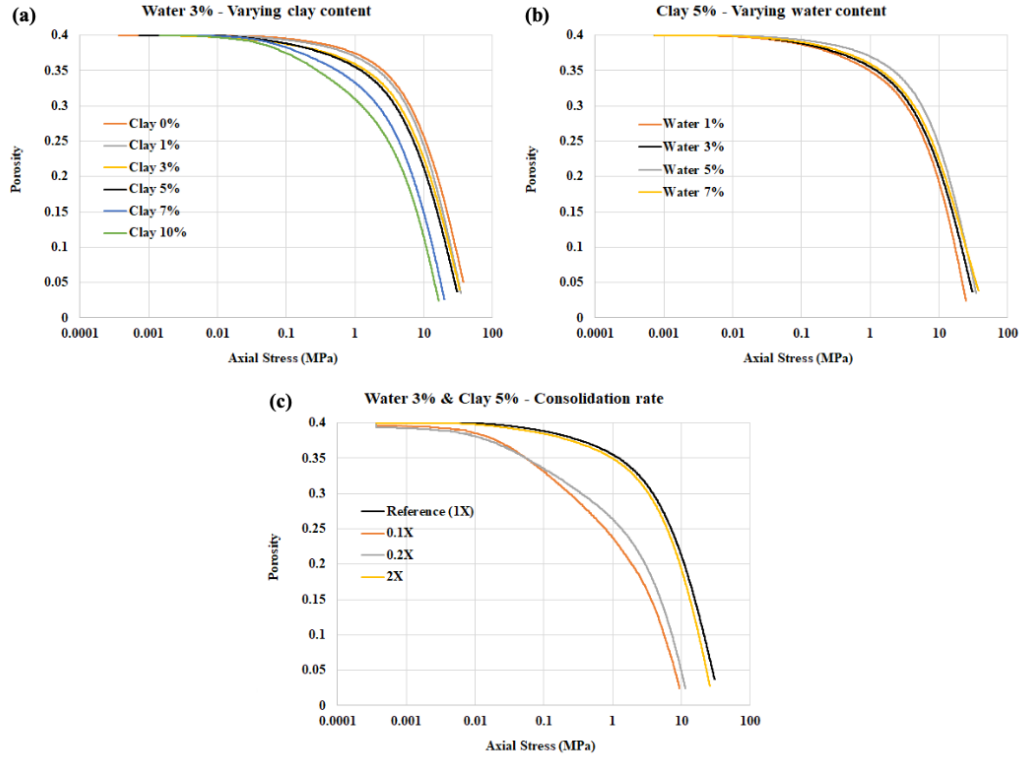


Fig. 3.2. Effects of (a) clay content, (b) water content, and (c) consolidation rate on the consolidation of crushed Avery Island salt with different percentages of water and bentonite clay. For testing, the reference case is 3% water and 5% clay content (by weight).

were cored to be 1 inch in diameter and 2 inches in height and were placed in a rubber jacket. Two Isco pumps (up to 10000 lbf) were used to control the axial and confining pressures, while temperature was controlled by a thermal wrapping unit attached to the core holder. During testing, the axial strain, volumetric strain, axial stress, confining pressure and temperature of the system were gauged. The LVDTs used for testing are currently under calibration to estimate the final measurements. The triaxial testing setup

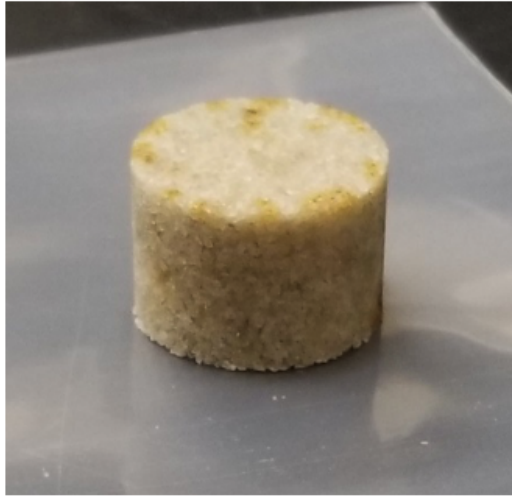


Fig. 3.3. Consolidated AI salt sample with water (3% wt) and clay (5% wt) contents.

can be seen in Figure 3.6.

Initially, the test specimens are progressively brought up to the target confining pressure and target temperature in hydrostatic condition, until the system stabilized. Following that the constant strain rate was set and the experiment progressed until failure of the sample – apparent by stress measure drop in the monitored stress strain curves of the sample – or until a target strain of 25% was reached in the more ductile sample responses.

The different strain-stress and volumetric responses can be seen in Figure 3.7. As the confining pressure applied to the samples is increased, their triaxial strength is increased and the strain at which failure occurs is larger. For example, in the case of testing in 0.5 MPa and 20°C failure occurs at a differential stress of 20 MPa and an axial strain of 7.5%, while when testing at 5 MPa failure occurs at approximately 42 MPa and at a strain of 22%. A similar transition is observed for an increase in temperature from 20°C to 60°C.

The brittle – ductile failure transition phenomenon is observed with a change in confining pressure and temperature. As the confining pressure and temperature is increased, the behavior of the salt samples change from brittle with a sudden drop in stress at failure, to semi-brittle, to ductile with failure not appearing at target strain. The brittle and ductile behaviors of the sample can be observed at the state of the samples after testing, shown in Figure 3.8. The samples that failed in a brittle manner crumpled. The samples with semi-brittle failure bulged but also demonstrated a subtle failure zone. The ductile samples appeared to only bulge until target strain was reached.

3.3 Long-term creep experiment

A long term creep test was performed with Avery Island (AI) sieved salt (between #18 and #70) mixed with 3% water and 5% clay additions. Please see the 1D consolidation results for different compositions of these mixtures. A sample is packed to 1 inch diameter and 2 inch height using a thermal shrink plastic. Two Isco pumps (up to 10000 lbf) were used to control the axial and confining pressures. The creep testing setup is shown in Figure 3.9. The testing specimen was under a hydrostatic condition with a constant 20 MPa axial and confining pressure for 75 hrs before the axial stress increased to 30MPa for $\approx$ 190 hrs at

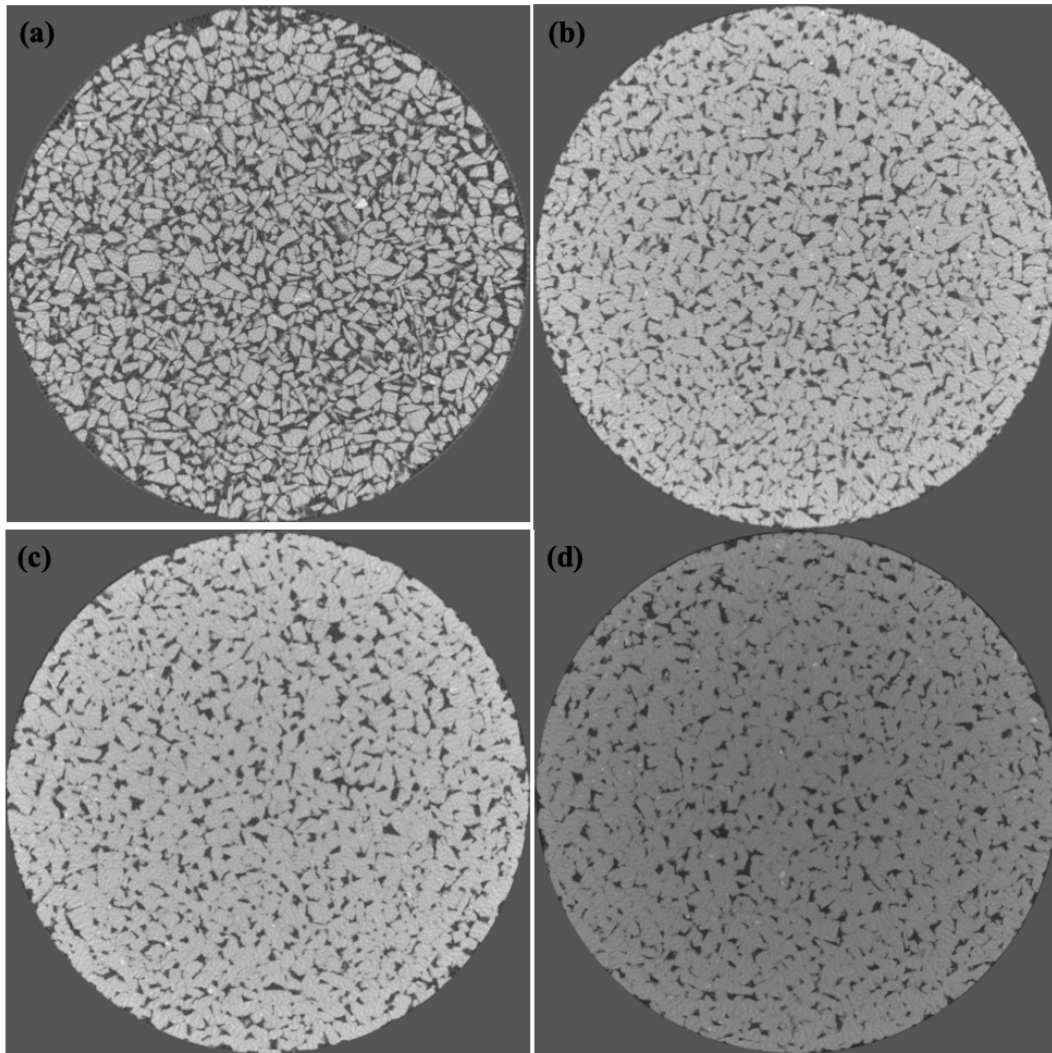


Fig. 3.4. MicroCT horizontal slice images of four different samples taken around the central region. (a) before consolidation testing and (b-d) after consolidation testing with (b) salt only, (c) 3% water content and no clay, and (d) 3% water and 5% clay contents.

60°C . Figure 1a shows the core holder setup with two Isco pump and thermal wrapping unit. The axial, confining pressure, temperature, displacement, and volumetric axial and confining unit changes were recorded in 10 second intervals during the testing period.

The recorded strain response over time can be seen in Figure 3.11. After the initial consolidation phase of the sample is complete, a decelerating creep effect is observed, especially in the later stages of the experiment. Currently, the calibration of LVDT and other compressibility corrections (confining fluid and sample jacket) are being performed to finalize the testing results. Figure 3.10 shows the 3D microCT images of the sample undergone the creep testing described above.

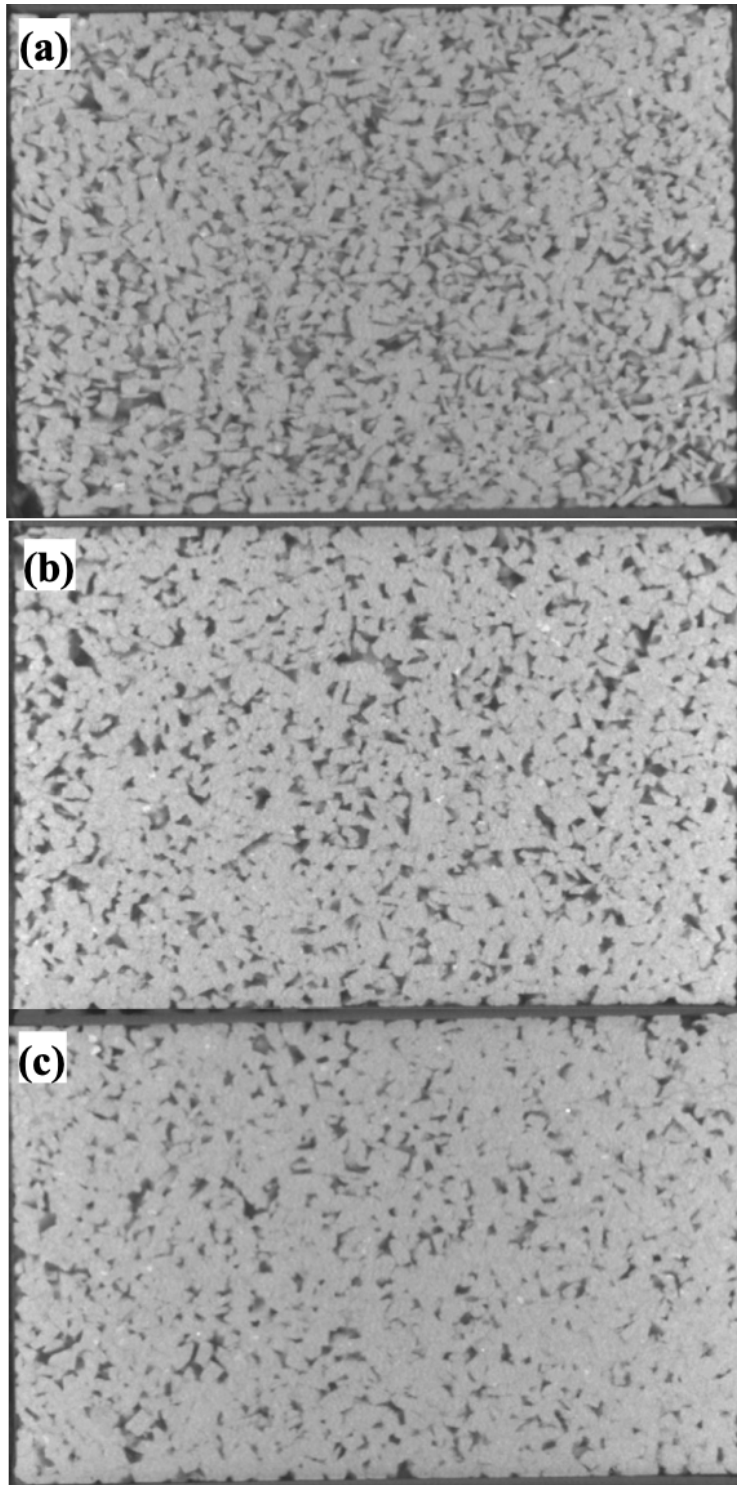


Fig. 3.5. MicroCT Vertical images of three different samples taken around the central region after consolidation testing. (a) salt only, (b) 3% water content and no clay, and (c) 3% water and 5% clay contents.

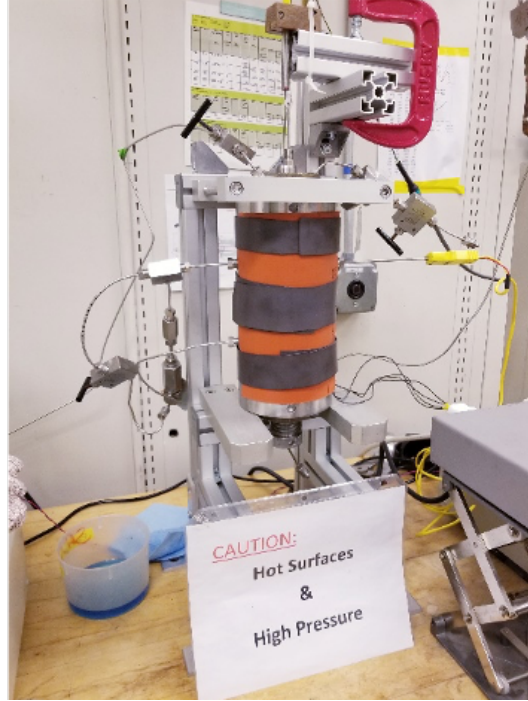


Fig. 3.6. Experimental setup for triaxial strength tests of the cored salt samples using two Isco pumps and a thermal unit.

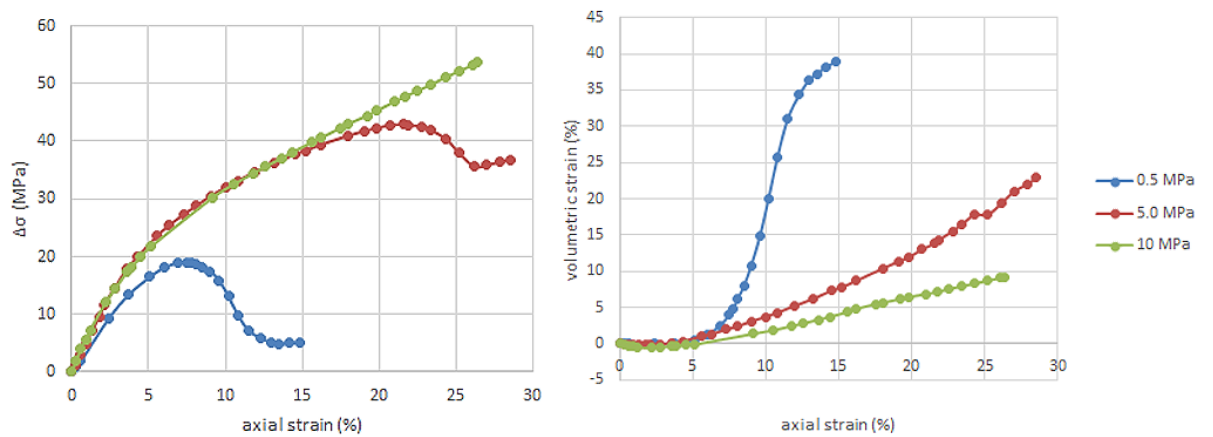


Fig. 3.7. Triaxial strength test results for salt samples at 20°C at confining pressures of 0.5 MPa, 5 MPa, and 10 MPa at a strain rate $\dot{\epsilon} = 10^{-5} \text{ s}^{-1}$.

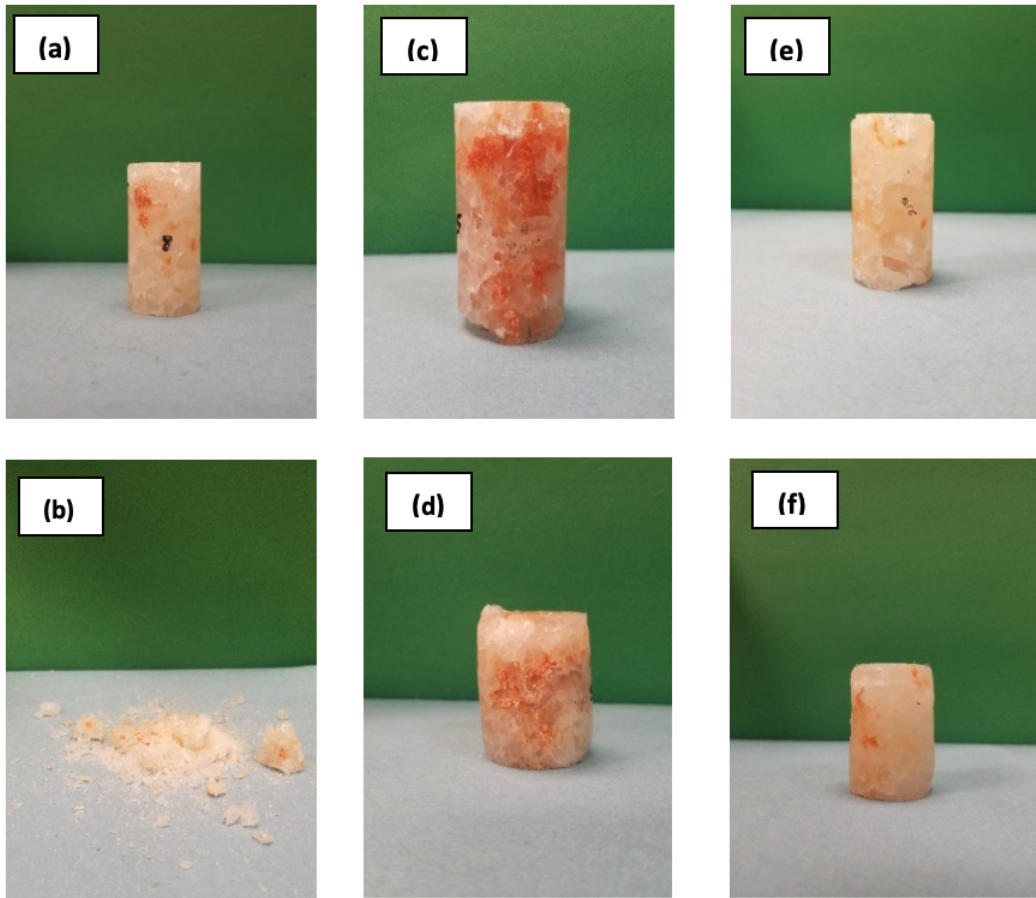


Fig. 3.8. Salt samples before (top) and after testing (bottom) at 20°C for confining pressures of 0.5 MPa (a, b), 5 MPa (c, d), and 10 MPa (e, f).

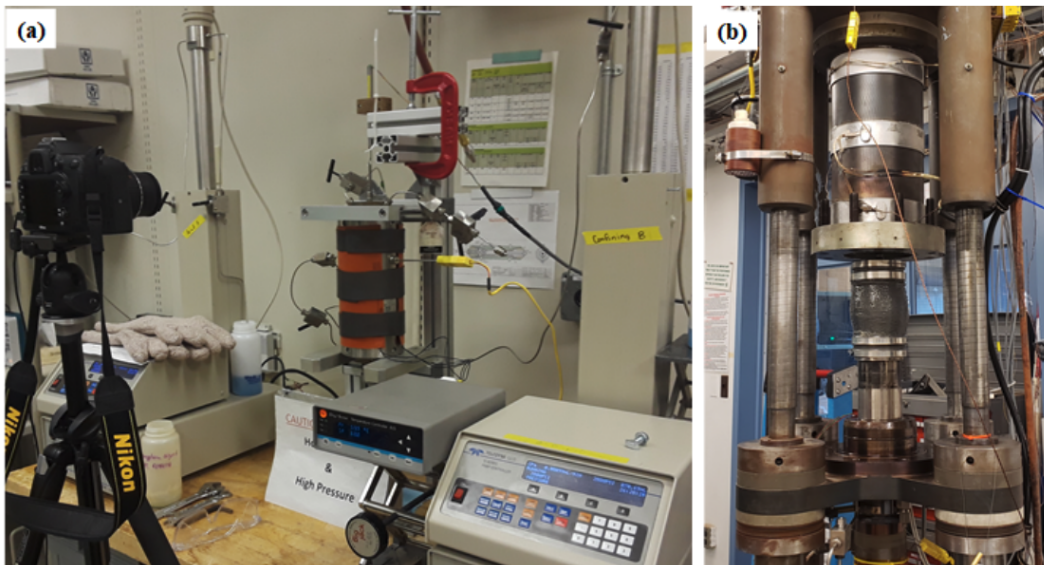


Fig. 3.9. Experimental setup for (a) a small sample (1 inch diameter) using two Isco pumps and thermal unit and (b) a large sample (≈ 4 inches diameter) with a lead jacket with thermal controlling unit.

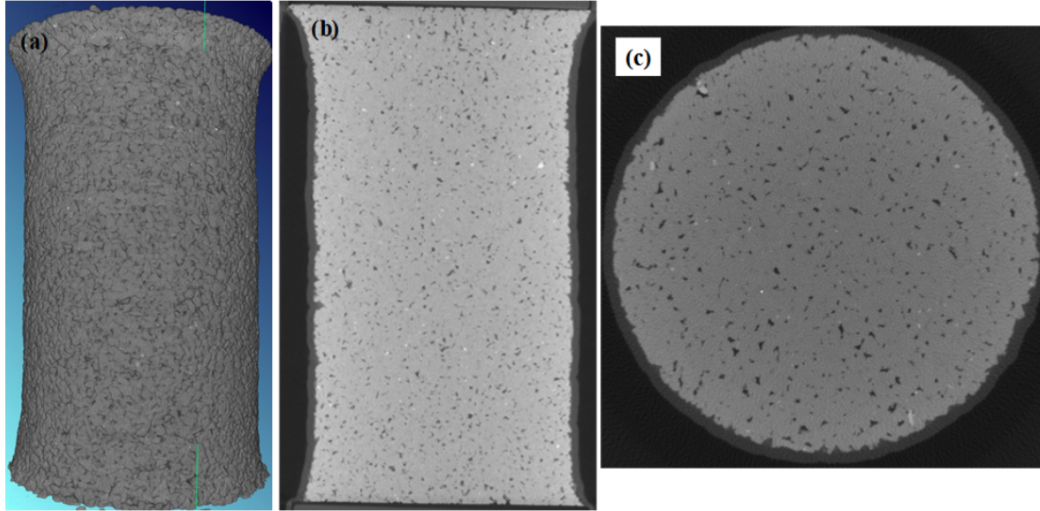


Fig. 3.10. MicroCT images of Avery Island salt sample with 3% water and 5% clay addition after creep testing. (a) 3D view of the sample and (b-c) vertical and horizontal views along the center of the sample.

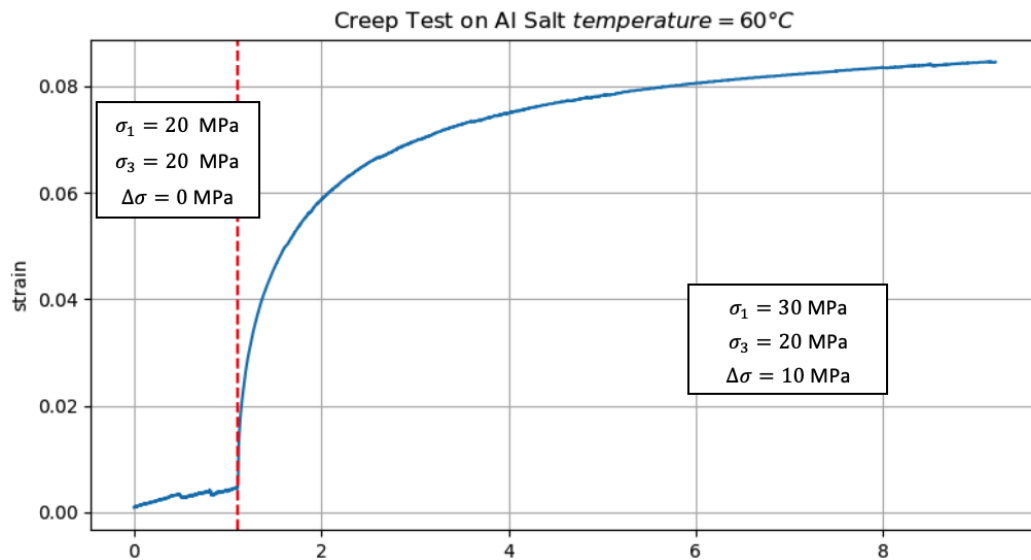


Fig. 3.11. Strain evolution for the creep testing of an 1-inch diameter salt sample (water 3% - clay 5%) at 60°C. The sample was initially under isotropic pressure of 20 MPa and was then subjected to differential stress of 10 MPa.

Chapter 4

Micro-mechanics of single-crystal salt

4.1 Introduction

The demands for safe and permanent disposal of nuclear waste in geologic formations dates back over decades. Natural rock salt found in domal and bedded formations and the re-consolidated counterpart formed in a high-pressure and high-temperature environment, have been used for geological repositories of nuclear waste disposal in the United States and Germany [[Kuhlman and Malama, 2013](#)]. Two operating facilities include, the Waste Isolation Pilot Plant (WIPP) in Carlsbad (New Mexico, USA), and the Endlager für radioaktive Abfälle Morsleben (ERAM) site in Morsleben, Germany [[von Berlepsch and Haverkamp, 2016](#)].



Fig. 4.1. Disposal operations for Transuranic (TRU) waste at the Waste Isolation Pilot Plant (WIPP). (Reproduced from [Hansen and Leigh \[2011\]](#).)

The decision to use salt formation for the storage and disposal of radial active wastes is attributed to the following desirable thermo-hydro-mechanical-chemical characteristics, i.e., (1) high thermal conductivity, (2) low permeability, (3) self-healing mechanism and, (4) biologically inactivity of rock salt (as compared with clay). Firstly, the heat generated from nuclear wastes can be dissipated to the surrounding area much faster in salt than

in other types of materials since the host salt rock exhibits high thermal conductivity [Urquhart and Bauer, 2015]. In addition, the permeability of the rock salt is sufficiently low that it is often idealized as impermeable. Therefore, it may function as a secured barrier for radioactive wastes [Bechthold et al., 2004]. Finally, the creeping property of salt enables micro-cracks or damages under mechanical load to be self-sealed, which may also naturally guarantee the necessary geological barrier function (e.g., von Berlepsch and Haverkamp [2016], Chan et al. [1998]).

While decades of experimental investigations provided insight on mechanisms of the brine migration, vapor transport, and related solution-precipitation creeps under nonisothermal conditions (e.g., Kuhlman and Malama [2013], Bradshaw and Sanchez [1969], Nowak and McTigue [1987], Beauheim et al. [1995], Schlöder et al. [2008], Hansen et al. [2012]), the numerical modeling of crystalline rock salt remains an active research area. Analysis of the complex multi-physical responses of natural and reconsolidated salt is an integral component for the design of short- and long-term life cycle of a salt repository. In the repository, the facility may have various phases of operation cycles that involve plugging, sealing, testing and re-consolidation of the saline materials [Hansen and Leigh, 2011]. Under such conditions, the reconsolidated salt is a poly-crystalline rock that is influenced by brine inclusions inside halite crystals, moisture content, and other materials such as clay or impurity that might present in the grain boundary. Within a certain temperature and confining pressure ranges (between 250°C to 790°C and between 0.15 and 12 MPa), the responses of salt may become even more complicated due to re-crystallization and grain boundary migration. This generally leads to precipitation creeping [Guillope and Poirier, 1979]. A number of previous studies, therefore, investigate the phenomenology and micro-mechanisms associated with the mechanical behavior of salt (e.g., Carter et al. [1982], Carter and Hansen [1983], Urai et al. [1986], Carter et al. [1993], Senseny et al. [1992], Urai et al. [2008], Hansen et al. [2014b]). These studies have explored the dislocation, dissolution-precipitation creep, healing behavior, and the related flow transport properties of natural or reconsolidated salt under a wide range of load and temperature conditions in a phenomenological setting where microstructural attributes of the rock salt are not explicitly modeled.

Since salt formations found in nature often contain varying amounts of impurities, minerals, and brine inclusion, how to capture these spatial heterogeneous effects without explicitly model the microstructures remains a major challenge. Various phenomenological constitutive models have been proposed to capture the macroscopic responses of salt (e.g., Olivella and Gens [2002], Chan et al. [1998], Senseny et al. [1992], Munson and Dawson [1979], Chan et al. [1995], Miao et al. [1995], Munson [1997], Aubertin et al. [1999], Zhu and Arson [2015a], Broome et al. [2015], Shen et al. [2016]). For instance, Olivella et al. [1994a] propose a general formulation for nonisothermal multiphase flow of gas and brine in saline media, which was further extended into a series of works for coupled thermo-hydro-mechanical analysis of saline materials (e.g., Olivella et al. [1996a,b], Olivella and Gens [2000]). Although the grain boundary, damage and healing, and the microstructural information were not incorporated explicitly, the complicated history- and rate-dependent behaviors of saline media, as well as the multiphase interactions among constituents including gas, liquid, and solid, are addressed by linking fluid flow, permeability change, thermal gradient, with phenomenological constitutive law for the solid skeleton.

While these phenomenological approaches inspired by micro-mechanics of crystals have achieved a level of success in the past, more physically consistent forward prediction may require stronger physical underpinning such that the interaction between the

impurity, the precipitated brine and the anisotropy of crystalline constitutive responses can be captured properly [Hansen and Leigh, 2011].

Yet, bridging the multiscale coupling effects from the grain scale to the field applications remains a challenging task due to the complexity of the physical natures of rock salt. In this work, we are motivated by the earlier work on crystalline rock [Anand and Kothari, 1996] in which the crystalline nature of rock is considered and captured. Our ultimate goal is to create a multiscale poly-crystalline material model suitable for field-scale application. As a first step toward this goal, this article will focus on the modeling thermo-mechanical responses of a single-crystal halite. In this paper, our objective is to derive, implement and validate an elasto-plastic model for a single-crystal halite subjected to different thermal and mechanical loadings relevant to the nuclear disposal applications. We will incorporate other important multiphysical and multiscale polycrystalline mechanisms such as the interactions across grain boundaries, the intra-crystalline and inter-crystalline brine inclusion, and the role of the impurity and additive on the precipitating and pressure solution of the grain boundaries in the future contributions of this series of work.

In particular, we present a unified mathematical framework that enables us to capture the anisotropic inelastic brittle and ductile behaviors of a single crystal halite. We achieve this by combining the thermal-sensitive rate-dependent crystal plasticity formulation that captures the anisotropic plastic deformation caused by the slip of crystallographic planes, and a multi-phase-field regularized fracture model that captures the interactions of the anisotropic intra-grainular and transgranular fractures of single-crystal halite under temperature range relevant to nuclear waste disposal applications, we introduce a finite element formulation.

The intrinsic anisotropy of halite stemming from the microstructure is incorporated through the crystal plasticity theory. The crystal plasticity theory is a micro-mechanics-based constitutive law where anisotropic plastic flow is associated with the activation(s) of slip system oriented according to the lattice structure of the crystalline materials [Rice, 1971, Peirce et al., 1982, Havner, 1992, Gurtin et al., 2010, de Souza Neto et al., 2011]. In this work, we use the strain energy equivalence theory such that the crystal plasticity framework can be integrated into a phase field model that captures the evolution of damage. As the creeping mechanisms, such as grain boundary diffusion and dislocation creep, and thermal activated glide are highly sensitive to temperature, the temperature-dependent energy dissipation due to creeping is captured in the crystal plasticity model (cf. Anand and Kothari [1996], Cuitino and Ortiz [1993], Borja and Wren [1993], Miehe and Schröder [2001], Borja and Rahmani [2012], Tjioe and Borja [2015]). The damage and crack growth of the single-crystal material is captured via a phase field approach, which is a sub-class of the smeared crack approach. This approach allow us to capture complicated crack patterns without introducing an embedded discontinuity (e.g., Miehe et al. [2010a,b], Clayton and Knap [2011, 2015a], Na et al. [2017a]). In particular, we use a sub-class of phase field models that employs multiple phase fields to capture damage accumulated in different directions. Multi-phase-field model has been a very popular approach to model a wide spectrum of material behaviors including anisotropic responses of regularized interfaces for planar dislocation (e.g. Koslowski and Ortiz [2004]), re-crystallization (e.g. Takaki et al. [2009]) and anisotropic fracture (e.g. [Oshima et al., 2014, Nguyen et al., 2017a,b]). See also Steinbach [2009] for a comprehensive review. In the content of anisotropic phase field model, one obvious advantage of the multi-phase-field approach is the lack of higher-order term that is typically required for the single-phase-field counterpart to capture non-convex anisotropic fracture energy Li et al. [2015]. As a result, the multi-phase-field model does not require specific finite element

space spanned by piece-wise smooth and globally C^1 -continuous basis functions. Instead, a simple low-order finite element can be used. This simplicity, although offset by the additional computational cost due to the additional degrees of freedom, motivate us to use the multi-phase-field model in this work. The number of independent phase-field field and their directional features are assumed to be consistent with the slip planes of halite. Finally, the crystal plasticity theory and anisotropic multiple phase-field approach are incorporated into a set of multi-physical field equations in which the balance of linear momentum, micro-forces for the plastic slip and phase field and the energy are considered. An operator-split integration scheme is introduced to construct a feasible algorithm for numerical simulations.

The organization of the paper is as follows. In Section 4.2, we first describe the kinematics of a halite grain with slip system. Then, the application of effective stress equivalence theory on the multi-phase-field model for the anisotropic fracture of crystalline materials is discussed. Within this mathematical framework, the general governing equations, including the balance of linear momentum, microforce equations, and balance of energy are derived. The specific choice of free energy function that combines the crystal plasticity and multiple phase-field approaches is provided. Subsequently, in Section 4.3, the stress update algorithm in effective stress space is described followed by the description of both rate-dependent and rate-independent settings. In Section 4.4, the finite element formulations, as well as solution strategies for nonlinear systems of equations, are presented. Numerical examples are then provided in Section 4.6, followed by a conclusion.

As for notations and symbols, bold-faced letters denote tensors; the symbol $\cdot$ denotes a single contraction of adjacent indices of two tensors (e.g., $\mathbf{a} \cdot \mathbf{b} = a_i b_i$ or $\mathbf{c} \cdot \mathbf{d} = c_{ij} d_{jk}$); the symbol $:$ denotes a double contraction of adjacent indices of tensor of rank two or higher (e.g., $\mathbf{C} : \boldsymbol{\epsilon}^e = C_{ijkl} \epsilon_{kl}^e$); the symbol $\otimes$ denotes a juxtaposition of two vectors (e.g., $\mathbf{a} \otimes \mathbf{b} = a_i b_j$) or two symmetric second order tensors (e.g., $(\boldsymbol{\alpha} \otimes \boldsymbol{\beta}) = \alpha_{ij} \beta_{kl}$). As for sign conventions, unless specify otherwise, we consider the direction of the tensile stress and dilative pressure as positive.

4.2 Governing equations

In this section, we present the field equations that capture the thermo-mechanical coupling effect of a single-crystal halite. We begin by reviewing kinematics of deformation of a crystalline solid with multiple slip systems. To capture the brittle-ductile transition and the brittle fractures that might occurs under low confining pressure, a multiple-phase-field model is used to capture the anisotropic damage evolution. Adopting the ideas originated from [Nguyen et al. \[2017b\]](#), we introduce multiple phase fields to model the anisotropic damage such that each phase field represents the damage along a particular preferential direction. Finally, the coupled anisotropic damage-plasticity behaviors of the single-crystal halite is captured via the effective stress theory in which one hypothesize the existence of fictitious undamaged configuration where stress experienced by the undamaged elasto-plastic body is considered effective [[Ekh et al., 2004](#), [de Borst and Verhoosel, 2016a](#)]. The total stress is then recovered from the damaged configuration which is related to the effective stress and the anisotropic gradient damage computed from the phase fields. Meanwhile, the field theory, including the balance of linear momentum and the balance of energy equation that describes the internal energy changes due to various mechanical and thermal dissipation and the structural heating due to the changes

of microstructural attributes of the halite crystal.

4.2.1 Strain energy equivalence for coupling phase field and plasticity model

In the proposed work, an anisotropic phase field fracture framework is coupled with the crystal plasticity model to simulate the anisotropic path-dependent behavior of single-crystal halite. Here we adopt strain energy equivalence principle used previously in isotropic and anisotropic damage mechanics [de Borst and Verhoosel \[2016a\]](#), [Lee et al. \[1997\]](#), [Simo and Ju \[1989\]](#) and assume that there exists a fictitious undamaged counterpart of the damaged halite such that a body composed of the fictitious undamaged material will experience an effective stress $\hat{\sigma}$ while the actual body will experience a stress σ . Furthermore, we assume that, in spite of the damage characterized by the driving force of the multiple phase fields is anisotropic, the effective stress and the total stress are co-axial. Consequentially, (1) the degradation function $g(\underline{d})$ becomes a scalar function of the collection of phase fields $\underline{d}$ and the relation between the total and effective stress when both the fictitious and the real material exhibit the same infinitesimal strain (i.e., $\epsilon = \hat{\epsilon}$) are,

$$\sigma = g(\underline{d})\hat{\sigma}, \quad (4.1)$$

and the corresponding infinitesimal stress rate is,

$$\dot{\sigma} = \sum_{i=1}^n \frac{\partial g(\underline{d})}{\partial d_i} \dot{d}_i \hat{\sigma} + g(\underline{d}) \dot{\hat{\sigma}} \quad (4.2)$$

where $d_i, i, 2, \dots, n$ is the phase field for each preferential direction and n is the total number of the phase field. An important implication of (4.1) is that while damage and plastic processes remain coupled, the plasticity model can be associated with the fictitious undamaged body and hence one may use a de-coupled plasticity algorithm to first obtain the effective stress from the strain field then update the total stress using the degradation function from each material point.

4.2.2 Kinematics for crystal deformation of the damaged halite

For completeness, we briefly review the kinematics of crystal deformation and restrict the formulation within the infinitesimal deformation range. More comprehensive treatment of this subject can be found in, for instance, [Borja \[2013\]](#), [Borja and Wren \[1993\]](#), [Miehe and Schröder \[2001\]](#), [Budiansky and Wu \[1961\]](#). Recall that we adopt the approach in [Simo and Ju \[1989\]](#) in which we assume that the actual (damaged) crystal and the fictitious counterpart undertake the same strain and strain history.

The elasto-plastic deformation of a single crystal grain is attributed to the interactions between crystal blocks along predefined crystallographic planes in a slip system. A *slip-system* is defined by a combination of crystallographic planes (i.e., *slip planes*) and the corresponding sliding directions (i.e., *slip directions*). The atomic arrangement of the slip system of the crystalline material leads to a preferential direction of the plastic flow and hence the overall constitutive responses of a single crystal is anisotropic. In Face Cubic Centered (FCC) crystal, for example, the slip-systems are defined by the densest packing planes in terms of atomic arrangement as: the slip-plane $\{111\}$ and slip-direction $\langle 110 \rangle$. This dense packing causes FCC materials, such as aluminum, copper, gold and silver exhibiting higher ductility than the Body Cubic Center (BCC) crystals such as, iron and chromium (although packing alone is not the only factor that influences ductility) [[Borja](#),

[2013]. One may view the halite structure as a FCC structure with secondary atoms in its octahedral holes. As a result, a set of slip-planes is known to be $\{110\}$ (rather than $\{111\}$ for FCC) and the corresponding slip directions are $\langle 1\bar{1}0 \rangle$. This plane is electrically neutral and motion on this plane avoids charged layers gliding over one another (cf. Hansen et al. [2014b], Carter and Norton [2007]).

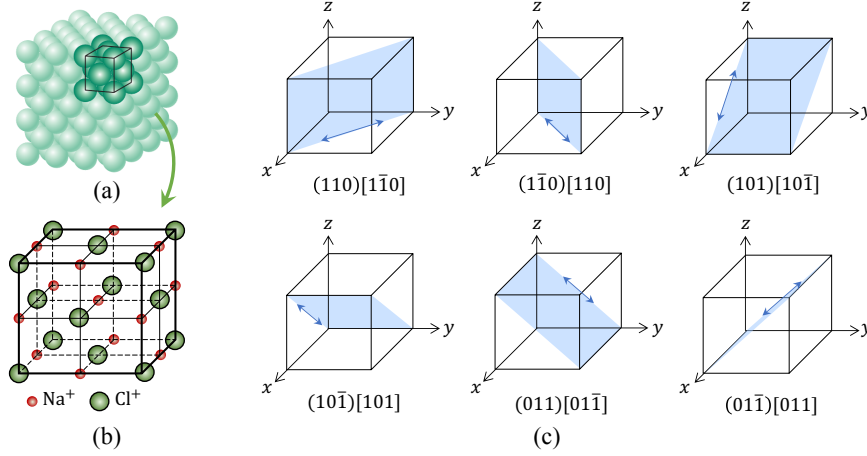


Fig. 4.2. The description of the slip-system of a single-crystal halite, (a) an aggregate of many atoms for the face centered cubic crystal structure (reproduced from Callister Jr and Rethwisch [2012] – adapted from the original Moffat et al. [1964]), (b) a unit cell for the sodium chloride (NaCl) crystal structure (reproduced from Callister Jr and Rethwisch [2012]), and (c) the slip-system $\{110\}\langle 1\bar{1}0 \rangle$.

We consider a continuum body ($\mathcal{B}$) with material points identified by the position vectors $\mathbf{x} \in \mathcal{B}$. The displacement of a material point with time t can be denoted by $\mathbf{u}(\mathbf{x}, t)$, and we define the strain measure ($\boldsymbol{\epsilon}$) as the symmetric part of the displacement gradient, i.e.,

$$\boldsymbol{\epsilon} := \nabla^{\text{sym}} \mathbf{u} = \frac{1}{2}(\nabla \mathbf{u} + (\nabla \mathbf{u})^T). \quad (4.3)$$

The additive decomposition of the infinitesimal total strain leads to,

$$\boldsymbol{\epsilon} = \boldsymbol{\epsilon}^e + \boldsymbol{\epsilon}^p, \quad (4.4)$$

where $\boldsymbol{\epsilon}^e$ and $\boldsymbol{\epsilon}^p$ indicate the elastic and plastic components, respectively. In crystals, the elastic strain describes distortion of the lattice structure which vanishes when the applied stress is released. On the other hand, the plastic strain is irreversible and evolves due to the slip on crystallographic planes. Among the multiple slip systems, we denote by $\mathbf{m}^\alpha$ the unit normal to a crystallographic plane containing α -slip system and by $\mathbf{s}^\alpha$ the corresponding direction of plastic slip. The slip-system α is defined by orthonormal vectors $(\mathbf{m}^\alpha, \mathbf{s}^\alpha)$ which disclose the microscopic information based on the crystallographic features of crystals. We then obtain the plastic strain by summing over all crystallographic slips,

$$\boldsymbol{\epsilon}^p = \sum_{\alpha} \gamma^{\alpha} \mathbf{S}^{\alpha}, \quad (4.5)$$

where γ^{α} indicates the plastic slip corresponding to the slip-system α . $\mathbf{S}^{\alpha}$ denotes the

symmetric part of the Schmid tensor (i.e., $\mathbf{m}^\alpha \otimes \mathbf{s}^\alpha$), which can be expressed as follows,

$$\mathbf{S}^\alpha = \frac{1}{2} (\mathbf{m}^\alpha \otimes \mathbf{s}^\alpha + \mathbf{s}^\alpha \otimes \mathbf{m}^\alpha). \quad (4.6)$$

Note that the summation convention is not employed in respect of indices relating to the slip systems.

The thermal expansion and structural heating will be included by using a thermal-sensitive elastic stored energy function (cf. Eq. (4.40)) for thermoelastic coupling. For simplicity, the thermal effect on elastic response is assumed to be isotropic, following the treatment of [Anand and Gurtin \[2003\]](#). The anisotropic thermal expansion can be considered by formulating the diagonal tensor of the thermal expansion coefficients with respect to the principal crystallographic directions (e.g., [Meisssonier et al. \[2001\]](#)). The characterization of anisotropic thermal parameters, however, is often neglected in the literature partly due to the lack of data to identify material parameters for anisotropic thermal behavior (e.g., thermal conductivity, expansion coefficients, etc.) in the field [Sun \[2015a\]](#). The thermal effect on the inelastic behavior of single crystal halite is incorporated via the flow rule that considers the impact of temperature on the plastic slip (Section 4.2).

4.2.3 Multi-phase-field approximation for anisotropic fracture

The phase field fracture modeling can be considered as a regularized smeared approach in which the cracks are not explicitly captured via embedded discontinuities but by implicit indicator function obtained from regularizing the strong discontinuities with a characteristic length [[Bourdin et al., 2008a](#), [Schmidt et al., 2009](#), [Wang and Sun, 2017a](#)]. Here we assume that this characteristic length is sufficiently smaller than the grain size but larger than the finite element mesh size we used for numerical simulations. In those cases, the strongly anisotropic damage and fracture behaviors can be captured either via a single phase field within a higher-order Cahn-Hilliard framework (e.g., [Clayton and Knap \[2015a\]](#), [Li et al. \[2015\]](#)) or via multiple phase fields, each constrained by governing equation without the forth-order terms (e.g., [Nguyen et al. \[2017a,b\]](#)). Due to the simplicity of the latter approach, we adopt the latter approach to capture the damage and fracture behavior in a single crystal halite.

In a regularized framework where the sharp crack topology is approximated by a diffusive representation, the phase-field variable ($d(\mathbf{x}, t) \in [0, 1]$) is an implicit function whose value indicates the location of the smeared crack(s). Let $\mathcal{T}$ denotes a set of discontinuous fractures inside a body $\mathcal{B}$. Then the total area of the crack surfaces can be described by an area integral over $\mathcal{T}$. By introducing crack density function $\Gamma_l(d, \nabla d)$ the total crack area can be described by the volume integral as,

$$\Gamma = \int_{\mathcal{T}} dA \approx \int_{\mathcal{B}} \Gamma_l(d, \nabla d) dV \quad \text{where} \quad \Gamma_l(d, \nabla d) := \frac{1}{2l} d^2 + \frac{l}{2} |\nabla d|^2. \quad (4.7)$$

Here l indicates a length scale that controls the width of the smooth approximation of the crack, and the assigned scalar phase-field values 0 and 1 represent the intact and completely damaged region, respectively. A phase-field value in between $0 < d < 1$ indicates a partially damaged material state at the corresponding material point. The regularized crack surface can be further extended to anisotropic case as,

$$\Gamma_l(d, \nabla d, \mathbf{w}) := \frac{1}{2l} d^2 + \frac{l}{2} \mathbf{w} : (\nabla d \otimes \nabla d) \quad (4.8)$$

where ω is a second-order structural tensor, which is invariant with respect to rotations for characterizing the material anisotropy [Clayton and Knap, 2015a]. This tensor can be defined as,

$$\omega = \mathbf{1} + \beta(\mathbf{1} - \mathbf{m} \otimes \mathbf{m}), \quad (4.9)$$

where $\mathbf{1}$ denotes the second-order identity tensor; $\mathbf{m}$ indicates the unit normal vector to the potentially preferred cleavage plane; $\beta \gg 1$ is the parameter penalizing fracture on planes not normal to the unit vector $\mathbf{m}$. The isotropic crack surface energy function in Eq. (4.7) can be recovered by setting $\beta = 0$.

To capture the anisotropy inherent to the crystalline materials, we associate the orientations of each slip plane as a potential cleavage plane. Adopting the multi-phase-field technique first introduced by Oshima et al. [2014] and further developed in [Nguyen et al., 2017a,b], we assign multiple phase fields $\underline{d} = \{d_1, d_2, \dots, d_n\}$ to quantify the damage accumulation on each slip plane normal to unit vector $\mathbf{m}^i$. Therefore, the total crack length can be defined by the summation of each anisotropic crack density function which is be rewritten as [Nguyen et al., 2017a,b],

$$\Gamma_1(\underline{d}, \nabla \underline{d}, \underline{\omega}) = \sum_i \left[\frac{1}{2l} (d_i)^2 + \frac{l}{2} \omega_i : (\nabla d_i \otimes \nabla d_i) \right],$$

where $\omega_i = \mathbf{1} + \beta(\mathbf{1} - \mathbf{m}^i \otimes \mathbf{m}^i).$ (4.10)

For convenience we use the underline to denote a set of variables. In analogous to Eq.(4.6), $\mathbf{m}^i$ corresponds to $\mathbf{m}^\alpha$ – the unit normal vector of each crystallographic slip plane. The length scale l and penalizing parameter β are assumed to be the same for each slip system in this study. These parameters may further differentiate for each slip system based on the microscopic information of crystalline materials.

4.2.4 Balance of linear momentum and microforce

In this section, we derive the balance of linear momentum and microforce for a single-crystal with damage via the principle of virtual power. The virtual-power formulation of the single-crystal using microforce balance is well established and has been described in the literature (cf. Gurtin et al. [2010]). Based on this standard theory, De Lorenzis et al. [2016] extended the virtual power formulation by introducing an additional microforce balance associated with a crack phase-field. In this work, our new contribution is to extend this work for the multi-phase-field formulation coupled with crystal plasticity. We postulate the existence of multiple microforces, each of them conjugate to a phase field for anisotropic damage in a single-crystal. Furthermore, we also extend this framework to incorporate thermal diffusion such that non-isothermal condition can be simulated. For simplicity, we limit our analysis within the small deformation range and the small temperature difference assumption (cf. Anand and Gurtin [2003]) is used. As described in Section 4.2.2, therefore, the thermal expansion effect is included in the elastic stored energy function Eq. (4.40) based on the standard virtual power formulation [Anand and Gurtin, 2003]. This treatment may be sufficient for a geological disposal under the limited temperature change condition for low-level radioactive waste materials, for example, generated by military activities (e.g., the Waste Isolation Pilot Plant, WIPP) [Hansen and Leigh, 2011, Bredehoeft et al., 1978].

The standard derivation procedure using the principle of virtual power for phase-field modeling of fracture in a single-crystal is straight forward and can be found in

great details in [Gurtin et al. \[2010\]](#), [De Lorenzis et al. \[2016\]](#). Here we briefly outline the expression of virtual power formulation including macroforce (i.e., the total Cauchy stress), microforce for a single-crystal, and additional microforces for multiple phase-field, and introduce the resultant balance equations. We use the symmetric Cauchy stress considering the infinitesimal strain theory. Considering a domain $\mathcal{B}$, first, the internal power ($\mathcal{J}$) over a subregion $\mathcal{P} \subset \mathcal{B}$ may take the form of,

$$\mathcal{J}(\mathcal{P}) = \int_{\mathcal{P}} \boldsymbol{\sigma} : \dot{\boldsymbol{\epsilon}}^e dV + \sum_{\alpha} \int_{\mathcal{P}} \pi^{\alpha} \dot{\gamma}^{\alpha} dV + \sum_i \left(\int_{\mathcal{P}} \boldsymbol{\xi}_i \cdot \nabla \dot{d}_i dV + \int_{\mathcal{P}} \pi_i \dot{d}_i dV \right), \quad (4.11)$$

where $\boldsymbol{\sigma}$ indicates the Cauchy stress with the power-conjugate to $\dot{\boldsymbol{\epsilon}}^e$; π^{α} denotes the microscopic force associated with the slip rate $\dot{\gamma}^{\alpha}$; $\boldsymbol{\xi}_i$ is the microscopic stress power-conjugate to $\nabla \dot{d}_i$; and π_i is the microscopic internal body force (power conjugate to $\dot{d}_i$). Note that the summation convention is not employed regarding indices related to multiple phase-field, and each phase-field variable corresponds to the direction of a crystalline slip system. As mentioned in Section 4.2.1, $\boldsymbol{\sigma}$ is a total stress considering damage of the material obtained by Eq. (4.1). The calculation of the total stress and a stress update algorithm will be explained in the later section. We then assume that the external power ($\mathcal{W}$) acting upon $\mathcal{P} \subset \mathcal{B}$ has the form as,

$$\mathcal{W}(\mathcal{P}) = \int_{\partial\mathcal{P}} \mathbf{t}(\mathbf{n}) \cdot \dot{\mathbf{u}} dA + \int_{\mathcal{P}} \mathbf{b} \cdot \dot{\mathbf{u}} dV + \sum_i \left(\int_{\partial\mathcal{P}} \chi_i(\mathbf{n}) \cdot \dot{d}_i dA + \int_{\mathcal{P}} \lambda_i \dot{d}_i dV \right), \quad (4.12)$$

where $\mathbf{t}$ is the traction vector with outward unit normal $\mathbf{n}$; $\mathbf{b}$ is the macroscopic body force; χ_i and λ_i are the microscopic external traction and body force, respectively, both power-conjugate to $\dot{d}_i$. Note that a scalar external virtual microscopic force power conjugate to $\dot{\gamma}^{\alpha}$ is not considered [[Gurtin et al., 2010](#)]. Additional assumption is made such that each field, that is $\dot{\mathbf{u}}$, $\dot{\boldsymbol{\epsilon}}^e$, $\dot{\gamma}^{\alpha}$, and $\dot{d}_i$, is known at some arbitrary known but fixed time, and can be independently specified within the kinematic constraints (Eqs. (4.3) to (4.5)) as

$$\nabla^{\text{sym}} \tilde{\mathbf{u}} = \tilde{\boldsymbol{\epsilon}}^e + \sum_{\alpha} \tilde{\gamma}^{\alpha} \mathbf{S}^{\alpha}. \quad (4.13)$$

By denoting each virtual field by $\tilde{\mathbf{u}}$, $\tilde{\boldsymbol{\epsilon}}^e$, $\tilde{\gamma}^{\alpha}$, and $\tilde{d}_i$, a generalized virtual velocity $\mathcal{V}$ can be defined by [[Gurtin et al., 2010](#), [Anand and Gurtin, 2003](#), [De Lorenzis et al., 2016](#)],

$$\mathcal{V} = (\tilde{\mathbf{u}}, \tilde{\boldsymbol{\epsilon}}^e, \tilde{\gamma}, \tilde{\mathbf{d}}). \quad (4.14)$$

Then the principle of virtual power is the requirement that, $\mathcal{J}(\mathcal{P}, \mathcal{V}) = \mathcal{W}(\mathcal{P}, \mathcal{V})$, that is,

$$\begin{aligned} & \int_{\mathcal{P}} \boldsymbol{\sigma} : \tilde{\boldsymbol{\epsilon}}^e dV + \sum_{\alpha} \int_{\mathcal{P}} \pi^{\alpha} \tilde{\gamma}^{\alpha} dV + \sum_i \left(\int_{\mathcal{P}} \boldsymbol{\xi}_i \cdot \nabla \tilde{d}_i dV + \int_{\mathcal{P}} \pi_i \tilde{d}_i dV \right) \\ &= \int_{\partial\mathcal{P}} \mathbf{t}(\mathbf{n}) \cdot \tilde{\mathbf{u}} dA + \int_{\mathcal{P}} \mathbf{b} \cdot \tilde{\mathbf{u}} dV + \sum_i \left(\int_{\partial\mathcal{P}} \chi_i(\mathbf{n}) \cdot \tilde{d}_i dA + \int_{\mathcal{P}} \lambda_i \tilde{d}_i dV \right), \end{aligned} \quad (4.15)$$

for all virtual velocities $\mathcal{V}$. Again, the derivation of balance equations are straight forward (e.g., [[Gurtin et al., 2010](#), [Anand and Gurtin, 2003](#), [De Lorenzis et al., 2016](#)]), thus resulting in the following final expressions.

$$\nabla \cdot \boldsymbol{\sigma} + \mathbf{b} = \mathbf{0}, \quad (\text{Linear momentum}) \quad (4.16)$$

$$\mathbf{t}(\mathbf{n}) = \boldsymbol{\sigma} \cdot \mathbf{n}, \quad (\text{Macroscopic traction}) \quad (4.17)$$

$$\pi^\alpha = \tau^\alpha \text{ with } \tau^\alpha = \mathbf{S}^\alpha : \boldsymbol{\sigma}, \quad (\text{Microscopic force balance for each slip system}) \quad (4.18)$$

$$\nabla \cdot \boldsymbol{\xi}_i - \pi_i + \lambda_i = 0, \quad (\text{Microscopic force balance for each phase field}) \quad (4.19)$$

$$\chi_i(\mathbf{n}) = \boldsymbol{\xi}_i \cdot \mathbf{n}. \quad (\text{Phase-field microscopic traction}) \quad (4.20)$$

Here τ^α indicates the Schmidt stress or the resolved shear stress, which represents the macroscopic stress $\boldsymbol{\sigma}$ resolved on the slip system α . In addition, the number of Eqs. (4.19) and (4.20) are consistent with the number of each phase-field corresponding to the slip-system of a single crystal halite in this study. We further note that the macro and microforces are related based on Eq. (4.18) such that τ^α represents the force applied on the lattice of a single crystal for dislocations on slip system α while π^α constitutes internal forces on slip system α [Gurtin et al., 2010]. The multiple phase-field variables, however, does not show clear relations associated with the macroforce and the microforce on the slip systems. This interaction is established based on our choice of a free energy functional, which will be covered in the later section.

4.2.5 Energy balance equation and dissipation inequality

To capture the thermo-mechanical behaviors of a single crystal, we present an energy balance equation which includes contributions of heat conduction, mechanical dissipation, and structural heating. The first law (energy balance) can be expressed in a local form,

$$\dot{e} = \boldsymbol{\sigma} : \dot{\boldsymbol{\epsilon}} - \nabla \cdot \mathbf{q} + r_\theta, \quad (4.21)$$

where e is internal energy per unit volume; $\mathbf{q}$ is the heat flux vector; r_θ is the heat source term. This equation can be rewritten in greater detail by including the dissipation of the plastic slip and the work done due to the growth of phase-field (Eq. (4.15)). Recall that the rate of the phase field change is power conjugate to the microforce (cf. Gurtin [1996a], Borden [2012]). The balance of energy therefore reads,

$$\dot{e} = \boldsymbol{\sigma} : \dot{\boldsymbol{\epsilon}} + \sum_\alpha \pi^\alpha \dot{\gamma}^\alpha + \sum_i (\boldsymbol{\xi}_i \cdot \nabla \dot{d}_i + \pi_i \dot{d}_i) - \nabla \cdot \mathbf{q} + r_\theta, \quad (4.22)$$

Then the second law of thermodynamics (Clausius-Duhem inequality) is,

$$\mathcal{D}_{\text{int}} = \dot{\eta} + \nabla \cdot \left(\frac{\mathbf{q}}{\theta} \right) - \frac{r_\theta}{\theta} \geq 0, \quad (4.23)$$

in which η is an entropy term and θ is absolute temperature. Here the the flux vector ($\mathbf{q}$) can be expressed using Fourier's law for heat conduction,

$$\mathbf{q} = -\boldsymbol{\kappa} \cdot \nabla \theta, \quad (4.24)$$

where $\boldsymbol{\kappa}$ is the thermal conductivity tensor, which is assumed to be isotropic in this study. In addition, we may recall Helmholtz free energy (ψ) which shows the relation between the internal energy (e) and entropy (η) as,

$$\psi = e - \theta \eta. \quad (4.25)$$

Therefore, the dissipation inequality ($\mathcal{D}_{\text{int}}$) can be rewritten as,

$$\mathcal{D}_{\text{int}} = \boldsymbol{\sigma} : \dot{\boldsymbol{\epsilon}} + \sum_\alpha \pi^\alpha \dot{\gamma}^\alpha + \sum_i (\boldsymbol{\xi}_i \cdot \nabla \dot{d}_i + \pi_i \dot{d}_i) - (\eta \dot{\theta} + \dot{\psi}) - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta \geq 0. \quad (4.26)$$

Now we consider a free energy function for thermoelastoplastic crystal materials of the form,

$$\psi = \hat{\psi}(\epsilon^e, \underline{s}, \theta, \underline{d}, \nabla \underline{d}), \quad (4.27)$$

which includes the elastic strain ϵ^e ; temperature θ ; a set of crack phase-fields $\underline{d}$, its gradient and its rate; s indicates a local scalar measure related to the plastic slip accumulation for each slip system defined by,

$$\dot{s}^\alpha := \dot{\gamma}^\alpha \geq 0 \quad \text{where} \quad s^\alpha(t) = \int_0^t \dot{\gamma}^\alpha d\tau. \quad (4.28)$$

The substitution of Eq. (4.27) into Eq. (6.61) induces the dissipation inequality as the following form,

$$\begin{aligned} & \left(\sigma - \frac{\partial \hat{\psi}}{\partial \epsilon^e} \right) : \dot{\epsilon}^e + \sum_\alpha \pi^\alpha \dot{\gamma}^\alpha - \sum_\alpha \frac{\partial \hat{\psi}}{\partial s^\alpha} \dot{s}^\alpha + \sum_i \left(\pi_i - \frac{\partial \hat{\psi}}{\partial d_i} \right) \dot{d}_i \\ & + \sum_i \left(\xi_i - \frac{\partial \hat{\psi}}{\partial \nabla d_i} \right) \cdot \nabla \dot{d}_i - \left(\eta + \frac{\partial \hat{\psi}}{\partial \theta} \right) \dot{\theta} - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta \geq 0. \end{aligned} \quad (4.29)$$

Well-known arguments (e.g., Standard Coleman–Noll arguments) lead to the constitutive relations for the Cauchy stress and entropy, respectively,

$$\sigma = \frac{\partial \hat{\psi}}{\partial \epsilon^e}, \quad \eta = - \frac{\partial \hat{\psi}}{\partial \theta}. \quad (4.30)$$

We then further derive the dissipation inequality by following the individual group terms. The inequality related the each multiple phase-field variable can be expressed as,

$$\sum_i \left(\pi_i - \frac{\partial \hat{\psi}}{\partial d_i} \right) \dot{d}_i + \sum_i \left(\xi_i - \frac{\partial \hat{\psi}}{\partial \nabla d_i} \right) \cdot \nabla \dot{d}_i \geq 0, \quad (4.31)$$

which leads to the phase-field microscopic constitutive equations as,

$$\pi_i = \frac{\partial \hat{\psi}}{\partial d_i} \quad \text{and} \quad \xi_i = \frac{\partial \hat{\psi}}{\partial \nabla d_i}. \quad (4.32)$$

Substitution of Eqs. (4.32) into Eq. (4.19) with the assumptions of no microscopic body force ($\lambda_i = 0$) leads to the multiple phase-field evaluation equations as,

$$\nabla \cdot \left(\frac{\partial \hat{\psi}}{\partial \nabla d_i} \right) - \frac{\partial \hat{\psi}}{\partial d_i} = 0. \quad (4.33)$$

Note that the rate independent phase-field evolution has been assumed in this study. The derivation for rate-dependent case can be further obtained by modifying the free energy function (Eq. (4.27)), which is represented in Ambati et al. [2015]. Based on Eqs. (4.30) and (4.32), therefore, the reduced dissipation inequality becomes,

$$\underbrace{\sum_\alpha \pi^\alpha \dot{\gamma}^\alpha - \sum_\alpha \frac{\partial \hat{\psi}}{\partial s^\alpha} \dot{s}^\alpha}_{D_p} - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta \geq 0, \quad (4.34)$$

where D_p denotes the plastic dissipation rate. Considering the definition of g^α the inequality (4.34) can be further reduced to,

$$\sum_{\alpha} (\pi^\alpha \dot{\gamma}^\alpha + g^\alpha \dot{s}^\alpha) - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta \geq 0, \quad \text{where } g^\alpha = -\frac{\partial \hat{\psi}}{\partial s^\alpha}, \quad (4.35)$$

in which the thermodynamic force g^α power-conjugate to $\dot{s}^\alpha$ is introduced.

Finally, we can obtain the balance of energy in temperature form using the the internal energy balance equation by substituting the free energy relation of Eq. (4.25) into Eq. (4.22):

$$\dot{\psi} + \eta \dot{\theta} + \eta \dot{\theta} - \boldsymbol{\sigma} : \dot{\boldsymbol{\epsilon}}^e - \sum_{\alpha} \pi^\alpha \dot{\gamma}^\alpha - \sum_i (\xi_i \cdot \nabla \dot{d}_i + \pi_i \dot{d}_i) + \nabla \cdot \mathbf{q} - r_\theta = 0. \quad (4.36)$$

This equation can be rewritten by combining the constitutive relations of Eqs. (4.30) and (4.32) as,

$$\begin{aligned} -\theta \frac{\partial^2 \hat{\psi}}{\partial \theta^2} \dot{\theta} &= \sum_{\alpha} (\pi^\alpha \dot{\gamma}^\alpha + g^\alpha \dot{s}^\alpha) + \sum_{\alpha} \left(\theta \frac{\partial^2 \hat{\psi}}{\partial \theta \partial s^\alpha} \dot{s}^\alpha \right) + \sum_i \left(\frac{\partial^2 \hat{\psi}}{\partial \theta \partial d_i} \dot{d}_i + \frac{\partial^2 \hat{\psi}}{\partial \theta \partial \nabla d_i} \cdot \nabla \dot{d}_i \right) \theta \\ &\quad + \theta \left(\frac{\partial^2 \hat{\psi}}{\partial \theta \partial \boldsymbol{\epsilon}^e} : \dot{\boldsymbol{\epsilon}}^e \right) - \nabla \cdot \mathbf{q} + r_\theta. \end{aligned} \quad (4.37)$$

Considering a choice of a free energy function in this study Eq. (4.40), we may reduce the equation as,

$$\underbrace{-\theta \frac{\partial^2 \hat{\psi}}{\partial \theta^2} \dot{\theta}}_{c_v} = \underbrace{\sum_{\alpha} (\pi^\alpha \dot{\gamma}^\alpha + g^\alpha \dot{s}^\alpha) + \theta \left(\frac{\partial^2 \hat{\psi}}{\partial \theta \partial \boldsymbol{\epsilon}^e} : \dot{\boldsymbol{\epsilon}}^e \right)}_{D_{\text{mech}} - H_\theta} - \nabla \cdot \mathbf{q} + r_\theta, \quad (4.38)$$

which can be written in a simplified expression by [Simo and Miehe \[1992\]](#),

$$c_v \dot{\theta} = [D_{\text{mech}} - H_\theta] - \nabla \cdot \mathbf{q} + r_\theta. \quad (4.39)$$

Note that the specific choice of free energy comprises the energy balance equation. Here c_v is the specific heat per unit volume at constant deformation; D_{mech} denotes the contribution to the dissipation due to pure mechanical load and/or thermal flow, which may be consistent in the form of $\beta \boldsymbol{\sigma} : \dot{\boldsymbol{\epsilon}}^p$ with β of Taylor-Quinney coefficient; H_θ is the non-dissipative (latent) thermoelastic structural heat or cooling. The influence of D_{mech} and H_θ in the energy balance equation may be significant or negligible depending on how the problem conditions and material properties are defined.

4.2.6 A specific free energy functional

We derive the explicit expressions for the balance equations by choosing the stored energy functional:

$$\psi = \hat{\psi}^e(\boldsymbol{\epsilon}^e, \theta, \underline{d}) + \hat{\psi}^p(\underline{s}, \underline{d}) + \hat{\psi}^c(\underline{d}, \nabla \underline{d}, \underline{\omega}) + \hat{\psi}^\theta(\theta). \quad (4.40)$$

The elastic part $\hat{\psi}^e$ is composed of the thermoelastic strain energy (w^e) considering the thermal expansion (e.g., [Anand and Gurtin \[2003\]](#), [Abeyaratne et al. \[1994\]](#), [Lubarda](#)

[2004], Yang et al. [2006], Peigney and Seguin [2013], Miehe et al. [2015a]), which is multiplied by the degradation function $g(\underline{d})$ for damage evolution:

$$\hat{\psi}^e(\epsilon^e, \theta, \underline{d}) = g(\underline{d})w^e(\epsilon^e, \theta) \quad \text{with} \quad w^e(\epsilon^e, \theta) = \frac{1}{2}\epsilon^e : \mathbb{C}^e : \epsilon^e - \alpha(\theta - \theta_0) \text{tr} \epsilon^e, \quad (4.41)$$

Firstly, the degradation function $g(\underline{d})$ for multiple phase-field can be assumed to have a simple form [Nguyen et al., 2017a,b] as,

$$g(\underline{d}) = (1 - k) \prod_i (1 - d_i)^2 + k. \quad (4.42)$$

This function has been chosen such that $g'(d_i = 1) = 0$ to guarantee that the strain energy density function takes a finite value as the domain is locally cracked. The small parameter $k \ll 1$ is introduced for maintaining the well-posedness of the problem for partially broken part of the domain (cf. Nguyen et al. [2017a,b], Heister et al. [2015]). Next $\mathbb{C}^e$ denotes the fourth-order elasticity tensor, which may constitute the isotropic or cubic symmetry for crystalline materials. α is the thermal expansion coefficients for isotropic behavior, and θ_0 denotes a fixed reference temperature. To avoid the crack propagations under compression, the elastic strain can be further decomposed into the positive part w_+^e and negative part w_-^e by the spectral decomposition or the volumetric and deviatoric split (e.g., Nguyen et al. [2017b], Ambati et al. [2015], Miehe et al. [2015a], Borden et al. [2016a]).

$$\hat{\psi}^e(\epsilon^e, \theta, \underline{d}) = g(\underline{d})w_+^e + w_-^e, \quad (4.43)$$

where the description of w_+^e and w_-^e for crystalline materials will be revisited in the following section.

We then take the contribution of plastic work for crack growth into account to simulate the ductile fracture behavior. The simple equation for the plastic work may have the form as:

$$\hat{\psi}^p(\underline{s}, \underline{d}) = g(\underline{d})^p \langle w^p - w_0^p \rangle, \quad \text{where} \quad g(\underline{d})^p = g(\underline{d}) \quad \text{and} \quad w^p = \frac{1}{2} \sum_{\alpha} h(s^{\alpha})^2. \quad (4.44)$$

Here the degradation function for plastic work is assumed to be identical to that of the elastic work, and a plastic work threshold w_0^p may be introduced for controlling the plastic deformation in ductile fracture (e.g., Miehe et al. [2015a], Borden et al. [2016a]). The angle bracket operator is defined as,

$$\langle x \rangle = \begin{cases} 0 & \text{if } x < 0 \\ x & \text{if } x \geq 0 \end{cases}. \quad (4.45)$$

The plastic work ω^p comes from the hardening contribution, which is consistent with the Taylor hardening in Eq. (4.58). Therefore, the thermodynamic force g^{α} power-conjugate to $\dot{s}^{\alpha}$ in Eq. (4.35) can be explicitly described as,

$$g^{\alpha} = - \sum_{\alpha} h s^{\alpha}. \quad (4.46)$$

The stored energy function $\hat{\psi}^c(\underline{d}, \nabla \underline{d}, \underline{\omega})$ describes the total anisotropic crack surface energy via the multiple phase-field method. The total crack density formulation is previously defined by Eq. (4.10) based on the diffusive crack topology using the multiple phase-field approximation (Section 4.2.3). By assuming the same fracture energy for each

slip direction (or the same energy release rate, G_c), the total crack energy function can be expressed as,

$$\hat{\psi}^c = G_c \sum_i \left[\frac{1}{2l} (d_i)^2 + \frac{l}{2} \boldsymbol{\omega}_i : (\nabla d_i \otimes \nabla d_i) \right]. \quad (4.47)$$

The purely thermal contribution on the stored energy function $\hat{\psi}^\theta$ constitutes the heat transfer, in which the equation of $\hat{\psi}^\theta$ may have a simple form as (e.g., [Abeyaratne et al. \[1994\]](#), [Lubarda \[2004\]](#), [Yang et al. \[2006\]](#), [Peigney and Seguin \[2013\]](#), [Miehe et al. \[2015a\]](#), [Bartels et al. \[2015\]](#), [Canadija and Mosler \[2011\]](#)),

$$\hat{\psi}^\theta = c_v [(\theta - \theta_0) - \theta \ln(\theta/\theta_0)], \quad (4.48)$$

where c_v is the specific heat defined in Eq. (4.37). The specific heat is assumed to be a constant for a single crystal halite based on [Urquhart and Bauer \[2015\]](#). In addition, we simplify the thermal-mechanical-fracture coupling problems by assuming that the stored thermal energy is not affected by fracture (e.g., [Miehe et al. \[2015a\]](#)).

Finally, the microscopic multiple phase-field evolution equation for each phase of Eq. (4.33) now becomes,

$$2(1 - d_i) \mathcal{H}_i + \frac{G_c}{l} d_i + G_c l \nabla \cdot (\boldsymbol{\omega}_i \cdot \nabla d_i) = 0, \quad (4.49)$$

where $\mathcal{H}_i$ is the strain history functional introduced to handle the irreversible crack propagations (cf. [Nguyen et al. \[2017a,b\]](#)). This parameter can be expressed by,

$$\mathcal{H}_i = \max_{\tau \in [0, t]} \left\{ \prod_{j \neq i} (1 - d_j)^2 [w_+^e + \langle w^p - w_0^p \rangle] \right\}. \quad (4.50)$$

4.3 Constitutive law

In this section, we present the constitutive law to capture the anisotropic behavior of crystalline materials. We combine the multiple phase-field method and the single crystal plasticity theory to describe the elasto-plastic behavior coupled with the damage evolutions. Provided the phase-field approach for brittle fracture can be conceived as a special gradient damage model [[de Borst and Verhoosel, 2016a](#)], we borrow this concept and extend it to the multiple phase-field method. As a result, the constitutive equations are calculated on a effective stress space based on hypothesis of strain equivalence (e.g., [Lemaitre \[1984\]](#), [Simo and Ju \[1987\]](#), [Ju \[1989\]](#), [de Borst et al. \[1999\]](#)), which allows one to use the existing constitutive models for undamaged materials.

The effective stress theory hypothesizes that there exists an effective stress space where local stresses are redistributed to the effective area such as undamaged skeleton of the body, undamaged material micro-bonds, the vicinity of growing voids, etc. Therefore the effective stress are higher than the nominal stresses. In addition, the hypothesis of strain equivalence states that the strain behavior of a damaged material by the applied stress is equivalent to the strain associated with its undamaged state under the effective stress. The local constitutive equations, therefore, can be resolved on the effective space, followed by the total stress updates for a damaged state.

Recalling the specific free energy function Eqs. (4.40) and (4.41) with the constitutive relation Eq. (4.30) reaches that the Cauchy stress, or total stress, can be resolved as,

$$\boldsymbol{\sigma} = \frac{\partial \hat{\psi}}{\partial \boldsymbol{\epsilon}^e} = g(\underline{d}) \hat{\boldsymbol{\sigma}} \quad \text{with} \quad \hat{\boldsymbol{\sigma}} = \mathbb{C}^e : \boldsymbol{\epsilon}^e - \alpha(\theta - \theta_0) \mathbf{1}, \quad (4.51)$$

where $\hat{\sigma}$ denotes the effective stress, or damage effective stress. It should be noted that the term effective stress of this study is different from that of porous media, in which the effective stress indicates the stress of the solid skeleton when saturated or partially saturated with fluid. (e.g., Sun [2015a], Coussy [2004], Sun [2013a], Sun et al. [2013a], Wang and Sun [2015], Na and Sun [2016, 2017a]). The stress update algorithm for the multiple phase-field approach coupled to crystal plasticity is described in Algorithm 1.

4.3.1 Elasticity of single-crystal halite

In this study the elastic response of single-crystal halite is described by the fourth-order elasticity tensor $\mathbb{C}^e$ as in Eqs. (4.41) and (4.51). As a crystal structure having cubic symmetry, single-crystal halite can be represented by three elastic constant, C_{11} , C_{12} , and C_{44} , when x , y , and z axes are aligned with the crystallographic axes (e.g., Hirth and Lothe [1982], Wachtman et al. [2009]). Apart from the anisotropic features originated from the microstructure, the elastic behavior of halite depends on temperature, pressure, and loading conditions (e.g., Durand [1936], Bartels and Schuele [1965]). Therefore, the experimental set-up with sample preparation process is an integral part to estimate the elastic parameters. There has been many efforts to measure the elastic constants of natural rock salt, and according to Birch [1978], the ultrasonics, one of the dynamical methods, may be the best technique to estimate the elastic properties of halite including its aggregates. One of the pioneering works was proposed by Oliver and Pharr [1992], in which the load-displacement data from indentation experiments is used for determining elastic modulus of materials. The proper estimation of elastic parameters for halite is out of the scope of this study, and for computational efficiency, we assume that the elasticity of halite is linear and isotropic. The assumed average bulk modulus (K) and Poisson's ratio (ν), which are obtained from single-crystal data [Carter and Norton, 2007], are presented in Table 4.1.

The thermoelastic strain energy (w^e) in Eq. (4.41) can be rewritten via tensor notation as,

$$w^e = \frac{1}{2}K(\epsilon_v^e)^2 + \mu(\epsilon_d^e : \epsilon_d^e) - 3\alpha K(\theta - \theta_0)\epsilon_v^e, \quad \text{with} \quad \epsilon^e = \frac{1}{3}\epsilon_v^e \mathbf{1} + \epsilon_d^e, \quad (4.52)$$

where K and μ are the elastic bulk and shear moduli, respectively, $\epsilon_v^e = \text{tr} \epsilon^e$ is the volumetric strain, and ϵ_d^e is the deviatoric strain tensor. By decomposing the elastic strain into volumetric and deviatoric contributions, we can further separate the thermoelastic strain energy as follows to prevent unrealistic crack propagation under compression:

$$\begin{cases} w_+^e := \frac{1}{2}K\langle\epsilon_v^e\rangle_+^2 + \mu(\epsilon_d^e : \epsilon_d^e) - 3\alpha K(\theta - \theta_0)\langle\epsilon_v^e\rangle_+, \\ w_-^e := \frac{1}{2}K\langle\epsilon_v^e\rangle_-^2 - 3\alpha K(\theta - \theta_0)\langle\epsilon_v^e\rangle_- \end{cases}, \quad (4.53)$$

where $\langle\cdot\rangle_{\pm} := \frac{1}{2}(\cdot \pm |\cdot|)$. This decomposition using volumetric and deviatoric strains is proposed by Amor et al. [2009a], while the positive and negative components based the spectral decomposition can be found Miehe et al. [2010b].

4.3.2 Single crystal plasticity

This section presents the stress update algorithm for single-crystal plasticity on effective stress space. The unified fully implicit return algorithm for both rate-independent and rate-dependent settings are implemented based on Miehe and Schröder [2001]. In this

framework a multisurface-type model with elastic domain is used as in [Cuitino and Ortiz \[1993\]](#), and the power law-type viscoplastic slip-rate is introduced for single crystal halite. This slip-rate equation takes temperature and rate-dependent effects into account to describe the dislocation creep. For the rate independent limit, the pseudo-inverse method based on the spectral value decomposition (SVD) is used for inverse of jacobian matrix in the constitutive equations (e.g., [Anand and Kothari \[1996\]](#), [Miehe and Schröder \[2001\]](#)).

Followed by Eq. (4.3), we first denote by the homogeneous strain rate, $\dot{\epsilon}$, in a crystal and consider its additive decomposition,

$$\dot{\epsilon} = \dot{\epsilon}^e + \dot{\epsilon}^p, \quad (4.54)$$

into elastic ($\dot{\epsilon}^e$) and plastic ($\dot{\epsilon}^p$) parts, respectively. The plastic component comes from slips on crystallographic planes and results in the following expression (cf. Eq. (4.5)) :

$$\dot{\epsilon}^p = \sum_{\alpha} \dot{\gamma}^{\alpha} S^{\alpha}. \quad (4.55)$$

Again, $\dot{\gamma}^{\alpha}$ and S^{α} denote the plastic slip rate and the symmetric part of the Schmid tensor on α -slip system, respectively.

Now we present the stress update algorithm for a crystal with $2N$ potentially active slip systems, $\mathcal{J} := \{1, 2, \dots, 2N\}$. Considering the microforce balance equation (4.18) for a slip system and crystal plasticity theory (e.g., [de Souza Neto et al. \[2011\]](#), [Miehe and Schröder \[2001\]](#), [Borja and Rahmani \[2012\]](#)), the yield condition can be expressed as a function of the Schmid resolved shear stress (τ^{α}) and the critical resolved shear stress (τ_Y^{α}) for each slip system. Here we set up the yield condition based on the effective stress and critical resolved shear stress (e.g., [Ekh et al. \[2004\]](#), [Qi and Bertram \[1999\]](#)),

$$f^{\alpha} = \tau^{\alpha} - \tau_Y^{\alpha}, \quad \text{where} \quad \tau^{\alpha} = \hat{\sigma} : S^{\alpha}. \quad (4.56)$$

Then a non-smooth convex elastic domain in the stress space can be defined as,

$$\mathbb{E} = \{(\hat{\sigma}, \tau_Y^{\alpha}) \mid f^{\alpha} \leq 0 \text{ for } \alpha = 1, 2, \dots, 2N\}. \quad (4.57)$$

The evolution of these resistances within an multislip deformation process are governed by the hardening equations,

$$\dot{\tau}_Y^{\alpha} = h \sum_{\beta=1}^{2N} \dot{\gamma}^{\beta}, \quad (4.58)$$

in which the hardening of the yield surface is characterized by a simple Taylor hardening law that introduces only one additional material parameter, the plastic modulus (h). Here we rewrite the plastic strain rate in Koiters's form for rate-dependent single-crystal plasticity as,

$$\dot{\epsilon}^p = \sum_{\alpha=1}^{2N} \dot{\gamma}^{\alpha} \frac{\partial f^{\alpha}}{\partial \hat{\sigma}} = \sum_{\alpha=1}^{2N} \dot{\gamma}^{\alpha} S^{\alpha}, \quad (4.59)$$

where the slip rates $\dot{\gamma}^{\alpha}$ satisfy a constitutive viscoplastic form of loading conditions to incorporate plastic flow due to dislocation creep motion in single-crystal halite as (cf. [Hansen et al. \[2014b\]](#), [Miehe and Schröder \[2001\]](#), [Wawersik and Zeuch \[1984\]](#)):

$$\dot{\gamma}^{\alpha} = C_0 \exp\left(-\frac{Q}{RT}\right) \left[\left(\frac{f^{\alpha+}}{\tau_Y^{\alpha}} + 1\right)^p - 1\right]. \quad (4.60)$$

Here C_0 represents a fitting parameter; Q is the activation energy; R is the universal gas constant; T is the temperature in Kelvine; p is a strain-rate-sensitivity exponent. The overstress function $f^{\alpha+}$ is defined by

$$f^{\alpha+} := \begin{cases} f^\alpha & \text{if } f^\alpha > 0 \\ 0 & \text{otherwise} \end{cases} \quad (4.61)$$

The slip rate $\dot{\gamma}^\alpha$ is reformulated to consider elastic region using a classical viscoplastic form based on the strain rate equation. We take the dislocation creeping observed in the experiment (e.g., [Wawersik and Zeuch \[1984\]](#)) into account the slip rate at each slip system to incorporate the rate and temperature dependence in single-crystal halite. The dislocation creep equation obtained from the experiment, however, is not for a microscopic slip-system response but for a macro response. We may reduce this discrepancy by material parameters calibration through resolving the inverse problem. The brief explanation about this procedure is discussed in the appendix.

The unified stress update algorithm on effective stress space is outlined for both rate-independent and rate-dependent crystal plasticity frameworks. Algorithm 1 describes the elastic predictor phase on effective space, while the plastic corrector and determination of active slip systems are performed in Algorithm 2. For the rate-dependent case the viscoplastic regularization of single-crystal plasticity for halite Eq. (4.60) is used. In the rate-independent limit, the slip rates $\dot{\gamma}^\alpha$ satisfy the classical Kuhn-Tucker conditions (e.g., [Miehe and Schröder \[2001\]](#), [Borja and Rahmani \[2012\]](#), [Budiansky and Wu \[1961\]](#), [Simo and Hughes \[2006\]](#)).

$$\dot{\gamma}^\alpha \geq 0, \quad f^\alpha \leq 0, \quad \dot{\gamma}^\alpha f^\alpha = 0, \quad (4.62)$$

for all α . Furthermore, the inverse of Jacobian ($\mathbf{D}$) is resolved by means of pseudo-inverse approach based the singular value decomposition (SVD). For brevity we drop the subscript $n + 1$ for variables pertaining to the time t_{n+1} . Note again that $\mathcal{J}$ denotes all potentially active slip systems in a crystal with $2N$.

Algorithm 1 Elastic predictor phase on effective stress space

- 1: At integration point compute strain increments $\Delta \epsilon$
 - 2: Compute effective trial stress $\hat{\sigma}^{\text{tr}} = \hat{\sigma}_n + \mathbf{C}^e : \Delta \epsilon$
 - 3: Assemble trial active set $\mathcal{A}^{\text{tr}} = \{\alpha \in \mathcal{J} \mid \hat{\sigma}^{\text{tr}} : \mathbf{S}^\alpha - \tau_{Y,n}^\alpha > 0\}$
 - 4: **if** $\mathcal{A}^{\text{tr}} = \emptyset$ **then**
Elastic response: set $\hat{\sigma} = \hat{\sigma}^{\text{tr}}$, $\tau_Y^\alpha = \tau_{Y,n}^\alpha$, $\mathcal{A} = \emptyset$,
 $\mathbf{C} = \mathbf{C}^e$, and exit
 - 5: **else**
Plastic response: call Algorithm 2
Update $\hat{\sigma}$, τ_Y^α , and $\mathcal{A}$
 $\mathbf{C} \leftarrow \mathbf{C}^{ep}$, and exit
 - 6: **end if**
 - 7: Resolve total stress $\sigma = g(\underline{d})\hat{\sigma}$
-

In Algorithm 1 we assemble the trial active set by checking the yield conditions for each slip system Eq. (4.56) under the given temperature θ condition. When the trial active set is blank set, the material is in the purely elastic regime. If the trial active set contains any element, Algorithm 2 is triggered and the Newton iteration for the plastic response is performed. With in this iteration step, the active slip system is successively checked and reassembled followed by two constraints. First, we check γ^α in the trial active set

Algorithm 2 Plastic integrator and determination of active slip systems

- 1: Initialize active set $\mathcal{A} = \mathcal{A}_n$
- 2: Set initial values for plastic slip iteration: $\gamma^\alpha = 0 \quad \forall \alpha \in \mathcal{J}$
- 3: Get current effective stress: $\hat{\sigma} = \hat{\sigma}^{\text{tr}} - \mathbb{C}^e : \left(\sum_{\alpha=1}^{2N} \gamma^\alpha \mathbf{S}^\alpha \right)$
- 4: Compute the residuals for active slips systems ($\alpha, \beta \in \mathcal{A}$) and construct the Jacobian ($\mathbf{D}$)

$$r^\alpha = \hat{\sigma} : \mathbf{S}^\alpha - \tau_Y^\alpha \sqrt{\frac{\eta}{\Delta t} \gamma^\alpha + 1}$$

$$\mathbf{D}^{\alpha\beta} = \mathbf{S}^\alpha : \mathbb{C}^e : \mathbf{S}^\beta + h \left(\frac{\eta}{\Delta t} \gamma^\alpha + 1 \right)^{1/p} + \tau_Y^\alpha \delta^{\alpha\beta} \frac{\eta}{p \Delta t} \left(\frac{\eta}{\Delta t} \gamma^\alpha + 1 \right)^{1/p-1}$$

$$\text{with } \tau_Y^\alpha = \tau_{Y,n}^\alpha + \sum_{\alpha \in \mathcal{A}} h \gamma^\alpha; \quad \eta = 1 / \left[C_0 \exp \left(-\frac{Q}{RT} \right) \right]$$

- 5: **if** $\mathbf{D}$ is singular **then**
 Compute singular value decomposition and obtain pseudo inverse: $\bar{\mathbf{D}}^{-1} = \mathbf{V} \bar{\boldsymbol{\Sigma}}^{-1} \mathbf{U}^T$
- 6: **else** Set $\bar{\mathbf{D}}^{-1} = \mathbf{D}^{-1}$ based on standard inversion
- 7: **end if**
- 8: Update incremental plastic slip: $\gamma^\alpha \leftarrow \gamma^\alpha + \sum_{\alpha \in \mathcal{A}} (\mathbf{D}^{-1})^{\alpha\beta} r^\beta$
- 9: **If** $\left(\sqrt{\sum_{\alpha \in \mathcal{A}} [r^\alpha]^2} > \text{tol} \right)$ **go to** 3
- 10: Check the minimum loaded system (I) :
- 11: **if** $\gamma^\alpha \leq 0$ for some $\alpha \in \mathcal{A}$ **then** $\mathcal{A} \leftarrow \{\mathcal{A} \setminus \alpha^*\}$ and **go to** 2
 where $\gamma^{\alpha^*} = \min[\gamma^\alpha] \quad \forall \alpha \in \mathcal{A}$
- 12: **else** **go to** 15
- 13: **end if**
- 14: Check the maximum loaded system (II):
- 15: **if** $f^\alpha > 0$ for some $\alpha \in \mathcal{J} \setminus \mathcal{A}$ **then** $\mathcal{A} \leftarrow \{\mathcal{A} \cup \alpha^*\}$ and **go to** 2
 where $f^{\alpha^*} = \max[f^\alpha] \quad \forall \alpha \in \mathcal{J} \setminus \mathcal{A}$
- 16: **else** Construct the consistent tangent-moduli:

$$\mathbb{C}^{ep} := \mathbb{C}^e - \sum_{\alpha \in \mathcal{A}} \sum_{\beta \in \mathcal{A}} (\mathbf{D}^{-1})^{\alpha\beta} (\mathbb{C}^e : \mathbf{S}^\alpha) \otimes (\mathbf{S}^\beta : \mathbb{C}^e)$$

- 17: **end if**
 - 18: Return to Algorithm 1
-

whether each parameter violates the discrete loading conditions (Eqs. (4.60) and (4.62)) in the sense $\gamma^\alpha \leq 0$. If this is the case, we drop the slip system from the active set $\mathcal{A}$ and restart the local Newton iteration. Second, we check the yield conditions again for the slip systems not in the current trial active set. We then reassemble the active set and restart the local Newton iteration by initializing $\gamma^\alpha = 0$ for all $\alpha \in \mathcal{A}$. Otherwise, Algorithm 2 is terminated and the updated parameters are forwarded to Algorithm 1 for the total stress calculation.

4.4 Variational formulation

This section describes a finite element formulation that combines multiple phase-field and crystal plasticity for modeling thermo-mechanical behaviors of a single crystal halite. We first derive the variational form of thermo-mechanical problems with multiple phase fields representing damage along preferential directions. This is followed by the description of a staggered scheme for solving the equilibrium (momentum and energy balances) and multiple phase-field equations in Section 4.5.

4.4.1 Galerkin form

We consider a domain $\mathcal{B}$ with its boundary $\partial\mathcal{B}$ composed of Dirichlet boundaries (solid displacement $\partial\mathcal{B}_u$, temperature $\partial\mathcal{B}_\theta$) and von Neumann boundaries (solid traction $\partial\mathcal{B}_t$, heat flux $\partial\mathcal{B}_q$) satisfying,

$$\begin{cases} \partial\mathcal{B} = \overline{\partial\mathcal{B}_u \cup \partial\mathcal{B}_t} = \overline{\partial\mathcal{B}_\theta \cup \partial\mathcal{B}_q} \\ \emptyset = \partial\mathcal{B}_u \cap \partial\mathcal{B}_t = \partial\mathcal{B}_\theta \cap \partial\mathcal{B}_q. \end{cases} \quad (4.63)$$

The prescribed boundary conditions (Dirichlet and von Neumann boundary conditions) read,

$$\begin{cases} \mathbf{u} = \bar{\mathbf{u}} & \text{on } \partial\mathcal{B}_u \\ \boldsymbol{\sigma} \cdot \mathbf{n} = \bar{\mathbf{t}} & \text{on } \partial\mathcal{B}_t \\ \theta = \bar{\theta} & \text{on } \partial\mathcal{B}_\theta \\ -\mathbf{n} \cdot \mathbf{q} = \bar{q} & \text{on } \partial\mathcal{B}_q, \end{cases} \quad (4.64)$$

where $\mathbf{n}$ is outward unit normal on surface $\partial\mathcal{B}$. For model closure, the initial conditions are imposed as,

$$\mathbf{u} = \mathbf{u}_0, \quad \theta = \theta_0 \quad \text{at } t = t_0. \quad (4.65)$$

In addition, we consider the trial space for the weak form that reads,

$$V_u = \left\{ \mathbf{u} : \mathcal{B} \rightarrow \mathbb{R}^3 \mid \mathbf{u} \in [H^1(\mathcal{B})], \mathbf{u}|_{\partial\mathcal{B}_u} = \bar{\mathbf{u}} \right\}, \quad (4.66)$$

$$V_\theta = \left\{ \theta : \mathcal{B} \rightarrow \mathbb{R} \mid \theta \in H^1(\mathcal{B}), \theta|_{\partial\mathcal{B}_\theta} = \bar{\theta} \right\}, \quad (4.67)$$

$$V_d = \left\{ \underline{d} : \mathcal{B} \rightarrow \mathbb{R} \mid \underline{d} \in H^1(\mathcal{B}) \right\}, \quad \text{i.e., } V_{d_i} = \left\{ d_i : \mathcal{B} \rightarrow \mathbb{R} \mid d_i \in H^1(\mathcal{B}) \right\}. \quad (4.68)$$

Here, H^1 denotes the Sobolev space of degree one. We further note that the multiple phase-field variables (or a set of phase-field values) are approximated in one element by d_i . The number of these variables depends on how we define preferential directions, which is consistent with the slip systems for a single crystal halite in this study. The corresponding admissible spaces of variations are defined as,

$$V_\eta = \left\{ \boldsymbol{\eta} : \mathcal{B} \rightarrow \mathbb{R}^3 \mid \boldsymbol{\eta} \in [H^1(\mathcal{B})], \boldsymbol{\eta}|_{\partial\mathcal{B}_u} = \mathbf{0} \right\}, \quad (4.69)$$

$$V_\psi = \left\{ \psi : \mathcal{B} \rightarrow \mathbb{R} \mid \psi \in H^1(\mathcal{B}), \psi|_{\partial\mathcal{B}_\theta} = 0 \right\}, \quad (4.70)$$

$$V_\phi = \left\{ \underline{\phi} : \mathcal{B} \rightarrow \mathbb{R} \mid \underline{\phi} \in H^1(\mathcal{B}) \right\} \quad \text{i.e., } V_{\phi_i} = \left\{ \phi_i : \mathcal{B} \rightarrow \mathbb{R} \mid \phi_i \in H^1(\mathcal{B}) \right\}. \quad (4.71)$$

Therefore, the weighted residual statement of the balance of linear momentum, energy, and phase-field are: find $\mathbf{u} \in V_u$, $\theta \in V_\theta$, and $\underline{d} \in V_\phi$ such that all $\boldsymbol{\eta} \in V_\eta$, $\psi \in V_\psi$, and

$$\underline{\phi} \in \underline{V}_\phi,$$

$$G(\mathbf{u}, \theta, \underline{d}, \underline{\eta}) = H(\mathbf{u}, \theta, \underline{d}, \psi) = \underline{L}(\mathbf{u}, \theta, \underline{d}, \underline{\phi}) = 0. \quad (4.72)$$

Consider the governing equations given by Eqs. (4.16), (4.37), and (4.49). Through the standard weighted residual procedure, we obtain the variational equations as,

$$\begin{aligned} G : V_u \times V_\theta \times \underline{V}_d \times V_\eta &\rightarrow \mathbb{R}, \\ G(\mathbf{u}, \theta, \underline{d}, \underline{\eta}) &= \int_{\mathcal{B}} \nabla \underline{\eta} : \sigma dV - \int_{\partial \mathcal{B}} \underline{\eta} \cdot \bar{\mathbf{t}} d\Gamma = 0, \end{aligned} \quad (4.73)$$

$$\begin{aligned} H : V_u \times V_\theta \times \underline{V}_d \times V_\psi &\rightarrow \mathbb{R}, \\ H(\mathbf{u}, \theta, \underline{d}, \psi) &= \int_{\mathcal{B}} \psi c_v \dot{\theta} dV - \int_{\mathcal{B}} \psi \left[\sum_{\alpha} (\pi^\alpha \dot{\gamma}^\alpha + g^\alpha \dot{s}^\alpha) \right] dV - \int_{\mathcal{B}} \psi (\alpha \mathbf{1} : \dot{\epsilon}^e) \theta dV \\ &\quad + \int_{\mathcal{B}} \nabla \psi \cdot \kappa \nabla \theta dV - \int_{\partial \mathcal{B}} \psi \bar{q} d\Gamma = 0, \end{aligned} \quad (4.74)$$

$$\begin{aligned} \underline{L} : V_u \times V_\theta \times \underline{V}_d \times \underline{V}_\phi &\rightarrow \mathbb{R}, \\ \underline{L}_i(\mathbf{u}, \theta, d_i, \phi_i) &= \int_{\mathcal{B}} \phi_i [2(1 - d_i) \mathcal{H}_i] dV + \int_{\mathcal{B}} \frac{G_c}{l} [\phi_i d_i + l^2 \nabla \phi_i \cdot \boldsymbol{\omega}_i \cdot \nabla d_i] dV, \end{aligned} \quad (4.75)$$

where $\mathcal{H}_i$ is previously defined by Eq. (4.50).

4.5 Operator-split solution strategies

Due to the non-linearity and path-dependence nature of the proposed model, linearizing the system of equations is necessary if an implicit solver is used. In the proposed work, the system of equations are multi-physical. As a result, this system of equations can be solved either in a monolithic or operator-split manner Kim et al. [2009], White et al. [2016a]. As previously numerical experiments with single phase field problems show that the operator splitting approach may potentially be more robustness (e.g., Miehe et al. [2010b], Heister et al. [2015]), we propose a semi-split iterative strategy in which the multiple phase fields are each advanced sequentially, while the thermo-mechanical solver which updates the displacement and temperature fields are advanced together. These sub-systems are iteratively updated until all the residuals are below the tolerance.

In the proposed operator-split setting, the crack driving force $\mathcal{H}_i$ – Eq. (4.50), is fixed while the multiphase field phase are updated. As a result, the split multiple phase-field incremental problem becomes linear. The schematic of solution strategies is summarized as follows:

$$\underbrace{\begin{bmatrix} \mathbf{u}_n \\ \theta_n \\ \underline{d}_n \end{bmatrix} \xrightarrow[\delta \underline{d}=0]{\mathcal{R}(\mathbf{u}, \theta)=0} \begin{bmatrix} \mathbf{u}_{n+1} \\ \theta_{n+1} \\ \underline{d}_n \end{bmatrix}}_{\text{Iterative solver}} \xrightarrow[\delta u=0, \delta \theta=0]{\substack{\text{Linear solver} \\ \mathcal{R}(\underline{d})=0 \text{ with } \mathcal{H}_{n+1}}} \begin{bmatrix} \mathbf{u}_{n+1} \\ \theta_{n+1} \\ \underline{d}_{n+1} \end{bmatrix}, \quad (4.76)$$

where $\mathcal{R}(\mathbf{u},)$ and $\mathcal{R}(\underline{d})$ are residuals expressed as follows:

$$\mathcal{R}(\mathbf{u}, \theta) : \begin{cases} \int_{\mathcal{B}} \nabla \boldsymbol{\eta} : \boldsymbol{\sigma}_{n+1} dV - \int_{\partial \mathcal{B}} \boldsymbol{\eta} \cdot \bar{\mathbf{t}}_{n+1} d\Gamma, \\ \int_{\mathcal{B}} \psi_{c_v} \left(\frac{\theta_{n+1} - \theta_n}{\Delta t} \right) dV - \int_{\mathcal{B}} \psi \left[\sum_{\alpha} \left(\pi_n^{\alpha} \left(\frac{\gamma_n^{\alpha} - \gamma_{n-1}^{\alpha}}{\Delta t} \right) + \delta_n^{\alpha} \left(\frac{s_n^{\alpha} - s_{n-1}^{\alpha}}{\Delta t} \right) \right) \right] dV \\ - \int_{\mathcal{B}} \psi \left[\alpha \mathbf{1} : \left(\frac{\boldsymbol{\epsilon}_n^e - \boldsymbol{\epsilon}_{n-1}^e}{\Delta t} \right) \right] \theta_{n+1} dV + \int_{\mathcal{B}} \nabla \psi \cdot \kappa \nabla \theta_{n+1} dV - \int_{\partial \mathcal{B}} \psi \bar{q}_{n+1} d\Gamma, \end{cases} \quad (4.77)$$

$$\mathcal{R}(d_i) : \left\{ \int_{\mathcal{B}} \phi_i [2(1 - d_{i_{n+1}}) \mathcal{H}_{i_{n+1}}] dV + \int_{\mathcal{B}} \frac{G_c}{l} [\phi_i d_{i_{n+1}} + l^2 \nabla \phi_i \cdot \boldsymbol{\omega}_i \cdot \nabla d_{i_{n+1}}] dV. \right. \quad (4.78)$$

These equations are consistent with Eqs. (4.73) to (4.75), in which the backward Euler method is used for time discretization for the heat transfer. Under the iterative solver in Eq. (4.76), the solutions are advanced followed by Algorithm 3. This procedure require

Algorithm 3 Solution strategies using iterative and linear solvers

Require: Compute $\mathbf{u}_{n+1}$, θ_{n+1} , and $\underline{d}_{n+1}$

- 1: Initialize: $k = 0$, $\mathbf{u}^k = \mathbf{u}_n$, $\theta^k = \theta_n$, and fix $\underline{d}_n$
 - 2: **while** $\| \mathcal{R}(\mathbf{u}, \theta) \| \geq \text{Tolerance}$ **do**
 - 3: Compute $\Delta \mathbf{u}$, $\Delta \theta$:

$$\delta \mathcal{R}(\mathbf{u}, \theta) \begin{bmatrix} \Delta \mathbf{u} \\ \Delta \theta \end{bmatrix} = -\mathcal{R}(\mathbf{u}, \theta)$$
 - 4: Update $\mathbf{u}^{k+1}$, θ^{k+1}

$$\begin{bmatrix} \mathbf{u}^{k+1} \\ \theta^{k+1} \end{bmatrix} = \begin{bmatrix} \mathbf{u}^k \\ \theta^k \end{bmatrix} + \begin{bmatrix} \Delta \mathbf{u} \\ \Delta \theta \end{bmatrix}$$
 - 5: **end while**
 - 6: $\mathbf{u}_{n+1} \leftarrow \mathbf{u}^{k+1}$, $\theta_{n+1} \leftarrow \theta^{k+1}$
 - 7: Update: $\mathcal{H}_{n+1}$ using updated $\mathbf{u}_{n+1}$ and θ_{n+1}
 - 8: Compute $\underline{d}_{n+1}$ by solving the linear problems of multiple phase-field variables
 - 9: **end**
-

the consistent tangent $\delta \mathcal{R}$ of the operator-split sub-problem, which is described in the following section. To obtain incremental updates of the multiple phase fields, the updated $\mathcal{H}_i$ from the displacement and temperature variables at time t_{n+1} are incorporated into the linear phase field solver. We then update the phase-field variables as in Eq. (4.76). To simplify the implementation, the temporal discretization of plastic dissipation and structural heating, i.e., the term $D_{\text{mech}} - H_{\theta}$ in Eq. (4.38) are treated explicitly. As shown in previous work such as Wang and Sun [2017a, 2016b,c], this semi-implicit approach can be effective if used properly. Finally, it should be noticed that one may choose other partition strategy to solve the same system of equations. For instance, the thermo-mechanical problem can also be solved using an isothermal or adiabatic approach. The explorations of different partition strategy and the construction of the proper pre-conditioners are important subjects, but are out of the scope of this study.

4.5.1 Linearization for the staggered algorithm

To obtain the numerical solution of the thermo-mechanical problems iteratively, the linearization of residuals of the governing equations is required. Due to the usage of the

iterative sequential solver, the operator-split residual will only update one set of known within one split iterative step and hence the linearization is only consistent numerically for the staggered solver but not for the monolithic counterpart. With this in mind, we first describe the linearization of the balance of linear momentum with respect to the displacement and temperature, as follows:

$$\delta G(\mathbf{u}, \theta, \underline{d}, \boldsymbol{\eta}) = \int_{\mathcal{B}} \nabla \boldsymbol{\eta} : \mathbb{C} : \delta \mathbf{u} dV - \int_{\mathcal{B}} \nabla \cdot \boldsymbol{\eta} [g(\underline{d})\alpha] \delta \theta dV, \quad (4.79)$$

in which $\mathbb{C} = g(\underline{d})\mathbb{C}^{ep}$ can be obtained from the constitutive laws based on the effective stress concept (Algorithms 1 and 2). Likewise, the linearization of energy balance equation can be derived as follows:

$$\begin{aligned} \delta H(\mathbf{u}, \theta, \underline{d}, \psi) = & \int_{\mathcal{B}} \psi c_v \delta \theta dV - \int_{\mathcal{B}} \psi \delta \left[\sum_{\alpha} (\pi^{\alpha} \dot{\gamma}^{\alpha} + g^{\alpha} \dot{s}^{\alpha}) \right] dV - \int_{\mathcal{B}} \psi \delta (\alpha \mathbf{1} : \dot{\epsilon}^e) \theta dV \\ & - \int_{\mathcal{B}} \psi (\alpha \mathbf{1} : \dot{\epsilon}^e) \delta \theta dV + \int_{\mathcal{B}} \nabla \psi \cdot \kappa \delta (\nabla \theta) dV. \end{aligned} \quad (4.80)$$

We note that the plastic dissipation and structure heating terms in the energy balance equation Eq. (4.38) can be considered as a source term due to internal mechanical work. The consistent linearization of these terms for fully implicit calculation requires complicated computations unless we employ the automatic differentiation tools (e.g., Albany). We therefore adopt the semi-implicit scheme for the energy balance equations by combining the backward Euler and forward Euler schemes Eq. (4.77). This procedure leads to the following tangent of energy balance equation,

$$\delta H(\mathbf{u}, \theta, \underline{d}, \psi) = \int_{\mathcal{B}} \psi c_v \delta \theta dV - \int_{\mathcal{B}} \psi (\alpha \mathbf{1} : \dot{\epsilon}^e) \delta \theta dV + \int_{\mathcal{B}} \nabla \psi \cdot \kappa \delta (\nabla \theta) dV. \quad (4.81)$$

For completeness, we include the linearization of the multiple phase-field equations as follows:

$$\delta L_i(\mathbf{u}, \theta, d_i, \phi_i) = \int_{\mathcal{B}} -2\phi_i \mathcal{H}_i \delta d_i dV + \int_{\mathcal{B}} \frac{G_c}{l} [\phi_i \delta d_i + l^2 \nabla \phi_i \cdot \boldsymbol{\omega}_i \cdot \delta (\nabla d_i)] dV. \quad (4.82)$$

Note again that this equation is for phase-field variable d_i , and other phase-field variables exhibit the identical expression.

4.6 Numerical Examples

We present numerical examples to validate and evaluate the applicability of the proposed combined multiple phase-field crystal plasticity model for a single crystal halite. Firstly, the plain strain (2D) tests are selected to investigate anisotropic behavior of a single crystal by changing Euler angle. In particular, we compare the stress-strain curve, plastic slip, and crack phase field to present the effect of changing orientations of the slip-system with respect to the loading direction. We further conduct the biaxial test by changing the boundary and initial conditions to evaluate the thermo-mechanical coupling effects of halite. A two dimensional tension test with a existing crack then is set up to explore how the loading rate affect fracture behavior. Finally we adopt a bicrystal as simplified polycrystalline rock salt, which gives the preliminary test for more complicated grain boundary conditions. The grain boundary is approximated by a single cleavage crystal

Table 4.1. Material properties of the specimens for the numerical simulations

Parameters	Description	Value	Unit
K	Bulk modulus	25.0	GPa
ν	Poisson's ratio	0.25	-
τ	Critical resolved shear stress	5.0×10^{-3}	GPa
h	hardening parameter	1.0×10^{-1}	GPa
G_c	Fracture energy	1.15	J/m ²
l	Length scale	1.0×10^{-5}	m
β	Anisotropy factor	40.0	-
Q	Activation energy	14.0	kcal/mol
R	Gas constant	1.986×10^{-3}	kcal/mol/K
C_0	Shape factor	1.0	-
p	exponent parameter	10.0	-
α	Thermal expansion coefficient	11.0×10^{-6}	1/K
c_v	Specific heat	2.0×10^6	J/m ³ /K
κ	Thermal conductivity	2.0	W/m/K

material with a finite thickness. By changing the fracture energy of grain boundary under the tensional loading conditions, we conducted three different simulations to provide an insight of how grain boundary characteristics influence the mechanical behavior including fractures.

The material properties, such as stiffness, strength, thermal parameters, activation energy, of halite or polycrystalline rock salt depend significantly on the environmental and loading conditions. Consequently, to analyze of those parameters itself has been an integral part in engineering application aspects (e.g., geological repository) as well as in material sciences (e.g., ceramics). In this numerical simulations, our main focus lies on investigating the applicability of our proposed formulations. Thus, the numerical parameters used in our study are adopted from the previous studies. Firstly, the elastic properties and fracture energy of a single crystal halite are used from the previous study by [Tromans and Meech \[2002\]](#). In addition, the critical resolved stress, hardening parameter, and creep parameters are borrowed from [Wawersik and Zeuch \[1984\]](#), [Wenk et al. \[1989\]](#). The thermal parameters including the specific heat, thermal conductivity, and thermal expansion coefficients of halite was studied by [Urquhart and Bauer \[2015\]](#). These material properties used in this study is summarized in Table 4.1.

The implementation of the numerical model leverages *Geocentric*, a massively parallel finite element code for geomechanics, which is built on the open source finite element library *deal.II* [[Bangerth et al., 2007a, 2013](#)] interfaced with *p4est* mesh handling library [[Burstedde et al., 2011a](#)] and *Trilinos* project [[Heroux and Willenbring, 2012a](#)]. The code base has been widely used in the previous studies including multiphysics problems (e.g., [Na et al. \[2017a\]](#), [Na and Sun \[2017a\]](#), [White et al. \[2016a\]](#), [Choo and Borja \[2015a\]](#), [White and Borja \[2011a, 2008a\]](#), [Borja and Choo \[2016a\]](#)).

4.6.1 Effect of crystal orientations

In this section, numerical examples are presented to investigate the anisotropic mechanical behaviors of a single crystal halite when subjected to various loadings under different loading rates and temperature range. Fig. 4.3 depicts the boundary conditions and the

definition of Euler angles for rotation of the slip system. For boundary conditions the horizontal directions are constrained on both top and bottom surfaces of the sample. The bottom surface are further constraint along the vertical direction, and the displacement boundary condition is applied at the tops surface to compress the sample. Note that the influence of confining on a single crystal is not considered, but its effect considering the grain boundary simulation will be covered in the future study. As can be seen from Figs. 4.2 and 4.3, two cleavage planes (or slip planes) projected onto 2D plane are used for directional information of multiple phase-field calculations. To be specific, unless changing orientations of the slip systems, each cleavage plane aligns to 45° and 135° directions onto 2D (x-horizontal, y-vertical) plane, respectively. Among the multiple possible combinations of the Euler angle ϕ and θ , we first use the default orientation (i.e., the slip orientations as in Fig. 4.2), and then set ϕ to 10° and 30° , respectively, to rotate the slip system on x-y plane. Due to the constraint at the top and bottom surfaces, we obtain the localized plastic and damage zones without adding other conditions (e.g., geometric or material defects).

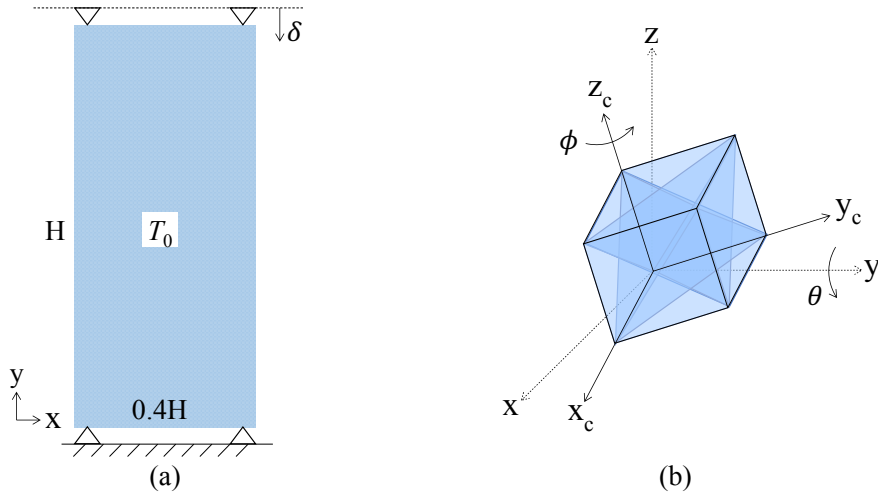


Fig. 4.3. Analysis of material anisotropy by changing the orientation of the slip system, (a) the set-up for plane strain compression test, (b) the definition of Euler angle (ϕ , θ) in the three-dimensional plane (reproduced from [Borja \[2013\]](#)), including the slip system of a single crystal halite (see each slip system in Fig. 4.2).

The number of numerical elements for the sample with $H = 1.0$ mm is 4,000 of bilinear quadrilateral element, which has 12,423 displacement unknowns and 8,282 phase-field unknowns (4,141 for each phase-field). Under three different orientation conditions (θ is fixed at 0° ; with $\phi = 0^\circ, 10^\circ$, and 30° , respectively), we observe the significant impact of material anisotropy on the mechanical responses without introducing any defects or inhomogeneity. The constraint boundary condition with material anisotropy facilitates the different responses and patterns of plastic and damage behaviors. In this simulation, the reference temperature, or the initial temperature condition is set to 25°C for all three test cases. The top boundary is moving downward during the simulation with the strain rate of $1.0 \times 10^{-5}/\text{sec}$.

To analyze the mechanical responses under plain strain tests, the differential stress and vertical strain curve with different orientations of the slip system are presented in

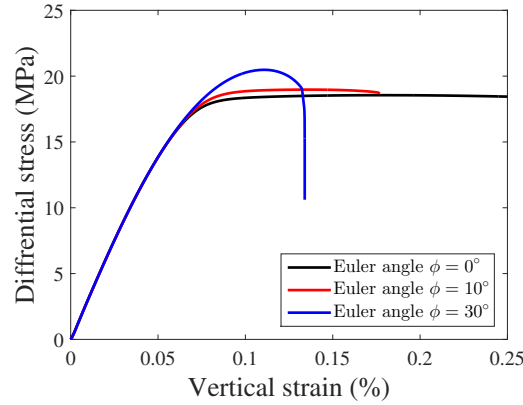


Fig. 4.4. Stress-strain curve of a single-crystal halite with different orientation of the slip-system (the Euler angle θ is fixed 0° while changing $\phi = 0^\circ$, 10° , and 30° , respectively).

Fig. 4.4. Although little difference is observed between the cases of $\phi = 0$ and $\phi = 10^\circ$, the resultant plastic slip patterns clearly show the impact of the orientation changes in Fig. 4.5. When the rotation of ϕ is within the range from 0 to around 10° , the damage is localized at the boundaries. In the case of $\phi = 30^\circ$, however, we observe the localized damage zone in the center of the domain not the edges. Furthermore, the plastic slip is also concentrated in accordance with the damaged zone. These patterns are consistent with the stress-strain curve in Fig. 4.4, which suggests the brittle fracture behavior. The temperature field for the effect of energy dissipation converging to heat is not presented, which has a minor role to influence on the mechanical behavior of halite.

4.6.2 Thermal effect on anisotropic creeping

We conduct the plain strain compression tests with different boundary conditions to analyze the thermal effect on halite. Firstly we designed the boundary conditions as depicted in Fig. 4.6. The left boundary is fixed along the horizontal direction, and the bottom surface is fixed against the vertical direction. The moving boundary with a constant strain rate (1.0×10^{-5} /sec.) applies the loading on the top surface as in the previous numerical example. Under the same mechanical loading condition, we set the three different initial reference temperatures on the sample domain: 25°C , 50°C , and 90°C , respectively. For three cases the same orientation of the slip system we use the same Euler angle, $\theta = 0^\circ$ and $\phi = 0^\circ$. Under different equilibrium temperature conditions, the mechanical responses including fracture and plastic behavior are evaluated.

First of all, we compare the mechanical responses under different temperature conditions using the differential stress and vertical strain curves in Fig. 4.6. For three cases the ductile fracture behaviors are observed while the plastic behaviors differ due to the difference in initial temperatures. Our crystal plasticity model adopts the creep behavior of halite that constitutes the temperature, which is essential in analyzing salt problems. Although the calibration has not been completed for temperature dependence and the fracture is initiated on the sample, the impact of temperature on the mechanical behavior of halite is clearly identified. Furthermore, we present the damage and plastic patterns at the same vertical strain, 0.23 % in Fig. 4.7. Followed by the stress-strain curves in Fig. 4.6, the clear fracture initiated from the top left edge is observed when the temperature

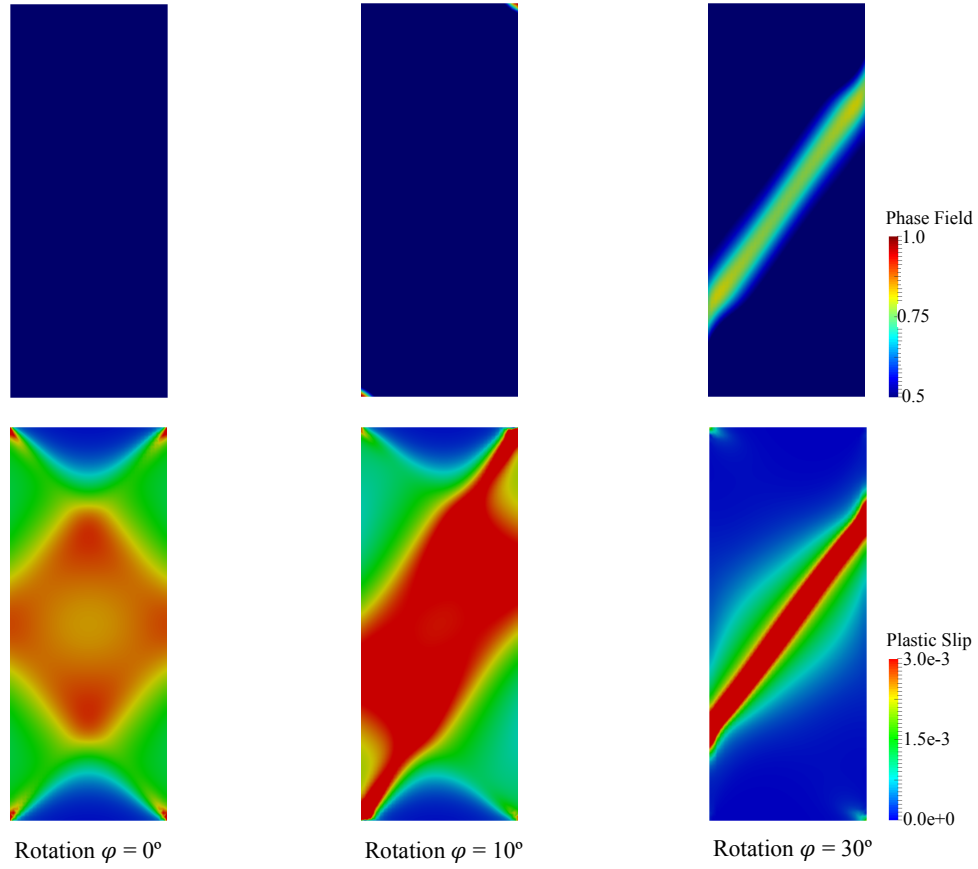


Fig. 4.5. Phase-field values (top) and plastic slip results patterns (bottom) with different orientations of the slip-system to investigate the material anisotropy under plain strain compression test condition. The patterns are captured at the last numerical step of each case. While fixing the Euler angle $\theta = 0^\circ$, ϕ varies 0, 10, and 30° .

is 25°C . The plastic zones including the localized region can be further identified. As the initial temperature increases, the less damage is observed at the same displacement, which can be easily expected from the stress-strain curves. Interestingly, the more plastic slip is concentrated at the right bottom area of the domain as the temperature increases. This indicates that the temperature may further influence the patterns of plastic zones under the same mechanical loading condition.

Now we adopt differently designed boundary value problem that mimics a storage room of a geological repository to evaluate the thermal effect in halite. The concept of the problem is adopted from the facility of the Waste Isolation Pilot Plant (WIPP) for investigation of room closer (cf. [Reedlunn \[2016\]](#)). Because the field problem requires the modeling of polycrystalline rock salt, the model for a single crystal halite cannot be used directly. Therefore, we focus on the problem that may happen in the room of repository facilities as in Fig. 4.8. The room is a type of tunnel that has a square shape section as depicted. Once the room is constructed, the disposed wastes fill the space and may generate heats depending on the type of wastes. Therefore, we may imagine a single crystal halite on the room surface that may be heated on a surface, in which the boundary

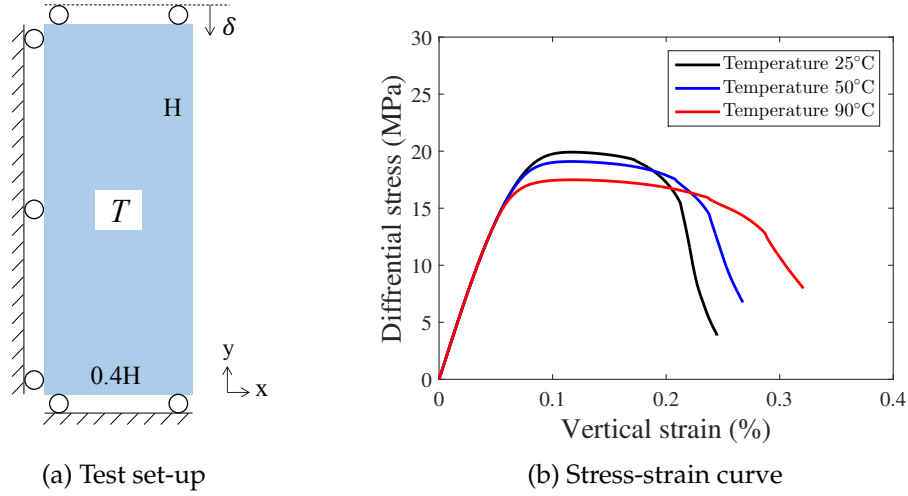


Fig. 4.6. Analysis of temperature effect on mechanical responses, (a) the set-up for plane strain compression test, (b) stress-strain curves for each temperature condition ($T = 25^\circ\text{C}$, 50°C , and 90°C , respectively).

condition is assumed as in Fig. 4.8 (c). The top, bottom, and right surfaces are constrained and insulated, but we apply heat flux on the left surface, that may be heated by nuclear wastes. Initially, we set the temperature on the domain 25°C and let the temperature increase around 100°C . Then we observe the sample behaves due to the thermal effect. The orientation of the Euler angle is set to $\theta = 0^\circ$ and $\phi = 20^\circ$.

In Fig. 4.9 we present the phase field value, plastic slip, and the horizontal displacement with its deformed shape. Due to the boundary conditions we set-up, the phase field and plastic slip are increased on the whole domain. To clearly identify the patterns, we set the ranges of phase-field value and plastic slip as in Fig. 4.9 for visualization purpose. Although we assume the isotropic thermal expansion behavior, the resultant damage and plastic patterns indicate anisotropic shapes. Furthermore, the sample domain is expanded to horizontal direction with the higher displacement concentration on the bottom parts. This further suggests the anisotropic behavior of halite due to thermal effect. Although the clear fracture is not observed, the impact of material anisotropy on the conceptual room closer problem is identified. This problem will be covered again using the polycrystalline rock salt models in the future study.

4.6.3 Loading rate effect

In this section, we conduct a simple tensile test on a specimen with a pre-existing crack. By changing the prescribed loading rates, we examine how the loading rate affects the interplay between the plastic deformation and the crack growth. The size of the square domain L is $1\text{mm} \times 1\text{mm}$ and the initial crack length is 0.5mm . The bottom surface of the specimen is constrained along both horizontal and vertical directions while the top surfaces are stretched vertically under a constant loading rate in each simulation.

As demonstrated in Fig. 4.10(b), the ductile-brittle transition of the halite crystal under different loading rate is captured. Increasing the loading rate leads to higher peak differential stress and the material also behaves more ductile in the softening regime. This is consistent with the behavior of crystalline rock [Paterson and Wong, 2005]. A

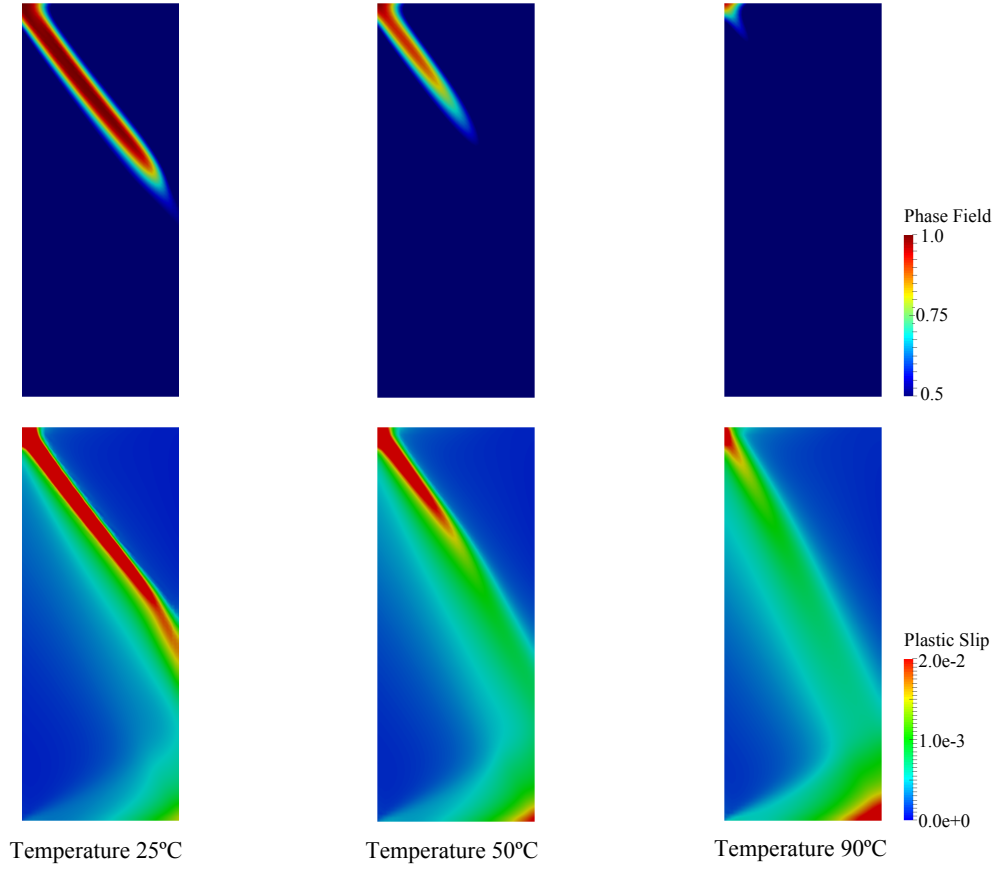


Fig. 4.7. Phase-field values (top) and plastic slip results (bottom) patterns with different initial temperature conditions at the same vertical strain, 0.23 %. The constant displacement loading is applied at the top, and the Euler angle of each temperature condition is fixed $\theta = 0^\circ$ and $\phi = 70^\circ$.

closer look at the crack pattern and the distribution of the plastic strain also reveals that the loading rate may not only affect the macroscopic responses, but also the induced anisotropy of the fracture and plastic deformation.

In particular, when the loading rate is slow ($\dot{\epsilon} = 1.0 \times 10^{-7}$ /sec.), the main crack tends to follow the cleavage plane directions (see right figure in Fig. 4.11). As the strain rate is increased to $\dot{\epsilon} = 1.0 \times 10^{-6}$ /sec, (the $10\dot{\epsilon}$ case in Fig. 4.11), a slightly zig-zag pattern is observed. It appears that initially the crack follows the cleavage plane but divert the propagation direction due to loading conditions and follow another cleavage plane at the left edge of the domain. Finally, as the loading rate increases (see left figure in Fig. 4.11), the crack propagation appears to be straight and the damaged zone is also more diffusive than the low-strain-rate counterpart. This result is perhaps attributed to the different amount of energy dissipation due to creeping deformation. As the plastic flow of each slip system is highly sensitive to the loading rate (cf. Eq. (4.60)), the changes of loading rate may affect the magnitude and direction of the overall plastic flow. In the the low strain rate case where the results resemble closely to the rate-independent crystal plasticity, the plastic flow is likely to be dominated by fewer slip system, which

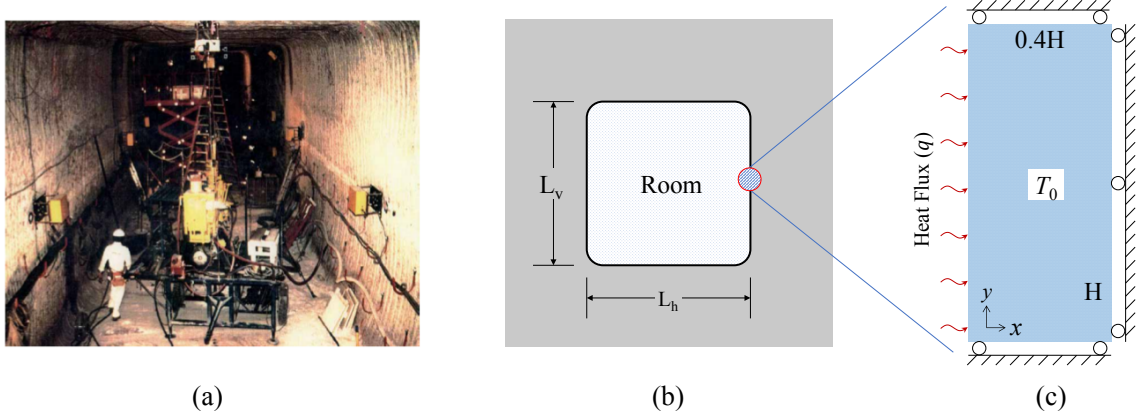


Fig. 4.8. The conceptual boundary value problem set-up for evaluating thermal impact on a single crystal halite, (a) a photo of room B (captured from [Reedlunn \[2016\]](#), [Munson et al. \[1990\]](#)), (b) the sections of rooms in WIPP (reproduced from [Reedlunn \[2016\]](#)), and (c) the problem set-up. The dimension of a room section (L_v and L_h) varies on each room in the WIPP. The top, bottom, and right surfaces are insulated, and the initial temperature T_0 is set to 25 °C. The whole domain temperature is increased to 100 °C by applying constant heat flux q . The Euler angle is set to

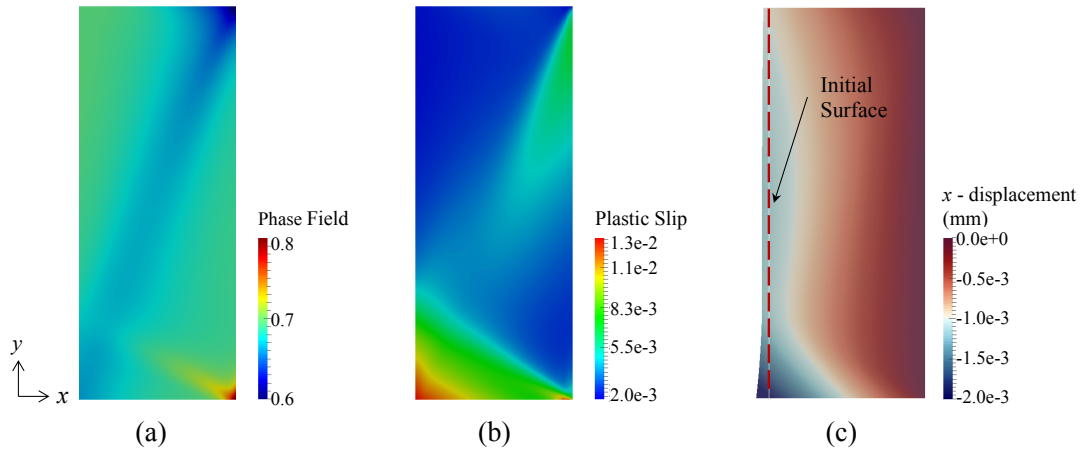


Fig. 4.9. The resultant patterns of (a) phase field for damages, (b) the plastic lip, and (c) the x direction displacement (scaled by $\times 10$) are illustrated when the overall temperature domain is increased up to 100°C. The ranges of each result are selected for visualization purpose.

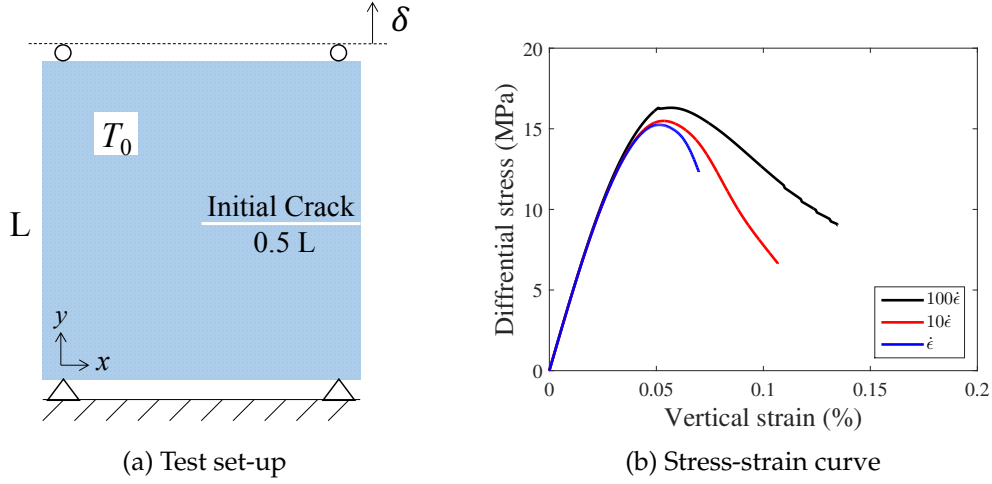


Fig. 4.10. Analysis of the loading rate effect under two-dimensional tension test, (a) the set-up for boundary value problem, (b) stress-strain curve with different loading rates ($\dot{\epsilon} = 1.0e^{-7}/\text{sec.}$). The initial temperature T_0 is set to 25 °C, and the Euler angle is set to $\theta = 0^\circ$ and $\phi = 0^\circ$.

causes plastic deformation appears to be more anisotropic. This in return changes the distribution of the driving force for each multiple phase field system in Eq. 4.50. As the multiple phase fields and plastic slip both evolve differently under different loading rates, this leads to the crack pattern appears to be more anisotropic in the low-strain-rate case.

4.6.4 Crack propagation in a bi-crystal halite

In this final example, we simulate a tensile test conducted on a bicrystal domain to investigate the interaction between grain and grain boundary. Similar boundary value problems have been used previously in [Oshima et al. \[2014\]](#) and [Nguyen et al. \[2017b\]](#) to model intra-grain and grain boundary fractures. In this study, our major point of departure is the introduction of crystal plasticity model to capture the plastic flow of the slip system inside the crystal and the modeling of plastic slip of the grain boundary. To simplify the modeling effort, the grain boundary is approximated using a crystalline material with finite thickness that has a single cleavage plane of which the slip direction is aligned to longitudinal direction of the grain boundary. The upshot of this treatment on grain boundary leads to a much simpler numerical model in which there is no need to capture the embedded discontinuities via cohesive element or enrichment function. However, as explained previously in [Wei and Anand \[2004\]](#), one must be cautious that representing strong discontinuities with regularized interfaces might leads to unrealistic large volume of fraction of grain-boundary regions if the thickness of the regularized interface is too large. The setup of the boundary value problem is depicted in Fig. 4.12, the grain boundary is located on the diagonal of the domain, which is separated into two grains having different orientations (30 and 60 degrees). This pre-existing crack is purposely put inside the gain such that the intra-grain crack may propagate toward the grain boundary.

We adopt the fracture energies reported by [Tromans and Meech \[2002\]](#), which estimates ideal fracture energies of both grain and grain boundary of a variety of minerals

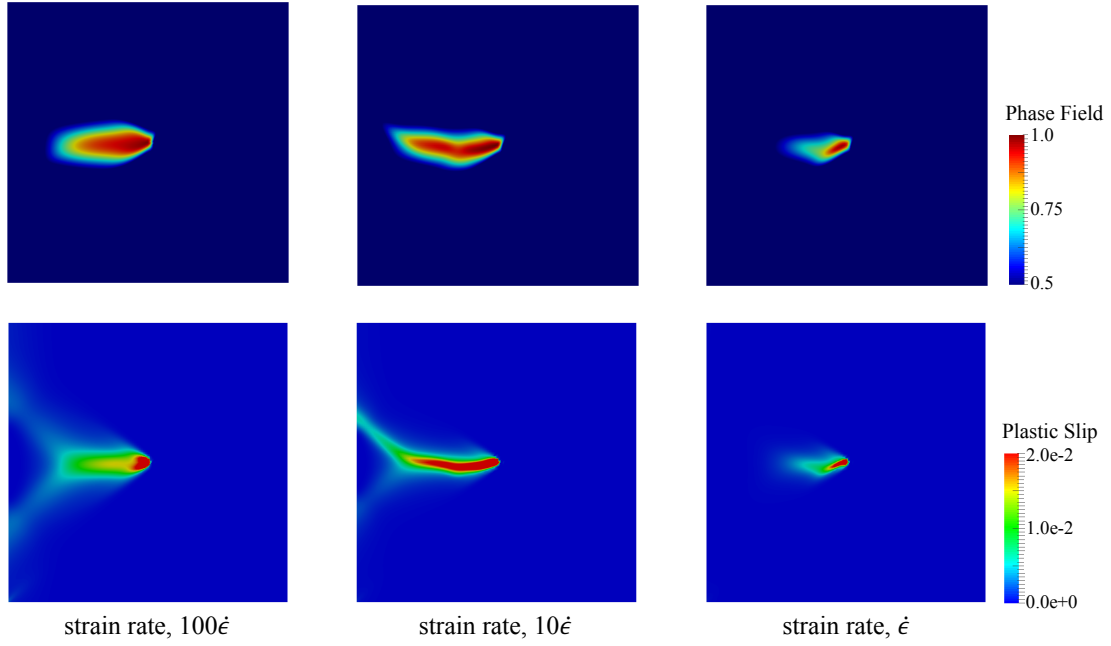


Fig. 4.11. Phase-field values (top) and plastic slip results (bottom) patterns with different loading rates are illustrated. The results are captured at the last numerical step of each simulation. The loading rate $\dot{\epsilon} = 1.0e^{-7}/\text{sec}$.

including halite. In this paper, the grain boundary fracture energy was estimated to 86% of the grain fracture energy: 1.155 J/m^2 for the grain, and 0.993 J/m^2 for the grain boundary, respectively. We further assume the critical resolved stress $\tau_Y = 7.5 \text{ MPa}$, hardening parameter ($h = 5 \text{ GPa}$), and activation energy $Q = 10.0 \text{ kcal/mol/K}$. To analyze the relation between the crack patterns and the grain boundary fracture energy, we conduct multiple simulations where the fracture energy of the grain boundary is assumed to be 86%, 75%, and 50% of the intra-grain counterpart. Other material properties of the grain boundary, that may differ from the grain due to diverse reasons, such as impurity, solid mass exchange, precipitation creep, etc., are out of the scope in this study but will be considered in the future. The material parameters used in the simulations can be found in Table 4.1. As discussed in Appendix .1, the material parameters of halite are highly sensitive to thermal and mechanical conditions. The well designed experiments, therefore, need to be conducted to calibrate the numerical models for particular engineering application purposes.

The simulated crack patterns of a bi-crystal with three different grain-boundary fracture energies (86%, 75% and 50% of intra-grain fracture energy), are shown in Fig. 4.13. In particular, The damage represented by the combined phase field (left figures) and the magnitude of the plastic slip (right figures) for the cases where the grain-boundary fracture energy is 86%, 75% and 50% are (a) and (b), (c) and (d), and (e) and (f) accordingly. In the 85% case, the crack and plastic strain initiates from the re-existing crack tip and propagates through the grain boundary until reaching the right end of the domain. In other words, the grain boundary causes the refraction of the crack, a phenomenon commonly observed in materials with inter-layer(s) [Nam et al., 2012].

When the fracture energy is reduced to 70% of the grain, as shown in Fig. 4.13(c) and

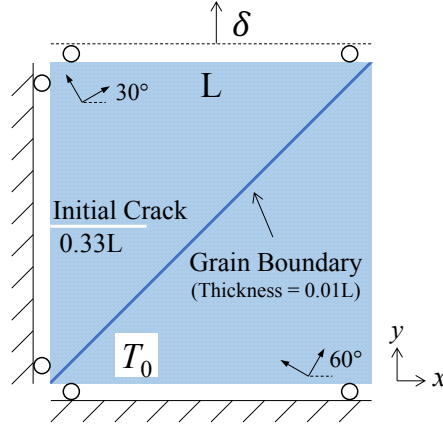


Fig. 4.12. The numerical set-up for a bicrystal is depicted to analyze the impact of the grain boundary fracture energy on crack propagations (reproduced from [Oshima et al. \[2014\]](#)). The Euler angles ϕ are set to -15° and 15° , respectively to left and right side grains. The single cleavage plane is assumed to the grain boundary with the direction of 45° along the diagonal. The initial temperature is set to 25°C , and the fracture energies of the grain boundary is adopted 86%, 75%, and 50% of the grain fracture energy, respectively.

(d), the main fracture behavior is similar to the previous case before reaching the grain boundary. However, when the crack reaches the grain boundary, it grows along the grain boundary before the crack refraction occurs. In the last case where the grain boundary energy is further reduced to 50% of the grain, the crack simply propagates along the grain boundary once reaching it. one interesting implication of this result is that the assumption that cracks only occur along the grain boundary may only be valid when the interface is sufficiently weak. Otherwise, it is possible that the grain boundary may lead to crack refraction, which may in return leads to damage and even fragmentation of crystal grains. Although this example is significantly simplified set-up, this result is an indicator that the intra-grain fractures could be significance in poly-crystalline materials in certain circumstance. In those cases, the phase field approach may provide a convenient mean to capture the interactions of intra-grain and grain-boundary fractures.

4.7 Conclusions

In this work, we developed a framework that combines the multiple phase-field method and crystal plasticity theory under non-isothermal conditions for modeling a single crystal halite, as a starting point for modeling coupled thermo-hydro-mechanical behavior of polycrystalline rock salt. Under the postulation of microforces associated with (1) plastic and (2) fracture behaviors, the microstructure information coming from its slip system is incorporated into the governing equations. Both the rate dependent and independent is captured via the crystal plasticity stress update algorithm, and the multiple phase-field method presents anisotropic damage of which each directional crack phase-field is associated with the slip planes. The energy balance equation is further encapsulated into the numerical framework to address the heat conduction, plastic dissipation, and thermoelastic structural heating. The numerical examples using a single crystal demonstrate

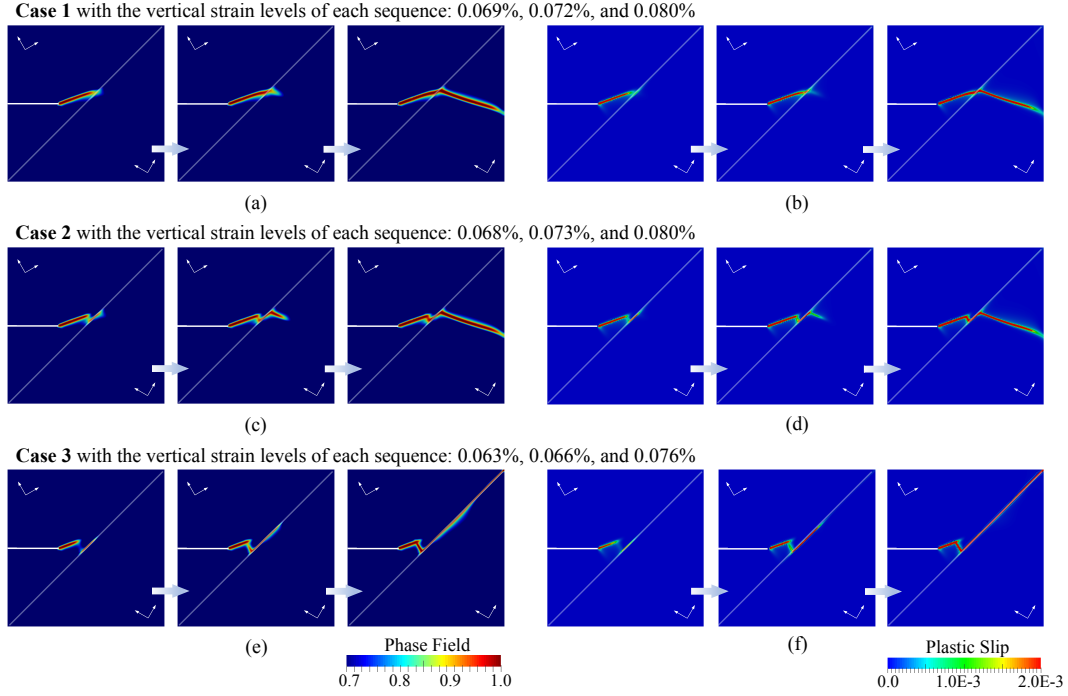


Fig. 4.13. Phase-field values (left) and plastic slip results (right) patterns with different fracture energies of the grain boundary are illustrated. Case 1 (a) and (b) assumes the fracture energy of the grain boundary with 86% of grain fracture energy; Case 2 (c) and (d) with 75%; Case 3 (e) and (f) with 50% of the grain fracture energy, respectively.

that, under the mechanical loading, the proposed numerical framework well captures the material anisotropy by rotating the slip system orientations and the variation of inelastic to fracture behavior under different temperature conditions. Furthermore, thermal loading by heat flux in a single crystal exhibits the anisotropic deformation, or distortion due to temperature increase. The tension test by changing the loading rate indicates the brittle-to-ductile transition along with the change of fracture patterns. Finally, the numerical example using a bicrystal presents the impact of fracture energy of the grain boundary on the inter- and intra-granular fracture, in which the grain boundary is simplified using a single cleavage crystal material. Although this framework for a single crystal is a starting point to be utilized as field scale simulations for polycrystalline rock salt, the results suggested here clearly indicate that this contribution provides quantitatively compatible physical behaviors of halite and potential applicability for practical engineering applications, e.g., a geologic repository for heating generating nuclear waste.

1 Stress-point simulations for model calibration

In this section, we performed the material calibration using the previous experimental work for a single crystal halite (e.g., [Carter and Heard \[1970\]](#)). Furthermore, we identified the significant sensitivity of material parameters for halite and indicated the limitation of the proposed numerical model and future direction toward modeling of polycrystalline rock salt.

As stated previously, the material parameters of halite or rock salt significantly depends on the environmental (e.g., temperature, etc.) and loading conditions (e.g., loading rate). As such, identifying the material properties of halite itself is an integral part for engineering applications. Furthermore, the calibration of numerical models suggested for different purposes to capture the specific behavior of halite under different test conditions is a challenging task. As a first step toward calibration for polycrystalline rock salt, we used the previous conducted experimental works by [Carter and Heard \[1970\]](#) to investigate how the proposed model captures the actual experimental work and evaluate the identified material properties of a single crystal halite. The calibration leverages the open source software toolkit *Dakota* [[Adams et al., 2009](#)], in which the gradient-based least square algorithm (NL2SOL) is used. This method minimizes the residual function, here we chose differential stress at each loading step, using its gradient. The variables for calibration includes the bulk modulus (K), critical resolved shear strength (τ_Y), hardening parameter (h), activation energy (Q), shape factor (C_0), and stress exponent (p). Here the isotropic elasticity is assumed for simplicity to halite, of which the bulk modulus is known to be around 26.0 GPa. Note that [Carter and Heard \[1970\]](#) used artificially grown single crystals of halite but the preparation of the sample lacked the details including the impurity, which may significantly influence the material properties. The further information can be found from [Carter and Heard \[1970\]](#). Furthermore, the information for elastic behavior within the small deformation regime was not clearly reported while a vast testing conditions were used for testing. However, this information of a single crystal halite is adopted for preliminary calibration testing in this study.

We first chose 8 sets of test results, which include two loading rates (1.0×10^{-4} /sec and 1.0×10^{-7} /sec) with four temperature conditions (25°C, 100°C, 200°C, and 300°C) with the vertical strain up to 10 %. Note that the proposed numerical model for stress point simulations may not be acceptable to capture these ranges of test conditions including large strain loading. However, we believe this process lead us to check the potential applicability of the proposed model and its limitation. As can be seen from Fig. 14, the overall behavior of the propose model is observed be similar to the mechanical behavior of halite obtained from the experiment. However, the calibration analysis did not reach its (relative) tolerance, which was set to 1.0×10^{-6} . In addition, this indicates limitations of the proposed model under these wide ranges of test conditions. Due to insufficient experimental information and severe nonlinearity within the small deformation regime ($< 1\%$), the significant errors within this region are observed, especially when the temperature was set to 25°C. Therefore, we reduced the experimental set for calibration and focused on the limited conditions of testing. The calibrated material parameters are presented as Case 1 in Table 2.

Table 2. Two sets of calibrated material parameters with

	K (GPa)	τ_Y (GPa)	h (GPa)	Q (kcal/mol)	C_0	p
Case 1	25.0	0.9	6.6	24.0	0.1	15.0
Case 2	1.1	1.9	80.1	8.0	1.0	10.0

We next streamlined the experimental results and used for stress-strain curves with two loading rates (1.0×10^{-4} /sec and 1.0×10^{-7} /sec) with two temperature conditions (25°C, 100°C). Furthermore, we analyzed the inverse problem within the small deformation regime ($< 2.0\%$). As a result, we observe the improved calibration results as in Fig. 15 for the small deformation with temperature conditions of 25°C and 100°C. The initial

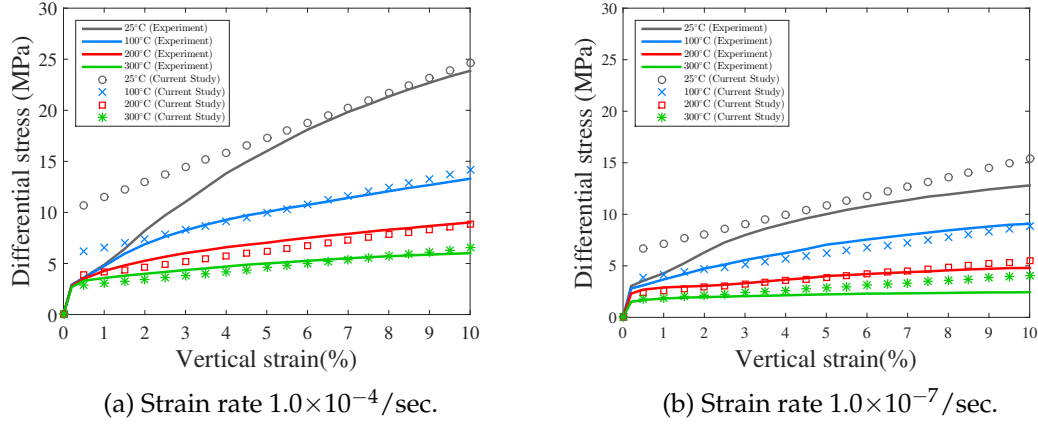


Fig. 14. Comparison of stress-strain curves from the experiment [Carter and Heard, 1970] and current study under different temperature (25°C, 100°, 200°, and 300°) and strain rate conditions, (a) with the strain rate 1.0×10^{-4} /sec, (b) the strain rate 1.0×10^{-7} /sec.

strain region, less than 0.2% of vertical strain, is still not calibrated enough due to the lack of information, whereas the calibrated behavior of halite is well matched with the experimental results. The calibrated material parameters are presented as Case 2 in Table 2. Compared to Case 1, the values of calibrated parameters are significantly changed including the bulk modulus K , hardening parameter h , and activation energy Q . This suggests that not only the plastic properties but the elastic parameters could be

This analysis further indicates that although the proposed model may not be suitable for a wide range of testing conditions, but we can still utilize this for a specific purpose. This conclusion not only leaves us room to improve the numerical model, but indicates the future direction of our research for polycrystalline rock salt which includes very complicated thermo-hydro-mechanical couplings. By combining the well designed experimental work for a geological repository stability, we may utilize material identification and calibration process to better capture the actual stability analysis. Note that the current calibration did not include the effect of damage. The calibration will be further expanded to actual sample considering damage in future study.

.2 Benchmark: multiple phase-field method for anisotropic crack propagation

This section describes how the multiple phase-field method captures the anisotropic crack propagations. The multiple phase-field method for anisotropic fracture using the elastic material has been proposed by Nguyen et al. [2017a,b], in which the derivation and the applications are well explained. Here we adopted this idea and conducted a benchmark simulation to check its implementation and ability to capture the anisotropic crack propagation using our numerical framework. Therefore, the ideas of the model and boundary value problem are all adopted from Nguyen et al. [2017a,b].

The test set-up for a guided crack propagation is described in Fig. 16 (a). The square domain with the length L is 1.0 mm, the higher fracture toughness is assigned to the top and bottom parts of the domain outside the guided line to prevent any crack propagation outside the guided line. For directional fracture, two phase-field variables are used with

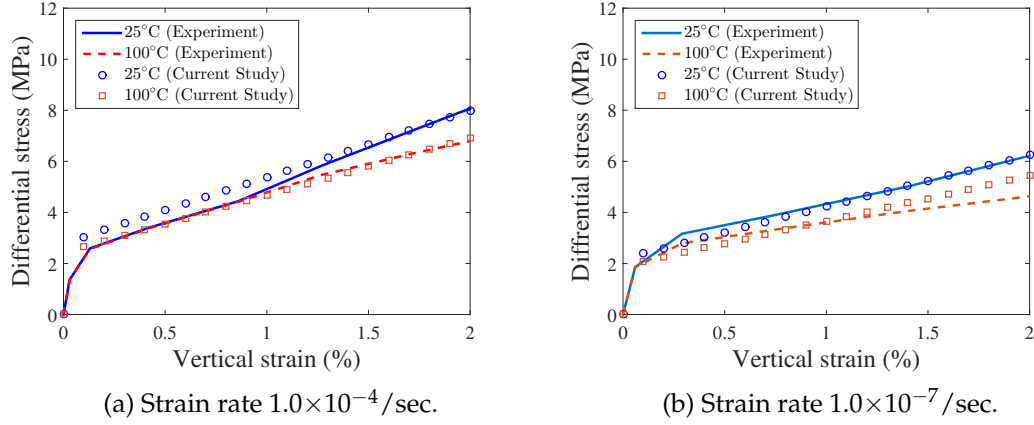


Fig. 15. Comparison of stress-strain curves from the experiment [Carter and Heard, 1970] and current study under different temperature (25°C and 100°) and strain rate conditions, (a) with the strain rate 1.0×10^{-4} /sec, (b) the strain rate 1.0×10^{-7} /sec.

the cleavage planes having normal vectors 135° and 45° directions, respectively. Note that the material is assumed to be elastic with the bulk modulus of 175 GPa, Poisson's ratio of 0.3, fracture energy of 2.7 kJ/m^2 , the length scale of 0.01 mm, and the anisotropy factor β of 50. As can be seen in Fig. 16 (c) and (d), the phase field 1 starts to grow from the edge of the existing crack while the phase field 2 does not grow until the crack hits the guided line. Then the crack due to phase field 2 starts to grow until it hits the guided line other side. This process continues and the combined crack indicates the zig-zag patterns as in Fig. 16. The combined crack phase field, or the equivalent phase field, is calculated using the given equation which is closely related to the degradation function for multiple phase-field method. The detailed explanations, derivations, and the anisotropic surface energy plots depending on the anisotropy factor β can be further found from Nguyen et al. [2017a,b]. We further note that this multiple phase-field method is combined with the crystal plasticity constitutive models for a single crystal halite and extended for thermal-mechanical behavior of salt in the current study.

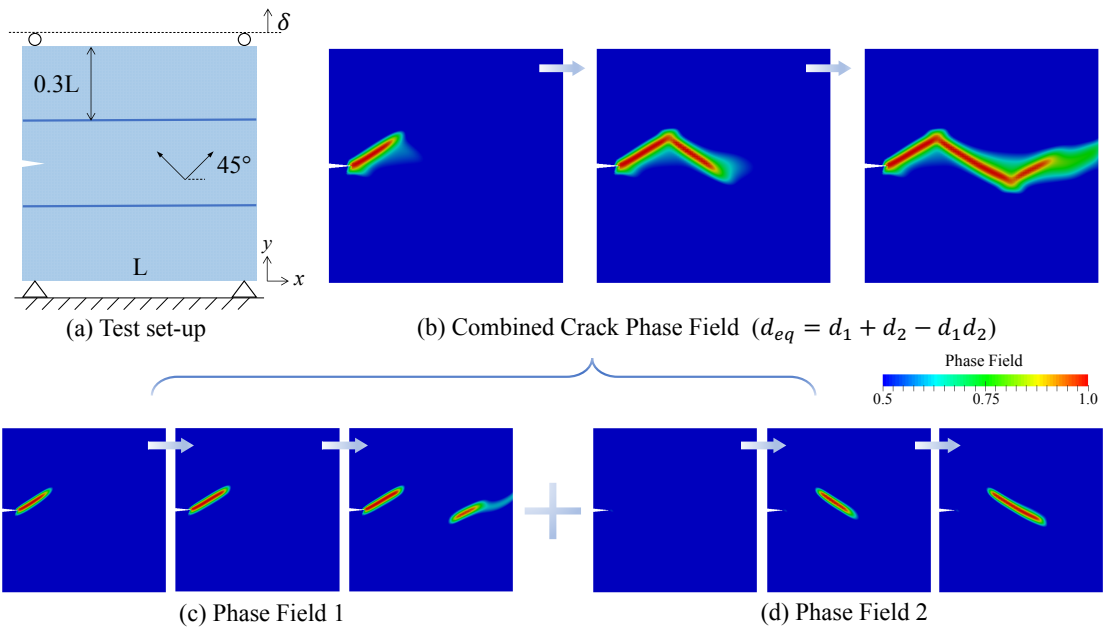


Fig. 16. The concept of multiple phase-field method for anisotropic crack propagation, (a) test set-up for guided crack propagation (the top and bottom parts of the domain outside the guided line have higher fracture toughness not to initiate cracks), (b) the combined crack phase field using the equation suggested by [Nguyen et al. \[2017a,b\]](#), (c) the crack propagation of phase-field 1, (d) the crack propagation of phase-field 2.

Chapter 5

Modeling size effect of clay in salt formation

We introduce a regularized anisotropic modified Cam-clay (MCC) model which captures the size-dependent anisotropic elastoplastic responses for clay, mudstone, shales, and sedimentary rock. By homogenizing the multiscale anisotropic effects induced by clay particle aggregate, clusters, peds, micro-fabric, and mineral contact across length scales, we introduce two distinctive anisotropic mechanisms for the MCC model at the material point and mesoscale levels. We first employ a mapping that links the anisotropic stress state to a fictitious isotropic principal stress-space to introduce anisotropy at the material point scale. Then, the mesoscale anisotropy is introduced via an anisotropic regularization mechanism. This anisotropic regularization mechanism is triggered by introducing gradient-dependence of the internal variables through a penalty method such that the resultant gradient-enhanced plastic flow may exhibit anisotropic responses non-coaxial to the stress gradient of the yield function. The influence of the size-dependent anisotropy on the formation of the shear band and the macroscopic responses of the effective media are analyzed in 2D and 3D numerical examples.

5.1 Introduction

Clay, mudstone and shales are materials that exhibit different anisotropic response across different length scales. This scale-dependent anisotropy originates from the fabric and microstructures of clay platelet and other fragments of minerals such as quartz and calcite. At the nanoscale, clay particles may aggregate together in sub-microscopic fabric units, which is often referred as domains. These domains then form clusters, which in return form peds that are large enough to be visible. Together with other features, such as joints and fissures, the peds then form a microfabric system that is inherently anisotropic. As shown in Fig. 5.1 (cf. Desbois et al. [2012b] and Bennett et al. [2015]), the anisotropy of clay and crystalline rock is often not just manifested by a single morphological feature (e.g. bedding orientation, inclusion of different materials). Instead, the anisotropy of the material response is the consequence of multiple microstructural mechanisms that may differ for effective media of different sampling sizes [Collins and McGown, 1974].

Furthermore, size effects are also related to the statistical distribution in the severity of flaws. For instance, Weibull's theory predicts that the stress that triggered fracture is proportional to the $V^{1/\beta}$ where V is the volume of the specimen and β is a material constant [Weibull, 1951]. On the other hand, experimental observations of size dependence in

geological materials have been reported in uniaxial compression and in Brazilian tests [Na et al., 2017a, Paterson and Wong, 2005]. In general, these tests often lead to the conclusion that shear strength consistently decreases with increasing dimensions. However, it is important to note that the size-dependent effect is often more profound when a sharp stress gradient presents due to the loading conditions (e.g., punch and indentation tests). This size dependence might also be suppressed when the confining pressure increases [Habib et al., 1966], but is of great importance in the brittle regime.

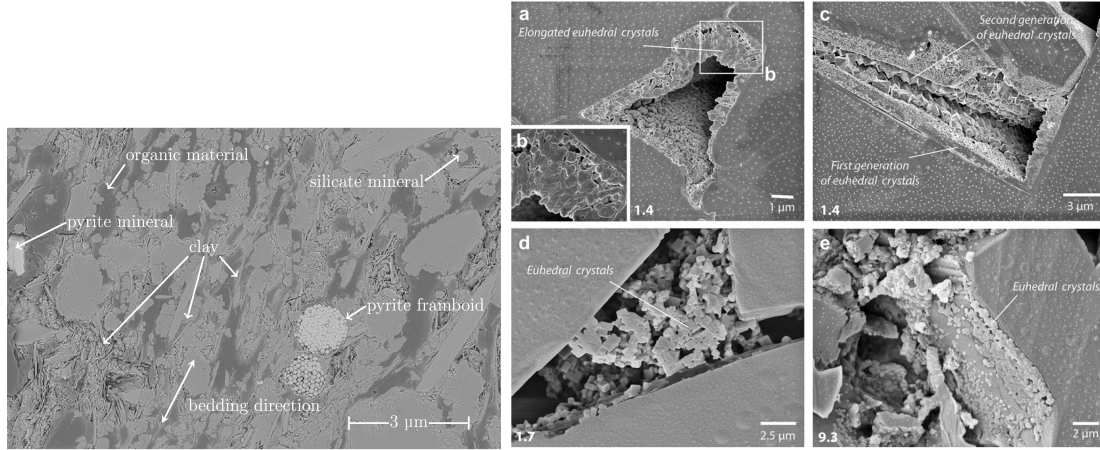


Fig. 5.1. Rock material heterogeneity at the micrometer scale, showing two different materials of significant engineering interest, both characterized by size-dependence of the anisotropy.

The major contribution of this work is the introduction of non-coaxial micromorphic regularization for the anisotropic MCC model. This treatment enables us to represent the distinct anisotropic characteristics at the particle and aggregate scales, via two mathematical treatments – (1) by introducing mapping tensor at the constitutive laws following Semnani et al. [2016] and (2) by introducing anisotropic micromorphic regularization via a penalty or relaxation functional following Forest [2016], Miehe et al. [2016], Forest et al. [2018]. The introduction of the map greatly simplifies the implementation of anisotropic constitutive laws, as demonstrated in previous works such as Semnani et al. [2016] and Bennett et al. [accepted]. Meanwhile, we introduce the anisotropic regularization by a variational model in which an Euler-Lagrange equation leads to the incremental update of the local constitutive law and two sets of Helmholtz equations that regularize the plastic flow and circumvent pathological mesh dependence. Rather than directly introducing gradient term to the plastic flow, we introduce coupling energy functionals that penalize the difference between the local internal variables and the global projected internal variables updated by the Helmholtz equations. This treatment enables us to bypass the identification of the plastic zone and the projection of the local internal variables typically required for gradient plasticity models [Stankiewicz and Pamin, 2006]. By leveraging the non-coaxiality of the local transversely isotropic plane and the diffusivity tensor in the Helmholtz equations, the new model is able to exhibit plastic flow of different direction than the stress gradient of the isotropically-regularized yield function, without introducing any plastic potential function, differently than the yield function or direction changes commonly employed in generalized plasticity models [Zienkiewicz et al., 1999]. To the best of our knowledge, this work is the first micromorphically regularized Cam-clay model, and is designed to capture the size-dependent anisotropy in geological materials.

As pointed out in previous work such as Wang and Sun [2016a], Sun [2015a], Sun et al. [2013a, 2014], Scovazzi et al. [2016], Wang and Sun [2018], isochoric plastic flow occurring at the critical state may cause significant numerical challenges due to volumetric locking and potential low or zero energy modes sometimes attributed to the failure of geomaterials [Wang and Sun, 2016a, Sun and Andrade, 2011, Sun, 2013b, Krischok and Linder, 2016a]. While the micromorphic regularization is already known to be an effective localization limiter, its effect on relaxing the volumetric locking has not yet been examined in detail. Our numerical examples have provided observations and numerical evidence to fill this knowledge gap. Finally, our numerical examples also indicate that the multiscale anisotropic model is capable of capturing the key morphological characteristic of the deformation band in anisotropic materials.

The organization of the rest of the paper is as follows. We first introduce the key elements of the anisotropic MCC model, including the usage of a mapping tensor for the local constitutive law and the gradient-based diffusive penalty stored work functional that provides a non-coaxial regularization for the post-bifurcation responses. Following this, the details of the implementation of the return mapping algorithm are discussed. Numerical examples are given and a brief summary of key results are presented in the conclusion.

5.2 Micromorphic anisotropic MCC model

In this section, we introduce the formulation of the micromorphic anisotropic MCC model capturing the size-dependent anisotropy of the plastic response. For simplicity, the anisotropically elastic response of the material is assumed to be linear. Then, we incorporate a non-coaxial anisotropic micromorphic regularization into the anisotropic MCC framework previously introduced in Crook et al. [2002] and codified in Semnani et al. [2016]. While both the anisotropic mapping and the anisotropic regularization both lead to transversely isotropic responses, they influence the plastic deformation differently. Local anisotropy introduced through the map may provides the response of a homogenized effective medium. In contrast, the anisotropic regularization provides a mechanism that introduces size-dependent anisotropy, wherein the anisotropic response is sensitive to the physical length scale of the material for a given orientation. Since these two anisotropic mechanisms are not necessarily co-axial, incorporating both can provide more flexibility to capture of the material responses of the microstructure composed of fabric, platelet, and minerals that span multiple length scales. The following assumptions are made throughout this paper.

- The deformation remains infinitesimal such that the infinitesimal strain measure applies, i.e. $\epsilon = (\nabla^X \mathbf{u} + \nabla^X \mathbf{u}^T)/2 = \nabla^X_s \mathbf{u}$, where $\mathbf{u}$ is the displacement field.
- The deformation process occurs while maintaining the isothermal condition such that the heat transfer can be neglected.
- The inertial force is negligible and hence the material is under a quasi-static condition.

In the geometrically linear regime, the additive strain decomposition of the infinitesimal elastic and plastic strains is valid, i.e.

$$\epsilon = \epsilon^e + \epsilon^p, \quad (5.1)$$

where superscripting e and p designates elastic and plastic parts of the strain, respectively. The stored work is partitioned into the elastic strain energy and the stored plastic work, i.e.

$$W(\epsilon^e, \alpha, \tilde{\alpha}) = W^e(\epsilon^e) + W^p(\alpha, \tilde{\alpha}), \quad (5.2)$$

where α is a collection of strain-like internal variables computed from the plastic strain ϵ^p . In contrast, $\tilde{\alpha}$ is a collection of field variables related to the internal variables by relaxation functionals in the stored plastic work W^p [Forest, 2009, Aldakheel and Miehe, 2017]. The total stored plastic work W^p is partitioned as

$$W^p(\alpha, \tilde{\alpha}) = W_\alpha^p(\alpha) + W_{\tilde{\alpha}}^p(\alpha, \tilde{\alpha}), \quad (5.3)$$

where $\alpha = \{\lambda, \epsilon_v^p\}$ is the set of history-dependent variables subjected to gradient regularization via a relaxation energy functional that penalizes the discrepancy of α and the set of corresponding field variables constrained by the Helmholtz equation, i.e. $\tilde{\alpha} = \{\tilde{\lambda}, \tilde{\epsilon}_v^p\}$. In this work, we introduce regularization both on the plastic multiplier λ and on the volumetric plastic strain ϵ_v^p . This strategy is more complex, more costly and requires more elaborated calibration effects to identify material parameters than the alternative where only one internal variable is regularized in Forest [2009], Aldakheel and Miehe [2017]. However, introducing the gradient regularization for both the plastic multiplier λ and the volumetric plastic strain ϵ_v^p also provides some benefits in capturing the critical state under which the plastic strain becomes isochoric [Sun et al., 2013a, Schofield and Wroth, 1968, Wood, 1990, Abboud and Scovazzi, 2018]. In particular, the second Helmholtz equation for the regularized field variable $\tilde{\epsilon}_v^p$ may penalize the sharp gradient of the local volumetric plastic strain by introducing a cost to generate such a sharp gradient. In principle, this technique can also be applied in an element-by-element manner through introducing an assumed strain formulation (Krischok and Linder [2016a]), reduced integration with hourglass control (e.g. Reese et al. [2017]) or nonlocal averaging on a patch [Cheng et al., 2016]. Preliminary studies presented in our numerical examples indicates that introducing the Helmholtz equation for the local volumetric plastic strain is sufficient to circumvent the volumetric locking with a more intuitive physical underpinning. Further analysis is certainly required to determine the optimal approach to capture the isochoric plastic flow at the critical state, but such an analysis is out of the scope of current study.

Note that this indirect approach enables the field variables within $\tilde{\alpha}$ to be defined not just inside the plastic zone but on the entire body $\mathcal{B}$ with boundary $\partial\mathcal{B}$ as:

$$\tilde{\lambda} : \mathcal{B} \rightarrow \mathbb{R}^+ \cup \{0\} \mid \hat{n} \cdot \omega_\lambda \cdot \nabla^X \tilde{\lambda} = 0 \text{ on } \partial\mathcal{B}, \quad \tilde{\epsilon}_v^p : \mathcal{B} \rightarrow \mathbb{R} \mid \hat{n} \cdot \omega_v \cdot \nabla^X \tilde{\epsilon}_v^p = 0 \text{ on } \partial\mathcal{B}, \quad (5.4)$$

where $\hat{n}$ is the boundary unit outward normal, and ω_λ and ω_v are second-order micro-morphic diffusivity tensors. These tensors and the specific form of $W_{\tilde{\alpha}}^p(\alpha, \tilde{\alpha})$ are discussed in Section 5.2.3.

5.2.1 Anisotropic elasticity

We employ a linear elasticity model such that the strain energy and elastic strain are related via

$$W^e = \frac{1}{2} \epsilon^e : \mathbf{C}^e : \epsilon^e, \quad (5.5)$$

where $\mathbf{C}^e$ is an super-symmetric fourth-order tensor expressing transverse isotropy of the elastic material response (for terminology cf. Itskov [2000], and for discussion of related

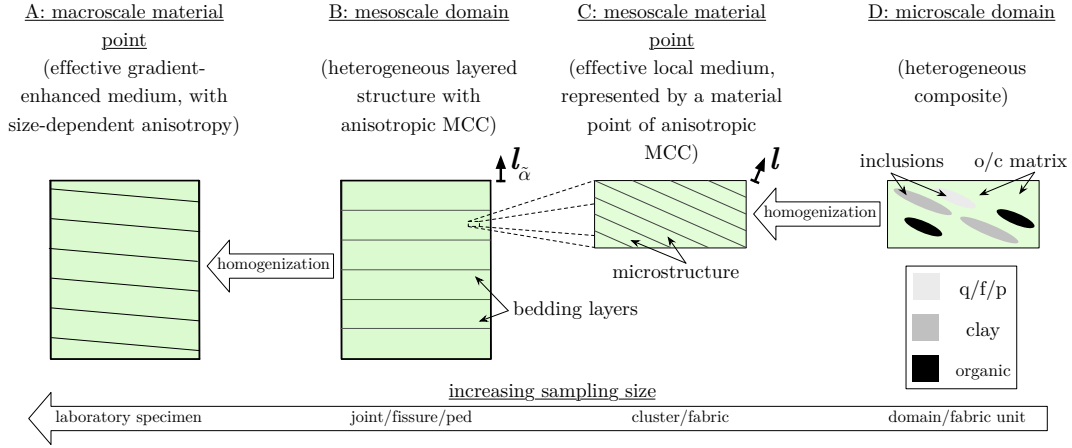


Fig. 5.2. Schematic of mesoscale and microscale-homogenized material effective media, where l is the microstructural direction whereas $l_{\tilde{\alpha}}$ is the direction characterizing the anisotropic micromorphic regularization of a plastic internal variable α , via the projected internal variable $\tilde{\alpha}$, governed by energy functionals as detailed in Section 5.2.3, with inhomogeneous microscale identification and constituent categorization after Bennett et al. [2015]; q/f/p means inorganic quartz, feldspar, or pyrite, and o/c matrix indicates organic and clay matrix.

Kelvin-notated matrix equivalents exhibiting symmetry such as the solid elastic tangent, see Appendix ??).

As such by Eq. (5.5), we discard in this work: nonlinearity of the material's elastic volumetric response, such that the pressure is semilogarithmic in the trace of the elastic strain during elastic unloading or rebounding; and, any potential coupling of the effective shear moduli to changes in the elastic bulk modulus with confining stress. In fact, elastic nonlinearity has previously been written in a strain energy functional amenable to a variational treatment, e.g. see Eq. (3.4-6) in Borja and Tamagnini [1998], given that our model's local minimization is already over the elastic strain (per later Section 5.2.5).

To introduce anisotropy of the elastic response corresponding to the orientation of the isotropic plane's normal, this elastic stiffness tensor C^e is expressed as a function of a second-order microstructural tensor, a dyadic tensor denoted as $\phi = l \otimes l$, where the microstructural direction l is a unit vector normal to the plane of isotropy, for $l \cdot l = 1$. For instance, l would be approximately vertical for many in-situ shale rock layers. The transversely isotropic elastic stiffness tensor is then represented via direction l (cf. Walpole [1984]):

$$C^e = c_1 E_1 + c_2 E_2 + c_3 (E_3 + E_4) + c_5 F + c_6 G, \quad (5.6)$$

where c_1 through c_6 are elastic moduli and E_1 through G are fourth-order tensors, related to the elastic input parameters, see Appendix ??.

In this work, we introduce a gradient-dependent constitutive law for geomaterials (e.g. clay, mudstone, shales and salt) that may exhibit size-dependent anisotropy due to the complex microstructures. As shown in the example illustrated in Fig. 5.2, a macroscopic representative elementary volume (e.g. A in Fig. 5.2) may be formed by mesoscale layers that introduce anisotropy at the macroscopic scale (e.g. the homogenization from B to A in Fig. 5.2). However, the composite materials that form each layer may also contain microscopic fabrics or domain units (e.g. D in Fig. 5.2) that introduce an mesoscale anisotropy originating from the microscale composite (e.g. the homogenization from D to

C in Fig. 5.2) but distinctive from those anisotropic effects introduced by the orientation of the homogenized layers (e.g. B in Fig. 5.2). As such, if we introduce a transversely isotropic effective medium at the scale comparable to the mesoscale layers, then the Euler angles between the effective principal directions corresponding to the homogenized principal strain and stress tensors of the effective medium may vary when different sizes of the representative elementary volume are subjected to homogenization. This effect is referred as size-dependent anisotropy throughout this paper. To capture this size-dependent anisotropy, we therefore introduce an anisotropic regularization for a transversely isotropic local constitutive law and use the mismatches among the isotropy plane of the local constitutive law and the principal directions of the diffusivity tensors of the gradient terms to replicate the size-dependence of anisotropy across length scales.

5.2.2 Anisotropic local plasticity

This section describes the local contribution W_α^P of the total stored plastic work W^P , which is partitioned as $W^P(\alpha, \tilde{\alpha}) = W_\alpha^P(\alpha) + W_{\tilde{\alpha}}^P(\alpha, \tilde{\alpha})$. In particular, we will review the relation between the hardening law and the stored plastic work (cf. Section 5.2.2) and the flow rules introducing via the mapping technique (cf. Section 5.2.2).

Hardening law

The stored work of hardening W_c^P in the purely local stored plastic work W_α^P is computed using an exponential constitutive relation. This admits model parameters (e.g. C_d) resembling the expression of the classical hardening law for the isotropic MCC counterpart (cf. Borja [2013], Schofield and Wroth [1968], Roscoe and Schofield [1963]),

$$p_c = p_{c0} \exp \left(\frac{\epsilon_v^P - \epsilon_v^P}{C_d} \right), \quad \dot{\epsilon}_v^P = \mathbf{b}_v : \dot{\epsilon}^P, \quad (5.7)$$

where $p_{c0} < 0$ is the reference pressure, and $C_d > 0$ is a material parameter related to the difference between the plastic and elastic compressibility of the materials. In the special case where (1) the bulk modulus of the material is $K = -p/C_r$ (which is not the case described in Section 5.2.1) and (2) the gradient regularization effect is vanished, then the bilogarithmic compressibility law $C_d = C_c - C_r$ introduced in Hashiguchi and Ueno [1977] and Butterfield [1979] is recovered over a finite load increment. In other words, the parameter C_d can be obtained from an one-dimensional compression (oedometer) test. However, special caution must be paid to ensure that the plastic deformation of the specimen remains *homogeneous* (such that the Laplacian/diffusion terms of the Helmholtz equations vanish) along the normal compression line and the hysteresis loops.

Note that in this content, $\dot{\epsilon}_v^P = \mathbf{b}_v : \dot{\epsilon}^P$ is not the increment of the volumetric plastic strain in the physical space but a strain measure in the fictitious space. Symmetric second-order tensor $\mathbf{b}_v$ is conventionally $\mathbf{1}$, such that ϵ_v^P is the volumetric plastic strain. For our small-strain kinematic assumption, this implies to a semilogarithmic relation between $-p_c$ and ϵ_v during plastic deformation, cf. Eqs. (3.14-19) in Borja and Tamagnini [1998]. Integrating p_c to find the stored work

$$W_c^P - W_{c0}^P = \int_{\epsilon_v^P}^{\epsilon_v^P} p_c d\epsilon_v^P = p_{c0} C_d \left[1 - \exp \left(\frac{\epsilon_v^P - \epsilon_v^P}{C_d} \right) \right], \quad (5.8)$$

where subscripting 0 indicates the reference state. In our numerical examples, we consider the idealized case in which the preconsolidation pressure p_{c0} is non-trivial but $\epsilon_v^p = 0$ and p_{c0} as indicated in Appendix ?? . For applications in the field scale, a more elaborated calibration is needed to establish the equilibrium state at the beginning of the simulations, as the residual stress and the body force may lead to a deformed initial configuration [Potts et al., 2001].

Flow rule in mapped space

In our proposed constitutive framework, anisotropy of the responses originates from two sources, the anisotropy from the local constitutive law and the counterpart from anisotropic micromorphic regularization. As to the local plastic anisotropy, we employ a eigen-space mapping strategy. The key idea of this eigen-space mapping idea is to create a mapping in between the physical space and an fictitious isotropic space, such that anisotropic constitutive response in the physical space can be obtained by mapping a isotropic constitutive response obtained from a isotropic plasticity model to the physical space. This strategy has applied as a stress-space mapping in different isotropic yield functions to introduce anisotropy, such as Hashagen and de Borst [2001] for the Hoffman yield function, Crook et al. [2002] and Semnani et al. [2016] for the MCC yield function, Versino and Bennett [2018] for the Von Mises yield function and Bennett et al. [accepted] for the Drucker-Prager yield function.

The upshot of this approach is that one may, in theory, create anisotropic constitutive laws from a template of isotropic constitutive law by introducing a linear and one-to-one mapping between the real configuration of the material and the fictitious isotropic configuration, by either stress-space or, as here, strain-space mapping tensors. The anisotropic plasticity mapping can be represented by a super-symmetric fourth-order tensor. This tensor is also built using I as, cf. Semnani et al. [2016]:

$$P^P = \beta P_1^P + (\alpha + \beta - 2\gamma) P_2^P + 2(\gamma - \beta) P_3^P, \quad (5.9)$$

where α , β , and γ are plastic anisotropy input parameters, and P_1^P through P_3^P are super-symmetric fourth-order tensors, see Appendix ?? . Input material parameters are selected such that P^P is invertible and, on that note, $P^P(\alpha, \beta, \gamma) = I$ for $\alpha = \beta = \gamma = 1$. Their plastic map defines the mean pressure and deviatoric stress in the mapped fictitious isotropic stress-space,

$$p^* = a_p : \sigma, \quad q^* = \sqrt{\frac{1}{2} \sigma : A_q : \sigma}, \quad (5.10)$$

where

$$a_p = \frac{1}{3} P^{p \text{ vol}} : \mathbf{1}, \quad A_q = 3 P^{p \text{ dev}} : P^{\text{dev}} : P^{p \text{ dev}}, \quad P^{\text{dev}} = I - P^{\text{vol}}, \quad P^{\text{vol}} = \frac{1}{3} \mathbf{1} \otimes \mathbf{1},$$

for distinct mappings $P^{p \text{ dev}}(\alpha^{\text{dev}}, \beta^{\text{dev}}, \gamma^{\text{dev}}) \neq P^{p \text{ vol}}(\alpha^{\text{vol}}, \beta^{\text{vol}}, \gamma^{\text{vol}})$. Superscripting 'dev' and 'vol' relates to independent deviatoric and volumetric maps, respectively. Absent superscript *, p and q are computed using I as the plastic mapping tensor. Fourth-order projection tensors P^{dev} and P^{vol} are idempotent and orthogonal, cf. Itskov [2000] or Section 2.6 in Simo [1998].

Following the treatment in Ortiz and Pandolfi [2004], we consider the plastic deformation obey the same associative flow rule and that a variational structure exists such that

the plastic flow rule is the primitive postulate of the theory, and latterly the elastic domain-describing yield criterion is the derived rule, also cf. [Weinberg et al. \[2006\]](#). While [Ortiz and Pandolfi \[2004\]](#) has demonstrated that such a framework is convenient for extending the isotropic MCC model into the finite deformation regime and enforcing material-frame indifference, our derivation in the subsequent sections indicates that this variational framework also simplify the extension of the MCC to incorporate a gradient-dependent flow rule. For the local anisotropic MCC plasticity, the flow rule is

$$\dot{\epsilon}^P = \dot{\lambda} \mathbf{n} \mid \dot{\lambda} \geq 0, \quad (5.11)$$

where the local equivalent plastic strain's rate $\dot{\lambda}$ is non-negative, $\mathbf{n}$ the second-order tensorial direction of plastic flow is not traceless, and $(\dot{\cdot})$ indicates the time derivative of $(\cdot)$. If yielding, plastic strain direction $\mathbf{n}$ satisfies the kinematic constraint

$$\mathbf{n} : \mathbf{B}_\lambda : \mathbf{n} = 1, \quad \mathbf{B}_\lambda = \frac{3}{M^2} \mathbf{P}^{\text{P vol } -1} : \mathbf{P}^{\text{vol}} : \mathbf{P}^{\text{P vol } -1} + \frac{2}{3} \mathbf{P}^{\text{P dev } -1} : \mathbf{P}^{\text{dev}} : \mathbf{P}^{\text{P dev } -1}, \quad (5.12)$$

where $M > 0$ is a dimensionless physical constant, later shown to be the slope of the critical state line, in Section 5.2.5 and 5.2.5. Our convention is that the symmetric fourth-order tensor $\mathbf{A}_{(\cdot)}$ maps stress σ from the real stress to fictitious isotropic stress-space, whereas $\mathbf{B}_{(\cdot)}$ maps kinematics-related tensorial internal variables, which convention is intended to be in-line with the general quadratic model for plasticity presented in Section 2.5.1 of [Simo and Hughes \[1998\]](#).

The rate $\dot{\lambda}$ is positive for change in the local equivalent plastic strain,

$$\dot{\lambda} = \sqrt{\dot{\epsilon}^P : \mathbf{B}_\lambda : \dot{\epsilon}^P} \geq 0.$$

Note that $\dot{\lambda}$ is degree one homogeneous in $\dot{\epsilon}^P$, because $(\partial \dot{\lambda} / \partial \dot{\epsilon}^P) : \dot{\epsilon}^P = \dot{\lambda}$. Furthermore, $\dot{\lambda} \in \mathbb{R}$ as $\dot{\epsilon}^P : \mathbf{B}_\lambda : \dot{\epsilon}^P \geq 0$ for all $\dot{\epsilon}^P$.

5.2.3 Anisotropic plastic regularization for non-coaxial plastic flow

The energy functional W^P in Eq. (5.3) contains two types of energy functionals, i.e. [\[Forest, 2009, Aldakheel and Miehe, 2017\]](#),

$$W_{\tilde{\alpha}}^P(\alpha, \tilde{\alpha}) = \overbrace{\frac{k_\lambda}{2} (\tilde{\lambda} - \lambda)^2 + \frac{k_v}{2} (\tilde{\epsilon}_v^P - \epsilon_v^P)^2}^{\text{penalty functionals}} + \overbrace{\frac{K_\lambda l_\lambda^2}{2} \nabla^X \tilde{\lambda} \cdot \omega_\lambda \cdot \nabla^X \tilde{\lambda} + \frac{K_v l_v^2}{2} \nabla^X \tilde{\epsilon}_v^P \cdot \omega_v \cdot \nabla^X \tilde{\epsilon}_v^P}_{\text{anisotropic regularization functionals}}, \quad (5.13)$$

where $\alpha = \{\lambda, \epsilon_v^P\}$ and $\tilde{\alpha} = \{\tilde{\lambda}, \tilde{\epsilon}_v^P\}$, k_λ and k_v are stiffness parameters, and penalize difference between local internal variables and field values of $\tilde{\lambda}$ and $\tilde{\epsilon}_v^P$. $K_\lambda > 0$ and $K_v > 0$ are moduli which have the same unit as a stiffness, and $l_\lambda > 0$ and $l_v > 0$ are length scale parameters corresponding to the regularized variables $\tilde{\lambda}$ and $\tilde{\epsilon}_v^P$ respectively.

The first type of energy functionals are the relaxation functionals that penalize the difference between local internal variables and field variables measured by the L_2 norm. The second type of energy functionals are weighted inner products of the gradient of the corresponding field variables that introduce the gradient dependence and the non-coaxiality of the plastic flow. The Euler-Lagrange equation of these two types of energy functional lead to two modified Helmholtz equations of which the corresponding Galerkin form is solved via the finite element method in this work. Notice that the second-order positive-definite diffusivity tensors ω_λ and ω_v , are weighting functions in the weighted

inner product. They are used to introduce anisotropy on the gradient-dependence of the field variables $\tilde{\lambda}$ and $\tilde{\epsilon}_v^p$. Since both ω_λ and ω_v are not isotropic tensors, a bias is therefore introduced in the regularization such that the gradient dependencies are stronger in the principal directions corresponding to the largest eigenvalues of ω_λ and ω_v than those corresponding to smaller eigenvalues. In other words, the plastic flow direction of each material point is therefore not only depending on the stress gradient of the yield function at the material point but also depends on the directionally biased influence of the constitutive responses of the neighbors characterized by ω_λ and ω_v . Consequently, the resultant gradient-enhanced plasticity model is associative, but the plastic flow direction is not coaxial to the stress gradient of the conventional MCC yield function due to the anisotropic regularization.

Note that the anisotropy introduced by the anisotropic diffusivity tensors ω_λ and ω_v are of different natures than those introduced at the local constitutive laws. In particular, the anisotropic effect introduced via the anisotropic diffusivity tensors exhibits scale effects. Furthermore, since the principal directions of the anisotropic diffusivity tensors are independent of the mapping operators used to introduce anisotropy into the local constitutive law, the Euler angles between the isotropic plane of the local plasticity model and the principal directions of tensors ω_λ and ω_v can be leveraged to generate a fuller anisotropic constitutive responses for complex materials for which the anisotropic responses are originated from multiple geometrical attributes across length scales (e.g. joints, layers, fabrics, slip systems, lattice).

Remark 1. To simplify the material identification procedure, analogously to the parameter selection in Mische et al. [2013] or Aldakheel [2017], we associate the volumetric term with the volumetric stiffness constant,

$$K_\lambda \sim a_\lambda = a_v, \quad K_v \sim a_v = \frac{1}{3} \mathbf{1} : \mathbf{C}^e : \mathbf{1}, \quad (5.14)$$

where our notation appropriates that of Walpole [1984]. Within the Walpole notation for elastic isotropy, $a = \mathbf{1} : \mathbf{C}^e : \mathbf{1}/3$, where the bulk modulus $K = a/3$. Calibration of the regularization might alternatively include inverse problems, against strain gradients obtained from triaxial tests [Wang et al., 2016]. That said, our 2D and 3D numerical examples consistently converge through strain softening.

Note that, for a given set of fixed length scale parameters, l_λ and l_v , the discrepancy between the local and the projected internal variables, and the spatial distribution of the projected internal variables are affected by the ratios K_λ/k_λ and K_v/k_v . If these ratios are low, then the discrepancy between the local and the field variables is lower but the field variables may exhibit sharper spatial gradients. If these ratios are high, then the sharp gradient of the projected internal variables may not be admissible, but the discrepancy between the local and field variables could be larger.

Now consider the tensor ω_λ (or ω_v) that describes microstructural attributes aligned with the unit vector l_λ for $l_\lambda \cdot l_\lambda = 1$.

Hence, one may express the tensor ω_λ as a function of the tensors $\phi_\lambda = l_\lambda \otimes l_\lambda$ and $\chi_\lambda = \mathbf{1} - l_\lambda \otimes l_\lambda$. Due to the orthogonality and idempotence of ϕ_λ and χ_λ , in the combined formalism of Clayton and Knap [2015b], Teichtmeister et al. [2017a], and Bryant and Sun [2018], the definition of this mapping relates

$$\omega_\lambda = (p_\lambda \otimes p_\lambda) : \mathbf{1} = \mathbf{1} + \phi_\lambda \phi_\lambda + \chi_\lambda \chi_\lambda, \quad p_\lambda = \phi_\lambda \sqrt{1 + \phi_\lambda} + \chi_\lambda \sqrt{1 + \chi_\lambda}, \quad (5.15)$$

where coefficients ϕ_λ and χ_λ are dimensionless, as is tensor ω_λ . Coefficients $\phi_\lambda \geq -1$ and $\chi_\lambda \geq -1$, such that ω_λ is positive semidefinite. Physically, $\phi_\lambda \gg 0$ penalizes field

variable diffusion on planes normal to $\mathbf{l}_\lambda$, whereas $\chi_\lambda \gg 0$ penalizes damage diffusion on planes not normal to normal vector $\mathbf{l}_\lambda$. For $\phi_\lambda = \chi_\lambda = 0$, $\mathbf{p}_\lambda = \mathbf{1}$ such that the mapping reduces to isotropy. $\phi_\lambda \neq \chi_\lambda$ indicates ω_λ is a tensor with one unique and one repeated eigenvalue. These statements apply equally to ω_v .

5.2.4 Incremental stored plastic work

Our objective is to an incremental form of the stored work functionals, such that the incremental constitutive update can be associated with the Euler-Lagrange equation of a discrete functional (seenext section). In particular, we employ a backward-implicit Euler method to integrate plastic work. Consider a finite set consisting of discrete snapshots of time instants $\{t_0, \dots, t_n, t_{n+1}\}$, for n the time step, with the data at prior time $t_1, t_2, \dots, t_n$ given. At the new time t_{n+1} , the elastoplastic strain terms are

$$\epsilon_{n+1} = \epsilon_{n+1}^e + \epsilon_{n+1}^p, \quad (\cdot)_{n+1} = (\cdot)_n + \Delta(\cdot)|_n^{n+1}. \quad (5.16)$$

The local stored plastic work has been defined in the mapped-isotropic strain-space by [Ortiz and Pandolfi \[2004\]](#), as

$$W_{\alpha, n+1}^p - W_{\alpha, n}^p = \underbrace{\frac{1}{2} [W_c^p(\epsilon_{v, n+1}^p) - W_c^p(\epsilon_{v, n}^p)]}_{\text{matched coefficients 1/2 and } M/2 \text{ return MCC yield criterion}} - \overbrace{\frac{M}{2} (\lambda_{n+1} - \lambda_n) \left[\partial_{\epsilon_v^p} W_c^p(\epsilon_{v, n+1}^p) \right]}^{\text{ensure } p_{c, n+1} \leq 0}. \quad (5.17)$$

Note the Lagrange multiplier term $M(\lambda_{n+1} - \lambda_n)p_{c, n+1}/2 = -\Delta\lambda\sigma_{q, n+1}$ enforces non-positivity of the preconsolidation pressure $p_{c, n+1}$. The limits of the partial derivatives

$$\frac{\partial W_{\alpha, n+1}^p}{\partial \Delta\lambda} = \frac{M}{2} \partial_{\epsilon_v^p} W_{c, n+1}^p = \sigma_{q, n+1}, \quad \frac{\partial W_{\alpha, n+1}^p}{\partial \epsilon_{v, n+1}^p} = \frac{1}{2} \partial_{\epsilon_v^p} W_{c, n+1}^p - \frac{M}{2} \Delta\lambda \partial_{\epsilon_v^p}^2 W_{c, n+1}^p = \sigma_{p, n+1}, \quad (5.18)$$

are

$$\lim_{\Delta\lambda \rightarrow 0} \sigma_{q, n+1} = \lim_{\Delta\lambda \rightarrow 0} \left(\frac{\partial W_{\alpha, n+1}^p}{\partial \Delta\lambda} \right) = -\frac{M}{2} p_{c, n+1}, \quad \lim_{\Delta\lambda \rightarrow 0} \sigma_{p, n+1} = \lim_{\Delta\lambda \rightarrow 0} \left(\frac{\partial W_{\alpha, n+1}^p}{\partial \epsilon_{v, n+1}^p} \right) = \frac{1}{2} p_{c, n+1}. \quad (5.19)$$

These results are later used to estimate the connection between the two-invariant yield surface and the incremental energy functional, following the treatments in [Ortiz and Stainier \[1999\]](#) and [Ortiz and Pandolfi \[2004\]](#) in Section 5.2.5.

Remark 2. Albeit not considered in this study, anisotropy-adapted viscoplastic regularization is similarly intuitive. One possible way to incorporate viscoplasticity in a variational framework is via appending to the local stored incremental plastic work a term similar to,

$$\frac{\eta_\lambda}{2\Delta t} (\lambda_{n+1} - \lambda_n)^2 + \frac{\eta_v}{2\Delta t} (\epsilon_{v, n+1}^p - \epsilon_{v, n}^p)^2,$$

where η_λ and η_v are material parameters and $\Delta t = t_{n+1} - t_n$ is the time increment (cf. [Ortiz and Pandolfi \[2004\]](#), [Ortiz and Stainier \[1999\]](#)).

5.2.5 Variational constitutive update with anisotropic micromorphic regularization

We derive an discrete energy functional whose Euler-Lagrange equation leads to the governing equation of the local constitutive law and the Helmholtz equations that regularize the boundary value problem. Meanwhile Following Eq. (3.3-13) in [Yang et al. \[2006\]](#) e.g., we suppose the mechanical equilibrium equation's numerical solution is staggered w.r.t. solution of the regularizing Helmholtz equations. This staggering is in-line with solution of the mechanically-coupled scalar diffusion equations in [Armero and Simo \[1992\]](#), for instance.

Furthermore, we employ the variational principle to obtain the Helmholtz equations for $\tilde{\lambda}$ and $\tilde{\epsilon}_v^p$ from the corresponding energy functional. Following the variational update for regularized viscoplastic models (e.g. [Aldakheel \[2017\]](#)), the local constitutive update and the corresponding equations that govern the micromorphic regularization can be obtained from one discrete incremental energy functional. From this single incremental energy functional, we may derive the discrete Euler-Lagrange equation, a system of nonlinear equations that constitute both the local constitutive update and the the micromorphic field equations [[Aldakheel and Miehe, 2017](#)].

This system of equations is solved via an operator-split scheme. The local constitutive updates are solved in a semi-implicit manner, in the sense that the incremental constitutive laws that updates the local internal variables and the stress are updated via a Newton solver while the incremental solutions of the Helmholtz equations are fixed. Meanwhile, when the incremental solutions of the Helmholtz equations are updated, the internal variables are frozen. Further discussions about the operator-split scheme can be found in [Miehe et al. \[2010b\]](#), [Sun \[2015a\]](#), [Aldakheel and Miehe \[2017\]](#), [Wheeler et al. \[2014\]](#), [Miehe et al. \[2015b\]](#), [Choo and Sun \[2017\]](#). In this work, the Cauchy stress is integrated incrementally via a return mapping algorithm. As result, we have,

$$\epsilon_{n+1}^e = \epsilon_{n+1}^{e \text{ tr}} - \Delta \epsilon^p, \quad \epsilon_{n+1}^{e \text{ tr}} = \epsilon_{n+1} - \epsilon_n^p, \quad (5.20)$$

where $\epsilon_{n+1}^{e \text{ tr}}$ is the trial elastic strain. The strain and hence elastic trial strain are fixed while solving the local optimization problem, as we employ a a standard return mapping algorithm. Thus

$$\left. \frac{\partial \epsilon_{n+1}^p}{\partial \Delta \epsilon^p} \right|_{\epsilon_{n+1}^{e \text{ tr}}} = I, \quad \left. \frac{\partial \epsilon_{n+1}}{\partial \Delta \epsilon^p} \right|_{\Delta \epsilon} = 0,$$

during the constitutive update, the algorithm by which we obtain $\sigma_{n+1} = C^e : \epsilon_{n+1}^e$. Then, the identity ϵ_{n+1}^e in Eq. (5.20) defines the local minimization problem in the elastic strains. This implies the generalized coordinates

$$\zeta_{n+1} = \{\epsilon_{n+1}^e, \tilde{\lambda}_{n+1}, \tilde{\epsilon}_{v \text{ } n+1}^p\}, \quad (5.21)$$

where Eq. (5.13) is used. For a given strain at an incremental step ϵ_{n+1} , the constitutive updates is associated with the Euler-Lagrange equation of the constrained optimization problem which reads,

$$\zeta_{n+1} = \arg \min_{\zeta_{n+1}} W(\zeta_{n+1}). \quad (5.22)$$

subjected to the following constraint,

$$\Delta \lambda = \sqrt{\Delta \epsilon_{n+1}^p : B_\lambda : \Delta \epsilon_{n+1}^p} \geq 0, \quad \Delta \epsilon_{v \text{ } n+1}^p = b_v : \Delta \epsilon_{n+1}^p. \quad (5.23)$$

Note that the inequality in (5.23) is satisfied if $\mathbf{B}_\lambda$ is positive semi-definite (or positive definite). The micromorphic field equation that governs the relations between the global and local internal variables are obtained from the stationary conditions of Eq. (5.22) with respect to the global internal variables $\tilde{\lambda}_{n+1}$ and $\tilde{\epsilon}_{v\ n+1}^p$, i.e.,

$$\frac{\delta W(\zeta_{n+1})}{\delta \tilde{\lambda}_{n+1}} = k_\lambda (\tilde{\lambda}_{n+1} - \lambda_{n+1}) - K_\lambda l_\lambda^2 \nabla^X \cdot (\boldsymbol{\omega}_\lambda \cdot \nabla^X \tilde{\lambda}_{n+1}) = 0, \text{ in } \mathcal{B}, \quad (5.24)$$

$$\frac{\delta W(\zeta_{n+1})}{\delta \tilde{\epsilon}_{v\ n+1}^p} = k_v (\tilde{\epsilon}_{v\ n+1}^p - \epsilon_{v\ n+1}^p) - K_v l_v^2 \nabla^X \cdot (\boldsymbol{\omega}_v \cdot \nabla^X \tilde{\epsilon}_{v\ n+1}^p) = 0, \text{ in } \mathcal{B}, \quad (5.25)$$

where the trivial boundary conditions, e.g. $\hat{\mathbf{n}} \cdot \boldsymbol{\omega}_\lambda \cdot \nabla^X \tilde{\lambda}_{n+1} = 0$ on $\partial\mathcal{B}$, are applied. On the other hand, the system of equations for the local constitutive updates can be obtained incrementally from the first variation of $W(\zeta_{n+1})$ with respect to the elastic strain ϵ_{n+1}^e (see Appendix ?? for the detailed derivation), i.e.,

$$\begin{aligned} \frac{\delta W(\zeta_{n+1})}{\delta \epsilon_{n+1}^e} = & \overbrace{\sigma_{n+1} - \sigma_{q\ n+1} \frac{\partial \Delta \lambda}{\partial \Delta \epsilon^p} - \sigma_{p\ n+1} \mathbf{b}_v}^{\text{purely local terms}} \\ & + \overbrace{k_\lambda (\tilde{\lambda}_{n+1} - \lambda_{n+1}) \frac{\partial \Delta \lambda}{\partial \Delta \epsilon^p} + k_v (\tilde{\epsilon}_{v\ n+1}^p - \epsilon_{v\ n+1}^p) \mathbf{b}_v}^{\text{micromorphic terms}} = 0, \end{aligned} \quad (5.26)$$

where we used the definitions of σ_{n+1} , $\partial W_{\alpha\ n+1}^p / \partial \Delta \lambda$, and $\partial W_{\alpha\ n+1}^p / \partial \epsilon_{v\ n+1}^p$.

Remark 3. Direct substitution of law Eq. (5.7) curtails hardening force-related expressions otherwise proliferating in local-to-global variational updates, cf. e.g. Aldakheel [2017]. Similarly per Section 5.2.4, viscoplasticity incorporates by a quadratic function of the plastic strain increment. Thus the regularized constitute update devolves to minimization Eq. (5.22).

Yield criterion

In the Euler-Lagrange Eq. (5.26), substitute $\partial \Delta \lambda / \partial \Delta \epsilon^p = \mathbf{B}_\lambda : \mathbf{n}_{n+1}$ such that

$$0 = \sigma_{n+1} - \sigma_{q\ n+1} \mathbf{B}_\lambda : \mathbf{n}_{n+1} - \sigma_{p\ n+1} \mathbf{b}_v + k_\lambda (\tilde{\lambda}_{n+1} - \lambda_{n+1}) \mathbf{B}_\lambda : \mathbf{n}_{n+1} + k_v (\tilde{\epsilon}_{v\ n+1}^p - \epsilon_{v\ n+1}^p) \mathbf{b}_v. \quad (5.27)$$

Interiority within the elastic domain can be determined as follows. Rearranging the Euler-Lagrange equation to solve for $\mathbf{n}_{n+1}$. At the trail state $\epsilon_{n+1}^e = \epsilon_{n+1}^{e\ \text{tr}}$, the tensorial flow direction evaluates as

$$\mathbf{n}_{n+1}^{\text{tr}} = \frac{1}{\sigma_{q\ n+1}^{\text{tr}} - k_\lambda (\tilde{\lambda}_{n+1} - \lambda_{n+1}^{\text{tr}})} \mathbf{B}_\lambda^{-1} : \left[\sigma_{n+1}^{\text{tr}} - \sigma_{p\ n+1}^{\text{tr}} \mathbf{b}_v + k_v (\tilde{\epsilon}_{v\ n+1}^p - \epsilon_{v\ n+1}^{p\ \text{tr}}) \mathbf{b}_v \right]. \quad (5.28)$$

If within the elastic domain, by the kinematic constraint on the tensorial direction in Eq. (5.12), the yield criterion can be written as

$$\varphi(\mathbf{n}_{n+1}^{\text{tr}}) = \mathbf{n}_{n+1}^{\text{tr}} : \mathbf{B}_\lambda : \mathbf{n}_{n+1}^{\text{tr}} - 1 \leq 0, \quad (5.29)$$

expressed quadratically in $\mathbf{n}_{n+1}^{\text{tr}}$ to recover the yield criterion in quadratic terms of the mapped stress invariants (next section). Note that Eq. (5.27), Eq. (5.28), and Eq. (5.29) combine to completely describe the variational update applied in Section 5.4.1. Following, change in the field variables $\tilde{\lambda}$ and $\tilde{\epsilon}_v^p$ not just regularizes but can also trigger the onset of local yielding.

Mapped two-invariant yield criterion

Now we establish the connection between our derivation based on the variational principle and the formulation in Semnani et al. [2016]. The superscript tr is dropped for brevity. First, assume that $\mathbf{b}_v = \mathbf{P}^{\text{vol} -1} : \mathbf{1}$. Then, we eliminate the gradient dependence of the plastic flow by setting $k_\lambda = k_v = 0$. By substituting the definition of $\mathbf{n}_{n+1}$ in (5.28) into the yield function and simplifying the expression, we obtain,

$$\frac{\sigma_{q,n+1}^2}{M^2} \varphi_{n+1} = \left(p_{n+1}^* - \sigma_{p,n+1} \right)^2 + \frac{1}{M^2} \left(q_{n+1}^{*2} - \sigma_{q,n+1}^2 \right) \leq 0. \quad (5.30)$$

Applying the relations in Eq. (5.19), Eq. 5.30 can be rewritten as,

$$\lim_{\Delta\lambda \rightarrow 0} \left(\frac{\sigma_{q,n+1}^2}{M^2} \varphi_{n+1} \right) = p_{n+1}^* \left(p_{n+1}^* - p_{c,n+1} \right) + \frac{q_{n+1}^{*2}}{M^2} \leq 0, \quad (5.31)$$

at time step t_{n+1} , i.e. a necessary condition for the stress to be admissible. As a result, our model may be reduced to the anisotropic yield function in Semnani et al. [2016] when the gradient regularization vanishes. It can also be reduced to the classical modified Cam-clay model (cf. Eq. (6.18) in Borja [2013] when the mapping becomes identity).

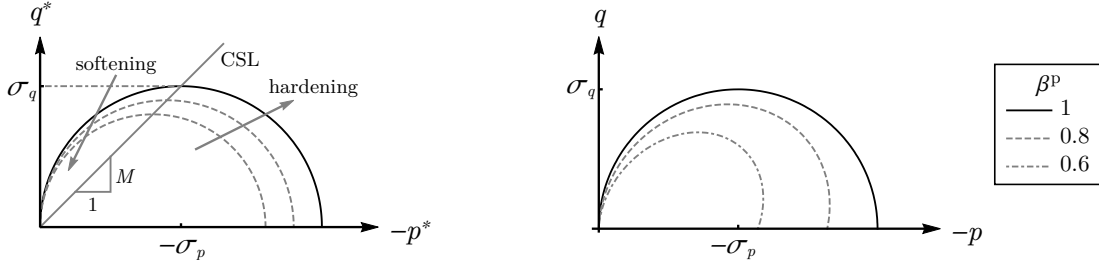


Fig. 5.3. Geometric interpretation of the mapped two-invariant yield criterion, Eq. (5.30): (a) introducing M as the slope of the critical state line (CSL) in the starred scalar space, after Ortiz and Pandolfi [2004], and with softening and hardening trends after Borja [2013]; and, (b) unmapped two-invariant yield criterion parameterized by the mapping coefficient, varying $\beta^{\text{dev}} = \beta^{\text{vol}}$ at fixed $\alpha^{\text{dev}} = \alpha^{\text{vol}} = \gamma^{\text{dev}} = \gamma^{\text{vol}} = 1$, after Semnani et al. [2016].

5.3 Balance and evolution equations

We solve the balance of linear equilibrium equation, i.e.

$$\nabla^X \cdot \sigma + \rho g = 0, \quad (5.32)$$

where σ is the Cauchy stress, ρ the density, and g the gravitational acceleration vector. We employ a hyperelastic energy functional such that the stress $\sigma = \partial W^e / \partial \epsilon^e$. Hence, the local internal variables λ and ϵ_p^p are updated incrementally once the solution of the nonlinear static equilibrium equation is obtained via an implicit solver, with the same local-to-global hierarchy described in Weinberg et al. [2006], regarding the variational constitutive update.

However, to introduce nonlocality in the constitutive law, we supply an additional set of governing equations to evolve the field variables $\tilde{\lambda}$ and $\tilde{\epsilon}_v^p$. Both evolution equations are characterized variationally via introduction of diffusive functionals in Eq. (5.13), which depends on the spatial gradients of $\tilde{\lambda}$ and $\tilde{\epsilon}_v^p$. In practice, we solve an equivalent nondimensionalized system (cf. Forest [2009]) with Laplacian coefficients of the regularization equation

$$\tilde{l}_\lambda = l_\lambda \sqrt{K_\lambda/k_\lambda}, \quad \tilde{l}_v = l_v \sqrt{K_v/k_v}. \quad (5.33)$$

Thus, each field variable accords with some Helmholtz equation, e.g.

$$\tilde{\alpha} - \tilde{l}^2 \nabla^X \cdot (\omega \cdot \nabla^X \tilde{\alpha}) = \alpha, \quad (5.34)$$

for $\tilde{\alpha}$ a field variable, α the corresponding internal variable, $\tilde{l}$ a length, and ω a dimensionless second-order tensor. The only imposed boundary condition for the Helmholtz equations is the trivial Neumann boundary condition.

This technique is referred to as micromorphic regularization in Forest [2009] and Miehe et al. [2013]. The nondimensionalizations $\sqrt{K_\lambda/k_\lambda}$ and $\sqrt{K_v/k_v}$ help us to explain convergence towards mesh independence even during material softening. See the results section, and for explanatory analysis Aldakheel [2017].

5.4 Local-global constitutive updates

Due to the introduction of the micromorphic regularization, the constitutive update is obtained from a global-local split algorithm, such that: the global evolution equations updates the strain and the micromorphic field variables, while the local return mapping algorithm provides the incremental updates of the elastic (and equivalently the plastic) strain, internal variables, and plastic flow direction [Aldakheel, 2017]. The governing equations for the micromorphic regularization are obtained from the stationary condition of the energy functional listed in Eq. (5.22). The elastic strain ϵ_{n+1}^e is defined from the strain update in Eq. (5.20), and results from optimality of the local minimization problem.

5.4.1 Local system

The local incremental stress update is obtained by a conventional return mapping algorithm. For clarity, the algorithm is summarized in Alg. 4. As necessary, we map or reconstruct required symmetric tensors from their Kelvin-notated vector equivalents per Eq. (??), and super-symmetric fourth-order tensors per. Eq. (??), Appendix ??.

Local nonlinearity is treated as follows. Define the local numerical system and residual in matrix-vector notation:

$$\left. \begin{aligned} \bar{\mathbf{x}}^k &= \left[\bar{\epsilon}_{n+1}^e \right]_{6 \times 1}^k \\ \bar{\mathbf{r}}^k &= \left[\mathbf{C}^e \bar{\epsilon}_{n+1}^e - \sigma_{q \ n+1} \mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1} - \sigma_{p \ n+1} \bar{\mathbf{b}}_v \right. \\ &\quad \left. + k_\lambda \left(\tilde{\lambda}_{n+1} - \lambda_{n+1} \right) \mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1} + k_v \left(\tilde{\epsilon}_{v \ n+1}^p - \epsilon_{v \ n+1}^p \right) \bar{\mathbf{b}}_v \right]_{6 \times 1}^k \end{aligned} \right\} \quad (5.35)$$

where k is the iteration, $\bar{\mathbf{x}}^k$ the Kelvin-notated vector equivalent of the local unknowns at k , $\bar{\mathbf{r}}^k$ the Kelvin-notated vector equivalent of the local residual at k , $\bar{\mathbf{b}}_v$ the Kelvin-notated

Algorithm 4 Local return-mapping

Require: From the global system, the strain increment $\Delta\epsilon$ as well as, for the micromorphic regularization, field variables $\tilde{\lambda}$ and $\tilde{\epsilon}_v^p$

- 1: A trial state is established
 - a. set $\epsilon_{n+1}^{e \text{ tr}} = \epsilon_{n+1}^e + \Delta\epsilon$, $\lambda_{n+1}^{\text{tr}} = \lambda_n$, $\epsilon_{v \text{ } n+1}^p \text{ tr} = \epsilon_v^p$, $\sigma_{q \text{ } n+1}^{\text{tr}} = \sigma_0$, and $\sigma_{p \text{ } n+1}^{\text{tr}} = p_0$
 - b. evaluate n_{n+1}^{tr} per Eq. (5.28)
 - 2: **if** $\varphi(n_{n+1}^{\text{tr}}) \leq 0$ **then**
 - a. the deformation is elastic per Eq. (5.29), set internal variables $(\cdot)_{n+1} = (\cdot)_{n+1}^{\text{tr}}$
 - b. set $\sigma_{n+1} = \mathbf{C}^e : \epsilon_{n+1}^e$ and the solid tangent $\mathbf{C}^e$
 - 3: **else**
 - a. the deformation is inelastic, obtain the local system by parameterizing the Euler-Lagrange equation with $n(\Delta\epsilon_{n+1}^p)$, $\lambda(\Delta\epsilon_{n+1}^p)$, and $\epsilon_v^p(\Delta\epsilon_{n+1}^p)$ under dependence $\Delta\epsilon^p(\epsilon_{n+1}^e)$
 - b. at local system iteration $k = 0$, guess the initial solution per Eq. (5.37)
 - c. using Newton's method or a variant, iteratively solve the local nonlinear system for ϵ_{n+1}^e
 - d. set $\sigma_{n+1} = \mathbf{C}^e : \epsilon_{n+1}^e$ and the solid tangent $\mathbf{C}_{n+1}^{\text{ep}}$ per Eq. (5.41)
 - 4: **end if**
 - 5: Return mapping completes
 - a. w.r.t the equilibrium equation, pass to the global system the stress σ_{n+1} and the solid tangent
 - b. for the micromorphic regularization, also pass the local variables λ_{n+1} and ϵ_v^p
-

vector equivalent of $\mathbf{b}_v$, and $\mathbf{B}_\lambda$ the Kelvin-notated vector equivalent of $\mathbf{B}_\lambda$ (for bold font and overline notation convention for Kelvin-notated equivalents, see Appendix ??). The local tangent operator is then

$$\left. \frac{\partial \bar{\mathbf{r}}}{\partial \bar{\mathbf{x}}} \right|_{\bar{\epsilon}_{n+1}^{e \text{ tr}}}^k = \left[\frac{\partial \bar{\mathbf{r}}}{\partial \bar{\epsilon}_{n+1}^e} \right]_{6 \times 6}^k, \quad (5.36)$$

as derived in Appendix ??, where

$$\begin{aligned} \frac{\partial \bar{\mathbf{r}}}{\partial \bar{\epsilon}_{n+1}^e} &= \mathbf{C}^e + \frac{\sigma_{q \text{ } n+1}}{\Delta\lambda} \left[\mathbf{B}_\lambda - (\mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1}) (\mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1})^T \right] - \frac{M}{2} \partial_{\epsilon_v^p} p_{c \text{ } n+1} \left[(\mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1}) \bar{\mathbf{b}}_v^T + \bar{\mathbf{b}}_v (\mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1})^T \right] \\ &\quad + \frac{1}{2} \left(\partial_{\epsilon_v^p} p_{c \text{ } n+1} - M \Delta\lambda \partial_{\epsilon_v^p}^2 p_{c \text{ } n+1} \right) \bar{\mathbf{b}}_v \bar{\mathbf{b}}_v^T - \frac{k_\lambda (\tilde{\lambda}_{n+1} - \lambda_{n+1})}{\Delta\lambda} \left[\mathbf{B}_\lambda - (\mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1}) (\mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1})^T \right] \\ &\quad + k_\lambda (\mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1}) (\mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1})^T + k_v \bar{\mathbf{b}}_v \bar{\mathbf{b}}_v^T, \end{aligned}$$

and

$$\bar{\mathbf{n}}_{n+1} = \frac{\mathbf{B}_\lambda \Delta \bar{\epsilon}^p}{\Delta\lambda} = \frac{\mathbf{B}_\lambda \Delta \bar{\epsilon}^p}{\sqrt{\Delta \bar{\epsilon}^p{}^T \mathbf{B}_\lambda \Delta \bar{\epsilon}^p + k_{\bar{\epsilon}}}} \quad \text{for} \quad \Delta \bar{\epsilon}^p = \bar{\epsilon}_{n+1}^{e \text{ tr}} - \bar{\epsilon}_{n+1}^e \quad \text{with} \quad \mathbf{B}_\lambda \bar{\mathbf{n}}_{n+1} = \mathbf{B}_\lambda^T \bar{\mathbf{n}}_{n+1}.$$

Small parameter $k_{\bar{\epsilon}}$ is taken as 1×10^{-15} .

Our local tangent matrix $(\partial \bar{\mathbf{r}} / \partial \bar{\mathbf{x}})^k$'s symmetry redounds to the model design's variational consistency, the hallmark of which is the cascading dependence $p_c(\Delta \bar{\epsilon}^p)$ and $\Delta \bar{\epsilon}^p(\bar{\epsilon}_{n+1}^e)$, in this the MCC context.

Trial state

Converging a local system requires passing an initial guess to Newton's method. Our guess, at iteration $k = 0$, approximates the elastic trial state:

$$\bar{\epsilon}_{n+1}^{e, k=0} = \left[\bar{\epsilon}_{n+1}^{e, \text{tr}} - \Delta\lambda \bar{\mathbf{n}}_{n+1}^{\text{tr}} \right]_{6 \times 1}^{k=0} \quad \text{for} \quad \Delta\bar{\epsilon}^{\text{p}, k=0} = \left[\Delta\lambda \bar{\mathbf{n}}_{n+1}^{\text{tr}} \right]_{6 \times 1}^{k=0}. \quad (5.37)$$

Thus at $k = 0$ we set $\bar{\epsilon}_{n+1}^e \approx \bar{\epsilon}_{n+1}^{e, \text{tr}}$ and $\Delta\bar{\epsilon}^{\text{p}} \approx \bar{\mathbf{0}}$, with $\Delta\lambda^{k=0}$ a small number taken as 1×10^{-10} .

Stored work update

In-line with the semilogarithmic relation, we use as derivatives

$$p_{c, n+1} = p_{c, n} \exp \left(\frac{\epsilon_{v, n+1}^e - \epsilon_{v, n+1}^{e, \text{tr}}}{C_d} \right), \quad \partial_{\epsilon_v^e} p_{c, n+1} = -\frac{p_{c, n}}{C_d} \exp \left(\frac{\epsilon_{v, n+1}^e - \epsilon_{v, n+1}^{e, \text{tr}}}{C_d} \right).$$

As such, the stored plastic work of consolidation's increments follow

$$W_{c, n+1}^{\text{p}} - W_{c, n}^{\text{p}} = p_{c, n} C_d \left[1 - \exp \left(\frac{\epsilon_{v, n+1}^e - \epsilon_{v, n+1}^{e, \text{tr}}}{C_d} \right) \right] = C_d (p_{c, n} - p_{c, n+1}). \quad (5.38)$$

5.4.2 Consistent tangent operator (CTO)

A consistent tangent operator $\mathbf{C}_{n+1}^{\text{ep}} = \partial \bar{\sigma}_{n+1} / \partial \bar{\epsilon}_{n+1}$ is useful to converge the global system for the displacements, cf. Eq. (7.127-136) in [de Souza Neto et al. \[2008\]](#), also see the next section. The CTO is evaluated after convergence of the local system, when

$$\frac{\partial \bar{\epsilon}_{n+1}^{e, \text{tr}}}{\partial \bar{\epsilon}_{n+1}^e} = \frac{\partial (\bar{\epsilon}_n^e + \bar{\epsilon}_{n+1} - \bar{\epsilon}_n)}{\partial \bar{\epsilon}_{n+1}^e} = \mathbf{I}, \quad \text{hence} \quad \frac{\partial \bar{\epsilon}_{n+1}^{e, \text{tr}}}{\partial \bar{\epsilon}_{n+1}^e} = \frac{\partial \bar{\epsilon}_{n+1}^{e, \text{tr}}}{\partial \bar{\epsilon}_{n+1}^e} : \frac{\partial \bar{\epsilon}_{n+1}}{\partial \bar{\epsilon}_{n+1}^e} = \frac{\partial \bar{\epsilon}_{n+1}}{\partial \bar{\epsilon}_{n+1}^e},$$

whereas

$$\left. \frac{\partial \Delta\bar{\epsilon}^{\text{p}}}{\partial \bar{\epsilon}_{n+1}^e} \right|_{\bar{\epsilon}^{e, \text{tr}}} = \left. \frac{\partial (\bar{\epsilon}_{n+1}^{e, \text{tr}} - \bar{\epsilon}_{n+1}^e)}{\partial \bar{\epsilon}_{n+1}^e} \right|_{\bar{\epsilon}^{e, \text{tr}}} = -\mathbf{I}, \quad \left. \frac{\partial \Delta\bar{\epsilon}^{\text{p}}}{\partial \bar{\epsilon}_{n+1}^{e, \text{tr}}} \right|_{\bar{\epsilon}_{n+1}^e} = \left. \frac{\partial (\bar{\epsilon}_{n+1}^{e, \text{tr}} - \bar{\epsilon}_{n+1}^e)}{\partial \bar{\epsilon}_{n+1}^{e, \text{tr}}} \right|_{\bar{\epsilon}_{n+1}^e} = \mathbf{I}.$$

As CTO is evaluated after convergence of the local system, we drop superscripts k and $n + 1$ and let $\bar{\mathbf{x}} = \bar{\epsilon}^e$, for the remainder of this section.

To identify the CTO, we rewrite the purely local Euler-Lagrange equation in residual form, as

$$\bar{\sigma} - \bar{\mathbf{s}}(\Delta\bar{\epsilon}^{\text{p}}) = \bar{\mathbf{0}} \quad (5.39)$$

where $\bar{\mathbf{s}}$ corresponds to the plastic back-stress as described in [Ortiz and Stainier \[1999\]](#), and is

$$\bar{\mathbf{s}}(\Delta\bar{\epsilon}^{\text{p}}) = \sigma_q \frac{\mathbf{B}_\lambda \Delta\bar{\epsilon}^{\text{p}}}{\sqrt{\Delta\bar{\epsilon}^{\text{p}^T} \mathbf{B}_\lambda \Delta\bar{\epsilon}^{\text{p}}}} + \sigma_p \bar{\mathbf{b}}_v \quad \text{and} \quad \Delta\bar{\epsilon}^{\text{p}} = \bar{\epsilon}^{e, \text{tr}} - \bar{\epsilon}^e \Rightarrow \left. \frac{\partial \bar{\mathbf{s}}}{\partial \bar{\mathbf{x}}} \right|_{\bar{\epsilon}^{e, \text{tr}}} = \mathbf{C}^e + \frac{\partial \bar{\mathbf{s}}}{\partial \Delta\bar{\epsilon}^{\text{p}}},$$

assuming invertible $\partial\bar{\mathbf{r}}/\partial\bar{\mathbf{x}}$, the local system's tangent at the converged state from Eq. (5.36). Apply the identity $\bar{\boldsymbol{\sigma}} = \mathbf{C}^e \bar{\boldsymbol{\epsilon}}^e$ and differentiate the Euler-Lagrange equation in residual form,

$$\frac{\partial}{\partial \bar{\boldsymbol{\epsilon}}^e} (\mathbf{C}^e \bar{\boldsymbol{\epsilon}}^e) - \frac{\partial \bar{\mathbf{s}}}{\partial \Delta \bar{\boldsymbol{\epsilon}}^p} \left(\frac{\partial \Delta \bar{\boldsymbol{\epsilon}}^p}{\partial \bar{\boldsymbol{\epsilon}}^e} \Big|_{\bar{\boldsymbol{\epsilon}}^e \text{ tr}} + \frac{\partial \Delta \bar{\boldsymbol{\epsilon}}^p}{\partial \bar{\boldsymbol{\epsilon}}^e \text{ tr}} \Big|_{\bar{\boldsymbol{\epsilon}}^e} \frac{\partial \bar{\boldsymbol{\epsilon}}^e \text{ tr}}{\partial \bar{\boldsymbol{\epsilon}}^e} \right) = \mathbf{C}^e - \frac{\partial \bar{\mathbf{s}}}{\partial \Delta \bar{\boldsymbol{\epsilon}}^p} \left(-\mathbf{I} + \mathbf{I} \frac{\partial \bar{\boldsymbol{\epsilon}}}{\partial \bar{\boldsymbol{\epsilon}}^e} \right) = \bar{\mathbf{0}}. \quad (5.40)$$

Rearranging for

$$\frac{\partial \bar{\boldsymbol{\epsilon}}}{\partial \bar{\boldsymbol{\epsilon}}^e} = \left(\frac{\partial \bar{\mathbf{s}}}{\partial \Delta \bar{\boldsymbol{\epsilon}}^p} \right)^{-1} \left(\mathbf{C}^e + \frac{\partial \bar{\mathbf{s}}}{\partial \Delta \bar{\boldsymbol{\epsilon}}^p} \right),$$

$\mathbf{C}^{\text{ep}}$ is evaluated via the chain rule. Noting that $\mathbf{C}^{\text{ep}} = \partial \bar{\boldsymbol{\sigma}} / \partial \bar{\boldsymbol{\epsilon}} = \mathbf{C}^e (\partial \bar{\boldsymbol{\epsilon}}^e / \partial \bar{\boldsymbol{\epsilon}}) = \mathbf{C}^e (\partial \bar{\boldsymbol{\epsilon}} / \partial \bar{\boldsymbol{\epsilon}}^e)^{-1}$,

$$\mathbf{C}^{\text{ep}} = \left[\mathbf{C}^e \left(\frac{\partial \bar{\boldsymbol{\epsilon}}}{\partial \bar{\boldsymbol{\epsilon}}^e} \right)^{-1} \right]_{6 \times 6} = \left[\mathbf{C}^e \left(\frac{\partial \bar{\mathbf{r}}}{\partial \bar{\mathbf{x}}} \Big|_{\bar{\boldsymbol{\epsilon}}^e \text{ tr}} \right)^{-1} \left(\frac{\partial \bar{\mathbf{r}}}{\partial \bar{\mathbf{x}}} \Big|_{\bar{\boldsymbol{\epsilon}}^e \text{ tr}} - \mathbf{C}^e \right) \right]_{6 \times 6}, \quad (5.41)$$

where we have substituted to write $\mathbf{C}^{\text{ep}}$ in $\mathbf{C}^e$, and $\partial \bar{\mathbf{r}} / \partial \bar{\mathbf{x}}$. As such, the consistent tangent computation exclusively reuses the solid elastic tangent, $\mathbf{C}^e$, and the local tangent at the converged state, $\partial \bar{\mathbf{r}} / \partial \bar{\mathbf{x}}$. On this point, contrast against [Semnani et al. \[2016\]](#). Consequently, our implementation is straightforward and generic with respect to additional (e.g. micromorphic or viscoplastic) physics.

Chapter 6

Modeling crystallization of rock salt

Cracking and damage from crystallization of minerals in pores center on a wide range of problems, from weathering and deterioration of structures to storage of CO₂ via in situ carbonation. Here we develop a theoretical and computational framework for modeling these crystallization-induced deformation and fracture in fluid-infiltrated porous materials. Conservation laws are formulated for coupled chemo-hydro-mechanical processes in a multiphase material composed of the solid matrix, liquid solution, gas, and crystals. We then derive an expression for the effective stress tensor that is energy-conjugate to the strain rate of a porous material containing crystals growing in pores. This form of effective stress incorporates the excess pore pressure exerted by crystal growth—the crystallization pressure—which has been recognized as the direct cause of deformation and fracture during crystallization in pores. Continuum thermodynamics is further exploited to formalize a constitutive framework for porous media subject to crystal growth. The chemo-hydro-mechanical model is then coupled with a phase-field approach to fracture which enables simulation of complex fractures without explicitly tracking their geometry. For robust and efficient solution of the initial-boundary value problem at hand, we utilize a combination of finite element and finite volume methods and devise a block-partitioned preconditioning strategy. Through numerical examples we demonstrate the capability of the proposed framework for simulating complex interactions among unsaturated flow, crystallization kinetics, and cracking in the solid matrix.

6.1 Introduction

Growth of mineral crystals in pores can give rise to severe damage and cracks in the host material. These coupled chemo-hydro-mechanical processes are now central to a number of problems in our society. A well-known example is weathering and deterioration of historic and building structures due to salt crystallization. Many of these structures are comprised of materials prone to invasion of salt water (e.g., stone), so they can be severely damaged when salt minerals grow inside the pores. See Fig. 6.1 for example. Preventing this type of damage has been a critical element of conservation of cultural heritage and structures around the world [NAP \[1982\]](#), [Tsui et al. \[2003\]](#), [Angeli et al. \[2007\]](#), [Espinosa-Marzal and Scherer \[2010\]](#), [Steiger et al. \[2014\]](#), [Flatt et al. \[2014\]](#). Also, crystallization of minerals in the subsurface can trigger ground heaving that severely damages buildings and geotechnical structures [Sass and Burbaum \[2010\]](#), [Serafeimidis and Anagnostou \[2013\]](#), [Alonso and Ramon \[2013\]](#). Furthermore, reaction-driven cracking during mineral hydration, carbonation, and oxidation is a key consideration for deploying

a promising strategy for geologic carbon storage that transforms CO₂ into solid carbonate minerals [Kelemen and Matter \[2008\]](#).



Fig. 6.1. Example of damage in building stones by crystallization of salts. (Photograph by Suzanne MacLeod, distributed under a CC-BY 2.0 license.)

Addressing the problem of cracking and damage from crystallization in pores requires us, as a first step, to understand its fundamental mechanism. Figure 6.2 schematically shows how a crystal is present inside a pore space. Importantly, the crystal is usually confined within a liquid solution, maintaining a thin liquid film between its surface and the solid pore wall (see Scherer [Scherer \[1999, 2004\]](#) for detailed explanations). The liquid solution surrounding the crystal allows it to continuously grow and push on the pore wall. This process of crystal growth generates an excess pressure on the solid matrix, which is commonly referred to as the crystallization pressure in a number of previous studies (e.g., [Scherer \[2004\]](#), [Correns and Steinborn \[1939\]](#), [Correns \[1949\]](#), [Steiger \[2005a,b\]](#), [Flatt and Scherer \[2008\]](#), [Kelemen and Hirth \[2012\]](#), [Desarnaud et al. \[2016\]](#)). These studies have proposed expressions of the crystallization pressure, showing that this pressure can far exceed the tensile strength of many porous materials. This affirms that the crystallization pressure is the direct cause of fracturing and damage during crystallization in pores, and thus, an accurate prediction of the crystallization pressure *per se* is an important research problem. However, an expression for the crystallization pressure alone is insufficient for tackling real-world problems such as those mentioned above, because the scales of these problems are orders of magnitude larger than the scale of pores. Also necessitated is a continuum-scale modeling framework that allows us to simulate and predict how crystallization pressures would evolve in space and time and ultimately affect the problem at the field scale. The necessity of such a predictive modeling framework is the motivation of this work.

Continuum modeling of fracturing by in-pore crystallization poses two significant challenges. First, it requires a mathematical formulation that encapsulates complex interactions among chemical reactions, fluid flow, and solid deformation in porous materials. A coupled formulation for such chemo-hydro-mechanical processes is not only difficult to develop in a theoretically consistent manner, but also challenging to solve numerically since the flow, transport, and reaction processes involve multiple length and time scales. Second, as can be seen from Fig. 6.1, the fracturing process of interest entails extremely

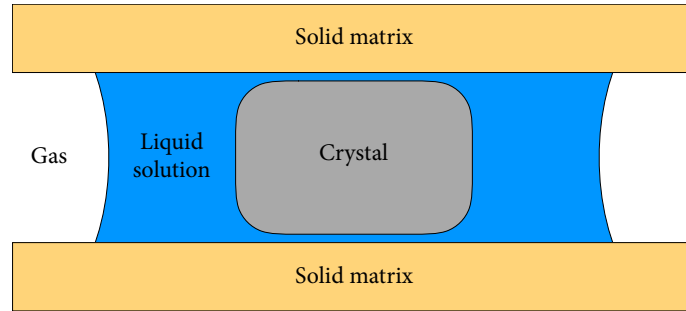


Fig. 6.2. Schematic illustration of a crystal inside a pore. Note that the crystal is confined within the liquid solution, not being in contact with the solid matrix. Similar illustrations have been presented in [Scherer \[2004\]](#), [Steiger \[2005a\]](#), [Derluyn et al. \[2014\]](#).

complicated geometries that cannot be idealized as a set of sharp discontinuities. Obviously, algorithmic capture of such complex geometries are very unwieldy and onerous. Since these two types of challenges are interwoven herein, theoretical and computational modeling of crystallization-induced fracturing is a particularly demanding task.

Modeling frameworks that address both of these theoretical and computational challenges remain scarce. To our knowledge, Coussy [Coussy \[2006\]](#) introduced the first chemo-hydro-mechanical framework for deformation and fracture from in-pore crystallization of minerals. His work presented significant contributions to theoretical aspects, but it remained unclear how the theory can be applied to construct an initial-boundary value problem of the crystallization-induced fracturing process. Some of more recent studies proposed computational models that paved the way to numerical simulation of the crystallization problem (e.g., [Derluyn et al. \[2014\]](#), [Koniorczyk and Gawin \[2012\]](#)). Yet, challenges remain for both the theory and computation. First, theoretical formulations in past studies show significant disagreement, particularly with respect to the definition of effective stress which governs the constitutive behavior of the solid matrix. Second, the existing computational models appear insufficient for addressing complex cracking and damage processes from crystallization. For example, Koniorczyk and Gawin [Koniorczyk and Gawin \[2012\]](#) used linear elasticity without consideration of fracture. Derluyn et al. [Derluyn et al. \[2014\]](#) employed a simple local damage model along with elasticity, but their work was limited to identification of crack nucleation by comparing the effective stress and the tensile strength in a 1-D setting. Such an approach would be inappropriate for delineation of the onset and evolution of damage and cracking zones in multi-dimensional problems. Also, their use of the standard, continuous finite element method (FEM) may not be an optimal choice, since it could suffer from numerical stability problems arising from the advective transport of minerals. To address the stability problem, Derluyn et al. [Derluyn et al. \[2014\]](#) smoothed equations related to crystallization kinetics. However, they also found that the smoothing parameters can plague the physics significantly. Another issue is that their work used a one-way coupled staggered scheme, which is presumably because computational cost for fully coupled chemo-hydro-mechanics is prohibitively expensive without a carefully designed solution strategy. All of the aforementioned aspects indicate that a significant amount of more work is necessary to advance the theory and computation of crystallization-induced cracking and damage in porous materials.

The purpose of this work is to develop a more theoretically grounded, computationally

efficient modeling framework for crystallization-induced deformation and fracture in porous materials. To this end, we first draw on recent advances in continuum modeling of coupled multiphysics in porous materials. For theoretically consistent modeling of the coupled chemo-hydro-mechanical problem at hand, we use continuum principles of thermodynamics which have been the basis of rigorous poromechanical frameworks in the literature [Borja \[2004, 2006b\]](#), [Borja and Koliji \[2009\]](#), [Song and Borja \[2014\]](#), [Choo et al. \[2016b\]](#), [Borja and Choo \[2016b\]](#). The use of thermodynamic arguments provides a systematic procedure to derive physically meaningful forms of effective stress and constitutive laws for coupled multiphysics processes. Notably, the poromechanical frameworks developed in this way have demonstrated their ability to reproduce a variety of real-world observations (e.g., [Borja and Choo \[2016b\]](#), [Borja and White \[2010a\]](#), [Borja et al. \[2016\]](#)). Then, for obtaining a stable numerical solution to the problem, we utilize the finite volume method (FVM) for the fluid flow and transport problem which involves advection phenomena, and the standard finite element method for the solid deformation problem which does not. Furthermore, we introduce a three-field block-partitioned solver that enables one to solve the coupled chemo-hydro-mechanical problem with an affordable computational cost.

As for the modeling of cracking and damage, we adopt a phase-field approach to fracture which has emerged as an efficient means for simulating complicated cracks without explicitly tracking their geometry (e.g., [Bourdin et al. \[2008b\]](#), [Amor et al. \[2009b\]](#), [Miehe et al. \[2010c,d\]](#), [Borden et al. \[2012\]](#), [Wilson et al. \[2013\]](#), [Borden et al. \[2014\]](#), [Mikelić et al. \[2015\]](#), [Miehe and Mauthe \[2015\]](#), [Borden et al. \[2016b\]](#), [Wilson and Landis \[2016\]](#), [Lee et al. \[2016a,b,c\]](#), [De Lorenzis et al. \[2016\]](#), [Lee et al. \[2017\]](#), [Mauthe and Miehe \[2017\]](#), [Ehlers and Luo \[2017\]](#), [Santillán et al. \[2017\]](#), [Na et al. \[2017b\]](#), [Cajuhi et al. \[2017\]](#), [Choo and Sun \[2018a\]](#)). This feature of phase-field modeling is particularly desirable for our purpose since most (if not all) observed cracks due to in-pore crystallization are extremely difficult to delineate geometrically. Also, in most cases, crystallization in pores gives rise to distributed microcracks, resulting in rounded damage zones like those shown in [Fig. 6.1](#). Such complex damage patterns can also be captured by the use of a phase-field model, because it can be regarded as a particular class of nonlocal gradient damage models [de Borst and Verhoosel \[2016b\]](#). As such, in this work we adopt a phase-field model of fracture to simulate both distributed damages and localized cracks by in-pore crystallization, in a mesh insensitive manner. In doing so, we also propose a way to estimate the length regularization parameter of the phase-field model, by drawing on an empirical relationship between the fracture toughness and the tensile strength of geomaterials.

The paper is organized as follows. In [Section 6.2](#), we formulate balance laws for a porous continuum containing liquid solution, gas, and crystals in the pores. Subsequently, in [Section 6.3](#) we use thermodynamic arguments in conjunction with the balance laws to derive suitable expressions for effective stress and multiphysics constitutive relations. In [Section 6.4](#), we derive a phase-field formulation of brittle fracture as a balance law such that it can be augmented to the coupled chemo-hydro-mechanics formulation developed in previous sections. We then discretize the resulting formulation in [Section 6.5](#) via a combination of finite element and finite volume methods, and devise a block-partitioned iterative solver for the coupled problem. In [Section 6.6](#) we present numerical examples demonstrating the validity and performance of the proposed framework for modeling complex chemo-hydro-mechanical and fracturing processes from crystallization of minerals in pores.

6.2 Conservation laws

In this section, we develop conservation laws for coupled chemo-hydro-mechanical processes in mineral-containing, partially saturated porous materials. We first introduce a continuum mixture representation of this type of porous material, and then formulate balance laws for its mass, linear momentum, and energy. At this point, we note that the purpose of this and the next section is to formulate a general mathematical model amenable to being combined with various methods for modeling cracking and damage. Later in Section 6.4, we will adopt a specific method, namely phase-field modeling.

6.2.1 Continuum representation

Using mixture theory we conceptualize the material of interest as a multiphase continuum in which the solid matrix, liquid solution, gas, and mineral crystals are overlapped. See Fig. 6.3 for an elementary volume representation of this four-phase mixture. We define the volume fractions of the constituent phases as

$$\phi^s = \frac{dV_s}{dV}, \quad \phi^l = \frac{dV_l}{dV}, \quad \phi^g = \frac{dV_g}{dV}, \quad \phi^c = \frac{dV_c}{dV}, \quad \phi^s + \sum_{\alpha=l,g,c} \phi^\alpha = 1, \quad (6.1)$$

where the index s refers to the solid matrix, l the liquid solution, g the gas, and c the crystals. It is noted that an index is used as a subscript when referring to an intrinsic property of a constituent phase, whereas it is used as a superscript when referring to a partial property of the mixture. We define the saturation ratios of the phases in the pore space—the liquid solution, the gas, and the crystals—as

$$S^l = \frac{\phi^l}{1 - \phi^s}, \quad S^g = \frac{\phi^g}{1 - \phi^s}, \quad S^c = \frac{\phi^c}{1 - \phi^s}, \quad S^l + S^g + S^c = 1. \quad (6.2)$$

Let ρ_s , ρ_l , and ρ_c denote the intrinsic mass densities of the solid, the liquid solution, and the mineral crystals, respectively. The partial mass densities of the constituent phases are given by

$$\rho^s = \phi^s \rho_s, \quad \rho^l = \phi^l \rho_l, \quad \rho^g = \phi^g \rho_g, \quad \rho^c = \phi^c \rho_c, \quad \rho^s + \sum_{\alpha=l,g,c} \rho^\alpha = \rho, \quad (6.3)$$

where ρ is the mass density of the entire mixture.

As shown in Fig. 6.3, the liquid solution itself is also a mixture composed of water and dissolved minerals. Let the index w denote the water and d denote the dissolved minerals. Their global volume fractions are given by

$$\phi^w = \frac{dV_w}{dV}, \quad \phi^d = \frac{dV_d}{dV}, \quad \phi^w + \phi^d = \phi^l, \quad (6.4)$$

and their saturation ratios are

$$S^w = \frac{\phi^w}{1 - \phi^s}, \quad S^d = \frac{\phi^d}{1 - \phi^s}, \quad S^w + S^d = S^l. \quad (6.5)$$

We also define local volume fractions of the water and dissolved minerals as

$$\psi^w = \phi^w / \phi^l = S^w / S^l, \quad \psi^d = \phi^d / \phi^l = S^d / S^l, \quad \psi^w + \psi^d = 1. \quad (6.6)$$

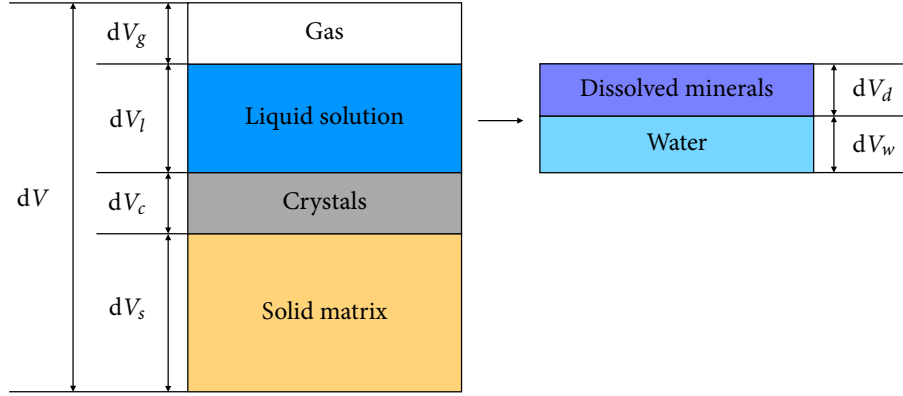


Fig. 6.3. Elementary volume representation of a four-phase mixture composed of the solid matrix, liquid solution, gas, and crystals. The liquid solution is also a sub-mixture of water and dissolved minerals.

Similarly, their partial densities are defined as

$$\rho^w = \phi^w \rho_w = \phi^l \psi^w \rho_w, \quad \rho^d = \phi^d \rho_d = \phi^l \psi^d \rho_d, \quad \rho^w + \rho^d = \rho^l. \quad (6.7)$$

Note that the intrinsic density of dissolved minerals ρ_d is identical to ρ_c , but we have used ρ_d for notational purposes. Using the above variables, we can write the mass fraction of minerals dissolved in the solution as

$$c = \frac{\rho^d}{\rho^l} = \frac{\rho^d}{\rho^w + \rho^d} = \frac{\psi^d \rho_d}{\psi^w \rho_w + \psi^d \rho_d}. \quad (6.8)$$

6.2.2 Balance of mass

To derive conservation laws for this mixture, we use a kinematic description that traces the motion of the solid matrix. Let an overdot denote the material time derivative with respect to the motion of the solid matrix. Then we can write balance of mass for the solid, the liquid solution, the gas, the dissolved minerals, and the mineral crystals as

$$\dot{\rho}^s + \rho^s \nabla^x \cdot \mathbf{v} = m_s, \quad (6.9)$$

$$\dot{\rho}^l + \rho^l \nabla^x \cdot \mathbf{v} + \nabla^x \cdot \mathbf{w}_l = m_l, \quad (6.10)$$

$$\dot{\rho}^g + \rho^g \nabla^x \cdot \mathbf{v} + \nabla^x \cdot \mathbf{w}_g = m_g, \quad (6.11)$$

$$\dot{\rho}^d + \rho^d \nabla^x \cdot \mathbf{v} + \nabla^x \cdot \mathbf{w}_d = m_d, \quad (6.12)$$

$$\dot{\rho}^c + \rho^c \nabla^x \cdot \mathbf{v} + \nabla^x \cdot \mathbf{w}_c = m_c, \quad (6.13)$$

respectively. Here, $\mathbf{v}$ is the velocity of the solid matrix, m_α is the rate of mass production for phase α , and $\mathbf{w}_\alpha$ is the Eulerian relative mass flow vector of phase α with respect to the solid matrix, which is given by

$$\mathbf{w}_\alpha = \rho^\alpha \tilde{\mathbf{v}}_\alpha, \quad \tilde{\mathbf{v}}_\alpha = \mathbf{v}_\alpha - \mathbf{v}, \quad \alpha = l, g, d, c, \quad (6.14)$$

with $\mathbf{v}_\alpha$ being defined as the velocity of phase α . For the dissolved minerals in the liquid solution, the relative mass flow is decomposed into convective and diffusive/dispersive parts as

$$\mathbf{w}_d = c \mathbf{w}_l + \mathbf{j}, \quad (6.15)$$

where j is the diffusive/dispersive mass flux of the minerals in the solution.

At this point, we introduce a few assumptions that simplify mathematical expressions in the succeeding development. First, we assume that the water and crystal phases are incompressible and the solid matrix undergoes infinitesimal deformations. Second, adopting the assumption in the fluid flow model of Castellazzi et al. [Castellazzi et al. \[2013\]](#), we postulate that only the dissolved and crystallized minerals exchange masses among the five constituent phases, i.e., $m_s = m_g = 0$ and $m_l = m_d = -m_c$. Third, we consider that momentum and pressure of the water and dissolved minerals in the liquid solution are under equilibrium.

6.2.3 Balance of linear momentum

Balance of linear momentum for the solid, liquid solution, gas, and crystals can be written as

$$\nabla^x \cdot \sigma^s + \rho^s g + h^s = \rho^s a, \quad (6.16)$$

$$\nabla^x \cdot \sigma^l + \rho^l g + h^l = \rho^l a_l - m_l v_l, \quad (6.17)$$

$$\nabla^x \cdot \sigma^g + \rho^g g + h^g = \rho^g a_g, \quad (6.18)$$

$$\nabla^x \cdot \sigma^c + \rho^c g + h^c = \rho^c a_c + m_c v_c, \quad (6.19)$$

where σ^α is the partial (Cauchy) stress tensor of phase α , a and a_α is the acceleration of the solid matrix and phase α , respectively, and h^α is the drag on phase α by the surrounding phases, which is subject to the constraint

$$h^s + h^l + h^g + h^c = 0. \quad (6.20)$$

Summing up the above equations gives an expression for balance of linear momentum for the entire mixture, which reads

$$\nabla^x \cdot \sigma + \rho g = \rho a + \sum_{\alpha=l,g,c} \rho^\alpha \tilde{a}_\alpha + m_c (\tilde{v}_l + \tilde{v}_c), \quad (6.21)$$

where $\tilde{a}_\alpha = a_\alpha - a$. Here, $\sigma = \sigma^s + \sigma^l + \sigma^g + \sigma^c$ is the total stress tensor in the mixture.

6.2.4 Balance of energy

Let P denote the total power, K the kinetic energy, and I the internal energy in the mixture. Then balance of energy for the mixture can be expressed as

$$P = \dot{K} + \dot{I}. \quad (6.22)$$

The rate of change of kinetic energy in an arbitrary volume $\mathcal{V}$ of the mixture is given by

$$\dot{K} = \int_{\mathcal{V}} \rho^s a \cdot v \, dV + \sum_{\alpha=l,g,c} \int_{\mathcal{V}} \rho^\alpha a_\alpha \cdot v_\alpha \, dV + \sum_{\alpha=l,g,c} \int_{\mathcal{V}} \frac{1}{2} m_\alpha v_\alpha \cdot v_\alpha \, dV, \quad (6.23)$$

and the rate of change of internal energy is

$$\dot{I} = \int_{\mathcal{V}} \rho \dot{e} \, dV, \quad (6.24)$$

where $\dot{e}$ is the rate of change of internal energy per unit total mass of the mixture.

The total power is the sum of the mechanical power P^m and the non-mechanical power P^n , i.e., $P = P^m + P^n$. In this work, we consider isothermal conditions in which $P^n = 0$. Let us introduce the infinitesimal strain tensor for the solid $\boldsymbol{\varepsilon} = \nabla^s \mathbf{u} = (\nabla^x \mathbf{u} + \nabla^{x^T} \mathbf{u})/2$ where $\mathbf{u}$ is the displacement vector. Similarly we introduce infinitesimal strain tensors $\boldsymbol{\varepsilon}_\alpha$ for phase α . The mechanical power is then given by

$$\begin{aligned} P^m = & \int_{\mathcal{V}} \left(\boldsymbol{\sigma}^s : \dot{\boldsymbol{\varepsilon}} + \sum_{\alpha=l,g,c} \boldsymbol{\sigma}^\alpha : \dot{\boldsymbol{\varepsilon}}_\alpha \right) dV \\ & + \int_{\mathcal{V}} (\nabla^x \cdot \boldsymbol{\sigma}^s \cdot \mathbf{v} + \mathbf{h}^s \cdot \mathbf{v} + \rho^s \mathbf{g} \cdot \mathbf{v}) dV \\ & + \sum_{\alpha=l,g,c} \int_{\mathcal{V}} (\nabla^x \cdot \boldsymbol{\sigma}^\alpha \cdot \mathbf{v}_\alpha + \mathbf{h}^\alpha \cdot \mathbf{v}_\alpha + \rho^\alpha \mathbf{g} \cdot \mathbf{v}_\alpha) dV. \end{aligned} \quad (6.25)$$

We now substitute above expressions into $\dot{I} = P - \dot{K}$, impose the balance of linear momentum for each phase, and localize the resulting integral. Then, we can express the rate of change of internal energy as

$$\rho \dot{e} = \boldsymbol{\sigma}^s : \dot{\boldsymbol{\varepsilon}} + \sum_{\alpha=l,g,c} \boldsymbol{\sigma}^\alpha : \dot{\boldsymbol{\varepsilon}}_\alpha - \sum_{\alpha=l,g,c} \frac{1}{2} m_\alpha \mathbf{v}_\alpha \cdot \mathbf{v}_\alpha. \quad (6.26)$$

This energy balance equation will be the starting point of the next section where we derive suitable forms of the effective stress tensor and constitutive laws.

6.3 Effective stress and constitutive framework

The purpose of this section is to identify physically meaningful constitutive relations that close the formulation. For this purpose, we begin by deriving a suitable form of the effective stress tensor, which is a necessary step for constitutive modeling of a deformable porous material infiltrated by fluid. We do this derivation based on a thermodynamic argument that the effective stress should be energy-conjugate to the strain rate tensor. From the effective stress derivation we identify several groups of energy-conjugate pairs, and we use them as a guide to develop constitutive relations for coupled chemo-hydro-mechanical modeling.

6.3.1 Effective stress

Our goal here is to find the form of stress measure energy-conjugate to the strain rate tensor for the solid matrix, which, by definition, corresponds to the effective stress in a fluid-infiltrated porous material. For this purpose we apply the procedure developed by Borja [Borja \[2006b\]](#), which was originally proposed for porous media without any crystals, to materials containing solid crystals in their pores.

In doing so, we postulate that the phases in the pores—the liquid solution, gas, and crystals—can only carry volumetric stresses. This postulate is reasonable for most pore fluids of interests. For the crystals, the postulate may be justified by the fact that they are usually floating in the liquid solution, as depicted in [Fig. 6.2](#) previously. See the same reasoning in [Na and Sun \[2017b\]](#) for incorporating ice crystals into effective stress in frozen soils. Then, we can express the partial stress tensors of these in-pore phases as

$$\boldsymbol{\sigma}^\alpha = -\phi^\alpha p_\alpha \mathbf{1}, \quad \alpha = l, g, c, \quad (6.27)$$

where p_α is the intrinsic pressure of phase α and $\mathbf{1}$ is the second-order identity tensor. Substituting the above expression into Eq. (6.26) and neglecting thermal terms gives

$$\rho \dot{e} = \sigma : \dot{\epsilon} - \sum_{\alpha=l,g,c} \phi^\alpha (\nabla^x \cdot \tilde{\mathbf{v}}_\alpha) p_\alpha - \sum_{\alpha=l,g,c} \frac{1}{2} m_\alpha \mathbf{v}_\alpha \cdot \mathbf{v}_\alpha. \quad (6.28)$$

From Eq. (6.28), we can see that the second term on the right hand side is directly related to the solid deformation (because $\nabla^x \cdot \tilde{\mathbf{v}}_\alpha = \nabla^x \cdot \mathbf{v}_\alpha - \nabla^x \cdot \mathbf{v}$), whereas the last term is irrelevant to the solid deformation. Thus we now seek to extract the contribution of $\phi^\alpha (\nabla^x \cdot \tilde{\mathbf{v}}_\alpha)$ to the solid deformation. The mass balance equations for the liquid solution, gas, and crystals can be rewritten as

$$\dot{\phi}^\alpha + \phi^\alpha \nabla^x \cdot \mathbf{v} + \phi^\alpha \nabla^x \cdot \tilde{\mathbf{v}}_\alpha + \nabla^x \phi^\alpha \cdot \tilde{\mathbf{v}}_\alpha = \frac{m_\alpha}{\rho_\alpha}, \quad \alpha = l, g, c, \quad (6.29)$$

and rearranging the above equations gives

$$\phi^\alpha \nabla^x \cdot \tilde{\mathbf{v}}_\alpha = -\phi^\alpha \nabla^x \cdot \mathbf{v} - \dot{\phi}^\alpha - \nabla^x \phi^\alpha \cdot \tilde{\mathbf{v}}_\alpha + \frac{m_\alpha}{\rho_\alpha}, \quad \alpha = l, g, c. \quad (6.30)$$

Now we look for an alternative expression for $\dot{\phi}^\alpha$ as it is also related to the solid deformation. Recalling that $\phi^\alpha = (1 - \phi^s) S^\alpha$ for $\alpha = l, g, c$, the material time derivative of ϕ^α is given by

$$\dot{\phi}^\alpha = (1 - \phi^s) \dot{S}^\alpha - S^\alpha \dot{\phi}^s, \quad \alpha = l, g, c, \quad (6.31)$$

The remaining challenge is to have an explicit expression for the last term, $\dot{\phi}^s$. Borja [Borja \[2006b\]](#) has shown that, in the case of barotropic (isothermal) flow for the solid, the mass balance equation for the solid can be expressed as

$$\dot{\phi}^s + (\phi^s - b) \nabla^x \cdot \mathbf{v} = 0, \quad (6.32)$$

where $b = K/K_s$, with K and K_s being the bulk moduli of the solid matrix and the solid constituent, respectively. Inserting Eq. (6.32) into Eq. (6.31), we get

$$\dot{\phi}^\alpha = (1 - \phi^s) \dot{S}^\alpha + S^\alpha (\phi^s - b) \nabla^x \cdot \mathbf{v}, \quad \alpha = l, g, c. \quad (6.33)$$

Using the above equations, we can rewrite Eq. (6.30) as

$$\phi^\alpha \nabla^x \cdot \tilde{\mathbf{v}}_\alpha = -[\phi^\alpha + S^\alpha (\phi^s - b)] \nabla^x \cdot \mathbf{v} - (1 - \phi^s) \dot{S}^\alpha - \nabla^x \phi^\alpha \cdot \tilde{\mathbf{v}}_\alpha + \frac{m_\alpha}{\rho_\alpha}, \quad \alpha = l, g, c. \quad (6.34)$$

Here, the coefficients of $\nabla^x \cdot \mathbf{v}$ can be simplified to

$$\phi^\alpha + S^\alpha (\phi^s - b) = (1 - \phi^s) S^\alpha + S^\alpha (\phi^s - b) = (1 - b) S^\alpha = B S^\alpha, \quad \alpha = l, g, c, \quad (6.35)$$

where $B = 1 - b = 1 - K/K_s$ is the well-known Biot coefficient. Finally, we can decompose $\phi^\alpha \nabla^x \cdot \tilde{\mathbf{v}}_\alpha$ in Eq. (6.28) as

$$\phi^\alpha \nabla^x \cdot \tilde{\mathbf{v}}_\alpha = -B S^\alpha \nabla^x \cdot \mathbf{v} - (1 - \phi^s) \dot{S}^\alpha - \nabla^x \phi^\alpha \cdot \tilde{\mathbf{v}}_\alpha + \frac{m_\alpha}{\rho_\alpha}, \quad \alpha = l, g, c. \quad (6.36)$$

Observe that the first term on the right hand side contains $\nabla^x \cdot \mathbf{v}$, which is the rate of volume change of the solid matrix.

Substituting Eq. (6.36) into Eq. (6.28), we can rewrite the energy balance equation as

$$\begin{aligned} \rho \dot{e} = & \sigma' : \dot{\epsilon} + \sum_{\alpha=l,g,c} (1 - \phi^s) \dot{S}^\alpha p_\alpha + \sum_{\alpha=l,g,c} (\nabla^x \phi^\alpha \cdot \tilde{\mathbf{v}}_\alpha) p_\alpha \\ & + \sum_{\alpha=l,g,c} \left(\frac{m_\alpha}{\rho_\alpha} \right) p_\alpha - \sum_{\alpha=l,g,c} \frac{1}{2} m_\alpha \mathbf{v}_\alpha \cdot \mathbf{v}_\alpha, \end{aligned} \quad (6.37)$$

where

$$\sigma' = \sigma + B \bar{p} \mathbf{1} = \sigma + B \sum_{\alpha=l,g,c} (S^\alpha p_\alpha) \mathbf{1}, \quad (6.38)$$

is the stress measure energy-conjugate to the strain rate tensor, which, by definition, is the effective stress. The “pore pressure” in this form of effective stress, denoted by $\bar{p}$, is

$$\begin{aligned} \bar{p} &= \sum_{\alpha=l,g,c} (S^\alpha p_\alpha) = S^l p_l + S^g p_g + S^c p_c \\ &= (S^l + S^c) p_l + S^g p_g + S^c (p_c - p_l), \end{aligned} \quad (6.39)$$

which is the mean of the liquid, gas, and crystal pressures weighted by their volume fractions (saturation ratios).

In the absence of crystals (i.e., $S^c = 0$), Eq. (6.38) boils down to the thermodynamically consistent effective stress tensor in unsaturated porous media derived by Borja [Borja \[2006b\]](#), which specializes to the well-known Terzaghi’s effective stress [Terzaghi \[1943\]](#) in the limit of full saturation and $B = 1$. However, when a crystal is growing within a liquid solution in confined pore space, the crystal growth exerts significant excess pressure on the solution film between the crystal and surrounding solid particles. This pressure—which is commonly referred to as the crystallization pressure in the literature—can make p_c far greater than p_l . Then the effective stress could become tensile even as the total stress is compressive, resulting in damage and fracture. Our derivation thus formally shows that the crystallization pressure can be a direct driver of deformation and fracture from crystallization in pores. Given its significance, the crystallization pressure is hereafter denoted by p_{cr} , i.e.,

$$p_{\text{cr}} := p_c - p_l. \quad (6.40)$$

6.3.2 Constitutive framework

Having derived the form of effective stress, we further exploit the energy-conjugate pairs in the energy balance equation to identify suitable forms of constitutive laws. We particularly extend the procedure of Borja and co-workers [Song and Borja \[2014\]](#), [Choo et al. \[2016b\]](#), which has been used for unsaturated porous materials without crystals, to materials containing crystals in pores. This procedure simply interprets the type and form of constitutive relationship that each energy-conjugate pair suggests, without performing a thorough thermodynamic analysis. The reason is that the standard argument will eventually require variables in each energy-conjugate pair to be linked via a constitutive relation, for ensuring the second law of thermodynamics. Therefore, in what follows, we interpret the implication of each energy-conjugate pair, and adopt one of the widely used constitutive models in the literature. The selected constitutive model may not agree with the most general form allowed by a thermodynamic analysis. Note, however,

that the specific model is just chosen for constructing a particular class of the general framework we propose in this work—it can readily be replaced by another constitutive model depending on the purpose of modeling. Also, for brevity, the following discussion focuses on new aspects emerging from the presence of the crystals. Detailed explanations of the other aspects unrelated to the crystals can be found in [Borja \[2004, 2006b\]](#), [Borja and Koliji \[2009\]](#), [Song and Borja \[2014\]](#), [Choo et al. \[2016b\]](#).

For a clearer interpretation, we first simplify some terms in Eq. (6.37). Since $S^l + S^g + S^c = 1$, we have $\dot{S}^l = -\dot{S}^g - \dot{S}^c$. Thus the second term in Eq. (6.37) can be expressed as

$$\sum_{\alpha=l,g,c} (1 - \phi^s) \dot{S}^\alpha p_\alpha = (1 - \phi^s) \dot{S}^c p_{cr} + (1 - \phi^s) \dot{S}^g p_{ca}, \quad (6.41)$$

where the capillary pressure (suction) is defined in the last term as $p_{ca} := p_g - p_l$. Also, the third and fourth terms in Eq. (6.37) can be combined as

$$\sum_{\alpha=l,g,c} \frac{m_\alpha}{\rho_\alpha} p_\alpha - \sum_{\alpha=l,g,c} \frac{1}{2} m_\alpha v_\alpha \cdot v_\alpha = \sum_{\alpha=l,g,c} m_\alpha \left[\frac{p_\alpha}{\rho_\alpha} - \frac{1}{2} \tilde{v}_\alpha \cdot \tilde{v}_\alpha \right]. \quad (6.42)$$

Therefore, under isothermal conditions, we can rewrite the energy balance equation as

$$\begin{aligned} \rho \dot{e} = & \sigma' : \dot{\epsilon} + (1 - \phi^s) \dot{S}^c p_{cr} + (1 - \phi^s) \dot{S}^g p_{ca} \\ & + \sum_{\alpha=l,g,c} (\nabla^x \phi^\alpha \cdot \tilde{v}_\alpha) p_\alpha + \sum_{\alpha=l,g,c} m_\alpha \left[\frac{p_\alpha}{\rho_\alpha} - \frac{1}{2} \tilde{v}_\alpha \cdot \tilde{v}_\alpha \right]. \end{aligned} \quad (6.43)$$

From the above equation we identify five groups of energy-conjugate pairs, which means that five types of constitutive relations are necessary to ensure non-negative entropy production. For each group, we can introduce a constitutive relation as follows.

The first energy-conjugate pair, from which we have defined the effective stress, is the mechanical power produced due to the deformation of the solid matrix. For this pair, it is natural to introduce a stress–strain relation of the form

$$\dot{\sigma}' = \mathbb{C} : \dot{\epsilon}, \quad (6.44)$$

where $\mathbb{C}$ is a fourth-order tangent stiffness tensor. In this work, we shall assume that the solid behavior is linear elastic. This assumption may not allow us to fully describe the complex stiffness of geomaterials [Choo et al. \[2011\]](#), [Jung et al. \[2013\]](#), [Choo et al. \[2013\]](#), but it is often good enough for modeling brittle fracture on which we focus in this work.

The second pair contains the crystallization pressure and the rate of volume change of the crystals. Obviously this pair implies a constitutive law for the crystallization pressure. Notably, the derived energy-conjugate relationship is consistent with an equation for crystallization pressure put forward by Kelemen and Hirth [Kelemen and Hirth \[2012\]](#), which can be expressed as

$$p_{cr} = - \frac{\Delta \psi}{\Delta V_s}, \quad (6.45)$$

where $\Delta \psi$ is the change of Helmholtz free energy and ΔV_s is the difference in volume between the solid products and the solid reactants. A number of specific expressions have been proposed for the crystallization pressure (e.g., [Scherer \[2004\]](#), [Correns and Steinborn \[1939\]](#), [Correns \[1949\]](#), [Steiger \[2005a,b\]](#), [Flatt and Scherer \[2008\]](#), [Kelemen and Hirth](#)

[2012], Desarnaud et al. [2016]). Here we use the classical Correns' equation Correns and Steinborn [1939], Correns [1949], given by

$$p_{cr} = \frac{RT}{V_m} \ln \left(\frac{c}{c_{eq}} \right), \quad (6.46)$$

where R is the gas constant, T is the temperature in Kelvin, V_m is the molar volume of the mineral, and c_{eq} is the equilibrium mass fraction at which the liquid is saturated by the mineral. This equation has later been shown to overlook the role of chemical activities in the crystallization process Steiger [2005a], Flatt et al. [2007], but it gives reasonable predictions compared with experimental data of salt crystallization.

The third pair contains the gas saturation ratio and the capillary pressure. Given that the gas saturation is determined by the liquid saturation, these variables may be related by a water retention law for unsaturated geomaterials. A common choice is the van Genuchten equation van Genuchten [1980], given by

$$S^l(p_{ca}) = S_1^l + (S_2^l - S_1^l) [1 + (p_{ca}/\alpha_{ca})^n]^{-m}. \quad (6.47)$$

This equation requires four material parameters: the residual liquid saturation $S_1^l \geq 0$, the maximum liquid saturation $S_2^l \leq 1$, the scaling capillary pressure α_{ca} , and the exponent n . Another exponent m is related to n via $m = 1 - 1/n$. This equation is originally developed for geomaterials infiltrated by freshwater, so it may be unable to accommodate some important aspects emerging from crystal growth in pores. However, the effects of crystallization on the retention behavior have only become a subject of research recently (e.g., Espinosa-Marzal and Scherer [2013]), and they are still far from being encapsulated into a water retention equation. For this reason, the original van Genuchten equation is used herein, but it can be modified easily when the effects of in-pore crystals become quantified. On a related note, Derluyn et al. Derluyn et al. [2014] also used the van Genuchten model and their simulation results showed good agreements with experimental data even when salt minerals were crystallized. Also noted is that a very recent phase-field model employing the van Genuchten equation has successfully reproduced qualitative patterns of drying-induced cracks, see Cajuhi et al. Cajuhi et al. [2017].

The variables contained in the fourth group of energy-conjugate pairs are the relative velocity, pressure, and volume fractions of every phase inside the pores. This means that we need constitutive laws for flows of pore-filling phases relative to the solid matrix. For the liquid solution, we use the multiphase extension of Darcy's law, which can be written as

$$\mathbf{q}^l = -\frac{k_r}{\mu_l} \mathbf{k} (\nabla^x p_l - \rho_l \mathbf{g}), \quad (6.48)$$

where $\mathbf{q}^l = \phi^l \tilde{\mathbf{v}}_l$ is the seepage velocity, μ_l is the dynamic viscosity of the liquid solution, k_r is the relative permeability, and $\mathbf{k}$ is the second-order absolute permeability tensor. We assume that the permeability tensor of the intact solid matrix is isotropic, i.e., $\mathbf{k} = k\mathbf{1}$, and will discuss its anisotropic evolution by fracturing in the next section. Usually k is assumed to be constant when the solid deformation is infinitesimal. In our problem, however, crystals can clog much of the pore space. To accommodate this clogging effect, here we consider k a function of pore volume, adopting the Kozeny–Carman equation. The equation can be written in a normalized form

$$k = k_0 \left(\frac{(1 - \phi_0)^2}{\phi_0^3} \right) \left(\frac{\phi^3}{(1 - \phi)^2} \right), \quad (6.49)$$

where $\phi = 1 - \phi^s$ is the porosity, and k_0 and ϕ_0 are the reference values of the absolute permeability and the porosity, respectively. The relative permeability is regarded as a function of saturation. When the water retention behavior is modeled by the van Genuchten equation, the relative permeability can be expressed as

$$k_r = \theta^{1/2} \left[1 - (1 - \theta^{1/m})^m \right]^2, \quad \theta = \frac{S^l - S_1^l}{S_2^l - S_1^l}. \quad (6.50)$$

The relative flow of the dissolved mineral is comprised of convective and diffusive/dispersive parts, as in Eq. (7.39). The convective part is described by Darcy's law for the liquid solution presented above. For the diffusive/dispersive part, we introduce a linear diffusion equation

$$\mathbf{j} = -\rho_l D \nabla^x c, \quad (6.51)$$

where D is the diffusion coefficient for the dissolved minerals. For this coefficient we adopt an equation used in Derluyn et al. [Derluyn et al. \[2014\]](#), which takes the form of $D = (D_m/\tau)(1 - \phi^s)(S^l)^{1.6}$ with D_τ being the molecular diffusivity of the mineral and τ being the tortuosity of the porous media. In what follows, we shall assume that the gas pressure is atmospheric (i.e., $p_g \approx 0$) and that the relative velocity of the crystal is negligibly small (i.e., $\tilde{v}_c \approx 0$). These assumptions, which have been introduced to other poromechanical models as well (e.g., [Na and Sun \[2017b\]](#), [Choo \[2016\]](#)), allow us to neglect constitutive laws for the relative flows of the gas and crystal phases.

Lastly, the fifth group of energy-conjugate pairs contains the rate of mass exchange, intrinsic density, pressure, and relative velocity of each phase inside the pores. Because $m_c = -m_l$ and $m_g = 0$, we only need to consider a constitutive law for m_c . It is noted that the velocity has already been related to the pressure, and the pressure related to the saturation. This means that a constitutive law that relates the mass exchange, density, and saturation terms can satisfy this energy-conjugacy. In fact, by definition, such a constitutive law corresponds to a kinetic equation for crystal growth and dissolution. Here we adopt the kinetic equation proposed by Espinosa-Marzal et al. [Espinosa-Marzal et al. \[2007\]](#), which has later become the common choice of computational salt crystallization models (e.g., [Derluyn et al. \[2014\]](#), [Koniorczyk and Gawin \[2012\]](#), [Castellazzi et al. \[2013\]](#)). This equation can be expressed as

$$m_c = \begin{cases} (1 - \phi^s) S^l K_c (U - U_{\text{thr}})^{g_c} & \text{if } U \geq U_{\text{thr}}, \\ -(1 - \phi^s) S^l K_c (1.0 - U)^{g_c} & \text{if } U < 1 \text{ and } S^c > 0, \end{cases} \quad (6.52)$$

where $K_c > 0$ and $g_c > 0$ are kinetic parameters, $U \geq 0$ is the supersaturation ratio, and $U_{\text{thr}} \geq 1$ is the threshold value of U for crystal growth which depends on the type of mineral. Usually $U_{\text{thr}} \geq 1$ for primary crystallization but $U_{\text{thr}} = 1$ once the crystallization process has begun. Multiple definitions are possible for the supersaturation ratio U , and here we define $U = c/c_0$ to be consistent with Eq. (6.46). Note that the first equation in Eq. (6.52) represents a crystal growth process ($m_c \geq 0$ if $U \geq U_{\text{thr}}$), whereas the second equation represents a crystal dissolution process ($m_c < 0$ if $U < 1$ and $S^c > 0$). Note, however, that complex changes in the material's internal structure by dissolution (e.g., the formation of a sensitive clay by leaching) are beyond the modeling capacities of this formulation employing linear elasticity.

So far, we have developed a general modeling framework for coupled chemo-hydro-mechanical processes in fluid-infiltrated porous materials containing dissolved and crystallized minerals. In doing so, we have constructed a particular class of the framework by

selecting a set of constitutive laws. It is again noted that our selection is just a specific choice, and other sets of constitutive laws would work equally well. Similarly, while we adopt a phase-field approach to fracture in the following, other approaches for similar purposes are also compatible with our development herein.

6.4 Phase-field formulation for fracture

In this section, we present a phase-field model of fracture driven by the effective stress derived in the previous section. To be consistent with the foregoing continuum mechanics approach, here we derive the phase-field model as a balance law of microforces. Microforce balance derivations of phase-field models of fractures have been presented in several previous studies [Wilson et al. \[2013\]](#), [Borden et al. \[2016b\]](#), [Wilson and Landis \[2016\]](#), [De Lorenzis et al. \[2016\]](#), [Choo and Sun \[2018a\]](#). Among them, we adopt the derivation procedure of Choo and Sun [Choo and Sun \[2018a\]](#), which differs from other microforce derivations because it views crack growth as a thermodynamically irreversible process. The motivation and implication of this derivation are explained in detail in Choo and Sun [Choo and Sun \[2018a\]](#).

Without loss of generality, in this section we consider a “dry” porous solid in which the liquid, gas, and crystal phases are absent. This simplification is just to prevent proliferation of numerous terms unrelated to the fracturing process. Again, an important premise in continuum poromechanics is that all mechanical processes—including the fracturing process which we will describe as the evolution of the phase-field variable—are driven by the effective stress. This means that terms other than the effective stress and its related ones will not affect the formulation that follow. Thus, for brevity, the terms unrelated to the deformation and fracture of the solid matrix are omitted in this section.

6.4.1 Phase-field approximation of fracture surfaces

We first introduce a phase-field approximation of fracture geometry, which, in essence, is an approximation of a sharp discontinuity as a diffuse interface. Let Γ denote a set of discontinuous fractures inside the body Ω . The total area of the fracture surfaces is given by

$$A_\Gamma = \int_\Gamma dA, \quad (6.53)$$

which is an area integral over Γ . Calculating this integral during the evolution of fracture is an infeasible task because tracing the change of Γ is extremely difficult in most cases. To circumvent this difficulty, we seek to transform an area integral over the evolving domain Γ into a volume integral over the fixed domain Ω . For this purpose we define a phase-field variable $d \in [0, 1]$, which denotes an intact state by $d = 0$ and a fully cracked state by $d = 1$. Naturally d can be understood as a damage variable. Using this phase-field variable, we introduce a crack density functional $\Gamma_d(d, \nabla^x d)$ such that

$$A_\Gamma \approx A_{\Gamma_d} = \int_\Omega \Gamma_d(d, \nabla^x d) dV, \quad (6.54)$$

where the last integral is defined over the volume Ω . In this work, we adopt a widely used crack density functional of the form

$$\Gamma_d(d, \nabla^x d) = \frac{d^2}{2l} + \frac{l}{2} |\nabla^x d|^2, \quad (6.55)$$

where $l > 0$ is the length parameter for the phase-field regularization. The smaller the length parameter l , the closer the phase-field approximation to the original sharp discontinuity.

6.4.2 Balance law derivation of phase-field evolution

To derive a governing equation for the phase-field variable, we make use of the microforce approach developed by Gurtin [Gurtin \[1996b\]](#), which has proven useful to derive phase-field models [Wilson et al. \[2013\]](#), [Borden et al. \[2016b\]](#), [Wilson and Landis \[2016\]](#), [De Lorenzis et al. \[2016\]](#), [Choo and Sun \[2018a\]](#) and other types of models [Gurtin \[2000\]](#), [Henann and Kamrin \[2014\]](#) within the framework of continuum mechanics. The first step of this approach is to postulate the existence of a microforce system in which the phase-field variable d (referred to as the order parameter in [Gurtin \[1996b\]](#)) is energy-conjugate to an internal microforce π and a surface microforce ζ . Consider an arbitrary volume $\mathcal{V}$ with boundary $\partial\mathcal{V}$ in this microforce system. The balance of microforce over the volume $\mathcal{V}$ is given by

$$\int_{\partial\mathcal{V}} \zeta \, dA + \int_{\mathcal{V}} \pi \, dV = 0. \quad (6.56)$$

Denoting the unit normal vector of $\partial\mathcal{V}$ by $\mathbf{n}$, we also introduce a microforce traction vector $\boldsymbol{\xi}$ such that $\zeta = \boldsymbol{\xi} \cdot \mathbf{n}$. Then, by applying the divergence theorem and noting the arbitrariness of $\mathcal{V}$, we obtain a localized form of Eq. (6.56) as

$$\nabla^x \cdot \boldsymbol{\xi} + \pi = 0. \quad (6.57)$$

The internal and surface microforces, which are energy-conjugate to the phase-field variable d , should evolve such that this balance law is satisfied. Therefore this microforce balance law serves as a governing equation for the evolution of phase field—equivalently, the damage and fracturing process. In the following, we derive specific forms of π and $\boldsymbol{\xi}$ based on thermodynamic arguments.

The mechanical power in the microforce system is given by

$$\tilde{P}^m = \int_{\partial\mathcal{V}} (\boldsymbol{\xi} \cdot \mathbf{n}) \cdot \dot{d} \, dA = \int_{\mathcal{V}} (\boldsymbol{\xi} \cdot \nabla^x \dot{d} - \pi \dot{d}) \, dV. \quad (6.58)$$

Incorporating this additional mechanical power as well as keeping the effective stress power only, we can rewrite the balance of energy as

$$\rho \dot{e} = \boldsymbol{\sigma}' : \dot{\boldsymbol{\varepsilon}} + \boldsymbol{\xi} \cdot \nabla^x \dot{d} - \pi \dot{d}. \quad (6.59)$$

We will exploit the second law of thermodynamics to derive expressions for the effective stress tensor and the microforce variables. To this end, we first consider a stored energy density function of the form

$$\psi(\boldsymbol{\varepsilon}, d) = g(d)W(\boldsymbol{\varepsilon}). \quad (6.60)$$

Here, $W(\boldsymbol{\varepsilon})$ denotes the strain energy stored in the undamaged material, and $g(d) \in [0, 1]$ is a so-called degradation function which should satisfy $g(0) = 1$ and $g(1) = 0$. For now we consider general forms of $W(\boldsymbol{\varepsilon})$ and $g(d)$, and will discuss their specific forms later in this section.

At this point, it is noted that the energy used to create a fracture surface does not enter the stored energy function because crack growth is considered fully dissipative

a priori herein. This is consistent with the stored energy functions used in variational frameworks for fracture (e.g., [Miehe et al. \[2010c\]](#), [Zhang et al. \[2016\]](#)), but different from those assumed in other balance law derivations of phase-field models based on microforce arguments [Wilson et al. \[2013\]](#), [Borden et al. \[2016b\]](#), [Wilson and Landis \[2016\]](#), [De Lorenzis et al. \[2016\]](#). More specifically, the stored energy functions in previous balance law derivations contain the fracture energy, so their derivations lead to a thermodynamic implication that crack growth is a reversible process in a rate-independent setting. This implication would be appropriate for clean fracture surfaces that can heal under highly controlled conditions. However, here we prefer to view crack growth as an irreversible process, since crack healing phenomena in geomaterials are usually inconsistent with the thermodynamic definition of a reversible process. See Choo and Sun [Choo and Sun \[2018a\]](#) for a more detailed discussion on this aspect.

Having defined the stored energy density, we can write the dissipation inequality as

$$\mathcal{D} = \sigma' : \dot{\varepsilon} + \xi \cdot \nabla^x \dot{d} - \pi \dot{d} - \dot{\psi} \geq 0. \quad (6.61)$$

The time derivative of the stored energy function is given by

$$\dot{\psi}(\varepsilon, d) = \frac{\partial \psi}{\partial \varepsilon} : \dot{\varepsilon} + \frac{\partial \psi}{\partial d} \dot{d}. \quad (6.62)$$

Substituting Eq. (6.62) into Eq. (6.61), we get an alternative expression for the dissipation inequality as

$$\mathcal{D} = \left(\sigma' - \frac{\partial \psi}{\partial \varepsilon} \right) : \dot{\varepsilon} - \left(\pi - \frac{\partial \psi}{\partial d} \right) \dot{d} + \xi \cdot \nabla^x \dot{d} \geq 0. \quad (6.63)$$

To ensure non-negative dissipation of the stress power irrespective of $\dot{\varepsilon}$, the effective stress should be related to the stored energy. This standard argument leads to a hyperelastic relation of the form

$$\sigma' = \frac{\partial \psi}{\partial \varepsilon}. \quad (6.64)$$

Next, as done in [Gurtin \[2000\]](#), [Henann and Kamrin \[2014\]](#) for deriving other types of models, we assume that the internal microforce is additively decomposed into two parts, the energetic (non-dissipative) part π^{en} and the dissipative part π^{dis} . In other words, $\pi = \pi^{\text{en}} + \pi^{\text{dis}}$. The same argument that we used to get Eq. (6.64) yields the following expression for the energetic part:

$$\pi^{\text{en}} = - \frac{\partial \psi}{\partial d}. \quad (6.65)$$

Inserting Eqs. (6.64) and (6.65) into Eq. (6.63) gives the reduced dissipation inequality of the form

$$\mathcal{D}^{\text{f}} = \xi \cdot \nabla^x \dot{d} - \pi^{\text{dis}} \dot{d} \geq 0. \quad (6.66)$$

Because the material is considered elastic, the dissipation is solely attributed to the evolution of the phase-field variable d , or crack growth.

To arrive at specific expressions for ξ and π^{dis} , we now postulate that cracks are created in a way that they maximize the energy dissipation. This postulate is consistent with the Griffith theory of brittle fracture [Griffith \[1921\]](#), and has been central to several

variational frameworks for phase-field fracture (e.g., Miehe et al. [2010c], Zhang et al. [2016]). Our task is then to find expressions for the microforce variables that maximize $\mathcal{D}^f$ defined in Eq. (6.66). Equivalently, we seek to minimize the negative of the reduced dissipation functional, given by

$$-\mathcal{D}^f = -\boldsymbol{\xi} \cdot \nabla^x \dot{d} + \pi^{\text{dis}} \dot{d} \leq 0. \quad (6.67)$$

The arguments of this functional, $\dot{d}$ and $\nabla^x \dot{d}$, are indeed subject to a constraint in that these variables should form a phase-field approximation of fracture. The specific expression for this constraint can be obtained by taking time derivative of the crack density functional, Eq. (6.55). This procedure gives

$$\dot{\Gamma}_d(d, \nabla^x d) = \left(\frac{d}{l} \right) \dot{d} + l \nabla^x d (\nabla^x \dot{d}). \quad (6.68)$$

In the cases we consider, $\dot{\Gamma}_d(d, \nabla^x d) \geq 0$ (crack irreversibility) and $\dot{\Gamma}_d(d, \nabla^x d) < \infty$ (finite speed of crack propagation). Given this constraint, we can write a Lagrangian for this constrained minimization problem as

$$\mathcal{L}(\dot{d}, \nabla^x \dot{d}, \lambda) = -\boldsymbol{\xi} \cdot \nabla^x \dot{d} + \pi^{\text{dis}} \dot{d} + \lambda \left[\left(\frac{d}{l} \right) \dot{d} + l \nabla^x d (\nabla^x \dot{d}) - \dot{\Gamma}_d \right], \quad (6.69)$$

where λ is the Lagrange multiplier of this problem. Invoking the stationary condition of this Lagrangian gives expressions for $\boldsymbol{\xi}$ and π^{dis} as follows:

$$\delta_{\nabla^x \dot{d}} \mathcal{L} = -\boldsymbol{\xi} + \lambda l \nabla^x d = 0 \quad \rightarrow \quad \boldsymbol{\xi} = \lambda l \nabla^x d, \quad (6.70)$$

$$\delta_{\dot{d}} \mathcal{L} = \pi^{\text{dis}} + \lambda \left(\frac{d}{l} \right) = 0 \quad \rightarrow \quad \pi^{\text{dis}} = -\lambda \left(\frac{d}{l} \right). \quad (6.71)$$

The remaining task is to identify the meaning of the Lagrangian multiplier, λ , in the context of our problem. To this end, we substitute the results of Eqs. (6.70) and (6.71) into $\mathcal{D}^f$ in Eq. (6.66), which is the energy dissipation per unit volume. Integrating the resulting dissipation density over the domain Ω with the crack surface Γ_d gives

$$\int_{\Omega} \mathcal{D}^f dV = \int_{\Omega} \lambda \left[\left(\frac{d}{l} \right) \dot{d} + l \nabla^x d (\nabla^x \dot{d}) \right] dV = \int_{\Omega} \lambda \dot{\Gamma}_d dV \approx \frac{d}{dt} \int_{\Gamma_d} \lambda dA \geq 0, \quad (6.72)$$

where the last approximation is attributed to the phase-field regularization of discontinuous surfaces. Equation (6.72) shows that the Lagrange multiplier λ can be interpreted as the energy dissipated by the creation of unit crack surface area. By definition, this energy corresponds to the critical fracture energy in fracture mechanics.

Let $\mathcal{G}_c$ denote the critical fracture energy. Now we can express the internal microforce as

$$\pi = \pi^{\text{en}} + \pi^{\text{dis}}, \quad \pi^{\text{en}} = -\frac{\partial \psi}{\partial d} = -g'(d)W(\varepsilon), \quad \pi^{\text{dis}} = -\mathcal{G}_c \left(\frac{d}{l} \right), \quad (6.73)$$

and the microforce traction vector as

$$\boldsymbol{\xi} = \mathcal{G}_c l \nabla^x d. \quad (6.74)$$

Substituting these expressions into Eq. (6.57), we can rewrite the governing equation for the phase-field variable as

$$-g'(d)W(\boldsymbol{\varepsilon}) - \mathcal{G}_c \left(\frac{d}{l} - l \nabla^x \cdot (\nabla^x d) \right) = 0. \quad (6.75)$$

Notably, this equation is the same as the governing equation for a phase-field model of brittle, rate-independent fracture obtained by variational and other balance law approaches, see [Miehe et al. \[2010c,d\]](#), [Wilson et al. \[2013\]](#), [Wilson and Landis \[2016\]](#) for example.

6.4.3 Stored energy function

Now we consider specific expressions for the stored energy function, $\psi(\boldsymbol{\varepsilon}, d) = g(d)W(\boldsymbol{\varepsilon})$. The stored energy function is central to the phase-field model of fracture since it determines the effective stress tensor $\boldsymbol{\sigma}'$, as in Eq. (6.64), and the energetic force driving the evolution of the phase-field variable d , as in Eq. (6.73).

First, as for the degradation function $g(d)$, we adopt the form most widely used by the phase-field modeling community, given by

$$g(d) = (1 - d)^2. \quad (6.76)$$

It is noted that other forms of degradation functions have also been suggested, e.g., the cubic degradation proposed by [Borden et al. \[2016b\]](#).

Next, we need to determine a suitable form of $W(\boldsymbol{\varepsilon})$, the strain energy function decoupled from the phase-field variable. In doing so, we should take into account that for physically realistic results, the strain energy associated with pure compression should not give rise to fracturing. For this reason, previous studies have proposed to decompose the stored energy function as $W(\boldsymbol{\varepsilon}) = W_+(\boldsymbol{\varepsilon}) + W_-(\boldsymbol{\varepsilon})$, where $W_+(\boldsymbol{\varepsilon})$ is the fracturing part related to the tensile strain energy, and $W_-(\boldsymbol{\varepsilon})$ is the non-fracturing part related to the compressive strain energy. This decomposition has been mainly done via either of the following two schemes: one that uses the sign of principal strains proposed by [Miehe et al. \[2010c\]](#), and another that uses a volumetric–deviatoric split of the strain energy function proposed by [Amor et al. \[2009b\]](#). Here we use the former one, and decompose the stored energy function of a linear elastic material as

$$W_+(\boldsymbol{\varepsilon}) = \frac{1}{2} \bar{\lambda} \langle \text{tr}(\boldsymbol{\varepsilon}) \rangle_+^2 + \bar{\mu} \sum_{a=1}^3 \langle \varepsilon_a \rangle_+^2, \quad W_-(\boldsymbol{\varepsilon}) = \frac{1}{2} \bar{\lambda} \langle \text{tr}(\boldsymbol{\varepsilon}) \rangle_-^2 + \bar{\mu} \sum_{a=1}^3 \langle \varepsilon_a \rangle_-^2, \quad (6.77)$$

where $\langle \cdot \rangle_{\pm} = (\cdot \pm |\cdot|)/2$, $\bar{\lambda}$ and $\bar{\mu}$ are the Lamé parameters, and ε_a are the principal strains.

After decomposing the strain energy as above, we apply the degradation function to the fracturing part only, i.e.,

$$\psi(\boldsymbol{\varepsilon}, d) = g(d)W_+(\boldsymbol{\varepsilon}) + W_-(\boldsymbol{\varepsilon}). \quad (6.78)$$

Then the effective stress is expressed as

$$\boldsymbol{\sigma}' = \frac{\partial \psi(\boldsymbol{\varepsilon}, d)}{\partial \boldsymbol{\varepsilon}} = g(d) \frac{\partial W_+(\boldsymbol{\varepsilon})}{\partial \boldsymbol{\varepsilon}} + \frac{\partial W_-(\boldsymbol{\varepsilon})}{\partial \boldsymbol{\varepsilon}}. \quad (6.79)$$

Likewise, the energetic microforce is expressed as

$$\pi_{\text{en}} = -\frac{\partial \psi(\boldsymbol{\varepsilon}, d)}{\partial d} = -g'(d)W_+(\boldsymbol{\varepsilon}). \quad (6.80)$$

Note that π_{en} does not take the non-fracturing part of the strain energy, $W_-(\boldsymbol{\varepsilon})$.

6.4.4 Thermodynamic restriction and crack irreversibility

Our derivation leads to an expression for the dissipation inequality which needs to be satisfied for thermodynamic consistency. Rewriting Eq. (6.66), we can express the rate of energy dissipation in a unit volume as

$$\mathcal{D}^f = \mathcal{G}_c \dot{\Gamma}_d \geq 0. \quad (6.81)$$

Because $\mathcal{G}_c > 0$ by definition, this is equivalent to

$$\dot{\Gamma}_d \geq 0, \quad (6.82)$$

which means that crack growth should be irreversible, as we postulated in the beginning of this derivation. Integrating this equation over the entire domain leads to an expression that is identical to the crack irreversibility condition presented in variational frameworks for phase-field fracture (e.g., Eq. (20) of Miehe et al. [Miehe et al. \[2010c\]](#)). Thus it can be concluded that our balance law derivation is consistent with the variational derivation with respect to the thermodynamic implication as well as the governing equation. Eq. (6.82) can also be written as

$$\left(\frac{d}{l}\right) \dot{d} + l \|\nabla^x d\| |\nabla^x d| \geq 0, \quad (6.83)$$

where we use an alternative form of the time derivative of the nonlocal term in the crack density functional. The above equation shows that the crack irreversibility condition boils down to $\dot{d} \geq 0$.

To enforce this crack irreversibility condition, $\dot{d} \geq 0$, we adopt the approach proposed by Miehe et al. [Miehe et al. \[2010d\]](#). The approach is to make the energetic force driving the phase-field evolution, which is denoted by π^{en} in our derivation, a non-decreasing function even as $W_+(\varepsilon)$ is decreasing. Following this idea, we introduce a strain energy history functional $\mathcal{H} \geq 0$ subject to the Karush–Kuhn–Tucker condition of

$$W_+ - \mathcal{H} \leq 0, \quad \dot{\mathcal{H}} \geq 0, \quad \dot{\mathcal{H}}(W_+ - \mathcal{H}) = 0. \quad (6.84)$$

Simply speaking, $\mathcal{H}$ is the maximum of the fracturing part of the strain energy during the course of loading. Replacing the stored energy term in Eq. (6.75) with $\mathcal{H}$ gives a modified phase-field equation of the form

$$-g'(d)\mathcal{H} - \mathcal{G}_c \left(\frac{d}{l} - l \nabla^x \cdot (\nabla^x d)\right) = 0. \quad (6.85)$$

Eq. (6.85) will be used as the governing equation for the phase-field model in the sequel.

6.4.5 Permeability evolution by phase-field fracture

Phase-field modeling of fracture in fluid-infiltrated porous media should take into account the impact of the fracturing process on fluid flow. Several types of approaches have been proposed for this purpose but with different emphases (e.g., [Mikelić et al. \[2015\]](#), [Miehe and Mauthe \[2015\]](#), [Lee et al. \[2016a,c, 2017\]](#), [Mauthe and Miehe \[2017\]](#), [Santillán et al. \[2017\]](#)). Most of them are concerned with hydraulic fracturing whereby the rate of mass transfer between the fracture and matrix systems (leak-off) is often far lower than the fluid injection rate. The problem at hand, however, involves a quite different situation

in that here crystal growth in matrix pores slowly drives fractures and significant fluid flow between the fracture and matrix systems is expected. In addition, we face a new problem that growing crystals increasingly clog the fracture aperture. Thus, an approach that views the fracture and matrix pores as a whole—rather than distinguished domains described by separate governing equations—would be more appropriate for our purpose.

Given these considerations, here we take an approach that augments an anisotropic permeability tensor describing Poiseuille flow to the absolute permeability tensor in Eq. (6.48). Such an approach has been advocated by Miehe and Mauthe [Miehe and Mauthe \[2015\]](#), [Mauthe and Miehe \[2017\]](#) and Wang and Sun [Wang and Sun \[2017b\]](#), among others. We now write the absolute permeability tensor as

$$\mathbf{k} = \mathbf{k}_{\text{matrix}} + d^2 \mathbf{k}_{\text{frac}}. \quad (6.86)$$

Here, $\mathbf{k}_{\text{matrix}}$ is the isotropic permeability tensor of the solid matrix which we have discussed in the previous section. On the other hand, $\mathbf{k}_{\text{frac}}$ is anisotropic, and given by

$$\mathbf{k}_{\text{frac}} = \frac{w^2}{12} (\mathbf{1} - \mathbf{n} \otimes \mathbf{n}), \quad (6.87)$$

where $\mathbf{n} = \nabla^x d / |\nabla^x d|$ is the unit normal vector perpendicular to the fracture direction, and w is the hydraulic aperture. Note that the hydraulic aperture herein should be a function of the crystal fraction as well as the fracture width. For simplicity, we assume that the aperture can be given by $(1 - S^c) \bar{w}$, where $\bar{w}$ is the hydraulic aperture in the absence of crystals. For $\bar{w}$, we adopt an equation proposed by Miehe and Mauthe [Miehe and Mauthe \[2015\]](#), specializing it to infinitesimal deformation conditions. The resulting form of the hydraulic aperture w is given by

$$w = (1 - S_c) l_{\perp} (\mathbf{n} \cdot \boldsymbol{\varepsilon} \cdot \mathbf{n}) \quad (6.88)$$

where $l_{\perp}$ is the characteristic length of a line element perpendicular to the fracture. For simplicity, we assign $l_{\perp}$ to be the mesh size h as in Miehe and Mauthe [Miehe and Mauthe \[2015\]](#).

6.4.6 Estimation of phase-field modeling parameters for geomaterials

Lastly, we describe how the phase-field modeling parameters are estimated in this work. A standard phase-field model of fracture requires two parameters: (1) the critical fracture energy $\mathcal{G}_c$, and (2) the length parameter l . While the two parameters have different origins—the former is from fracture mechanics theory and the latter is from a phase-field approximation of fracture—they together determine the fracturing response of a material [Amor et al. \[2009b\]](#), [Borden et al. \[2012, 2016b\]](#), [Zhang et al. \[2017\]](#). In particular, the peak stress under tension, which is usually reckoned as the tensile strength, is controlled by the fracture energy, length parameter, and elasticity moduli. It is thus of significant importance to assign a suitable combination of the phase-field modeling parameters. Among them, however, the length parameter is uneasy to estimate because it is not a physically measurable quantity.

Here we devise a way to estimate the length parameter for geomaterials, drawing on an empirical relationship between the tensile strength and the fracture toughness. Let σ_t denote the tensile strength of a geomaterial and K_{Ic} denote its mode I fracture

toughness. Experimental studies have found that, for many geomaterials under quasi-static conditions, the two properties are well correlated as a linear function [Harison et al. \[1994\]](#), [Zhang \[2002\]](#), [Wang et al. \[2007\]](#), given by

$$K_{Ic} = \beta \sigma_t, \quad (6.89)$$

where β is the linear regression coefficient having a unit of square root meter. Example values of β are 0.1453 for dozens of soft to hard rocks [Zhang \[2002\]](#) and 0.3546 for compacted clays tested by Wang et al. [Wang et al. \[2007\]](#). This relationship usually shows a coefficient of determination, r^2 , around 0.9. At the same time, the tensile strength of a phase-field model is determined by the two parameters, $\mathcal{G}_c$ and l , and Young's modulus, E . When the degradation function is $g(d) = (1 - d)^2$, the tensile strength is given by [Amor et al. \[2009b\]](#), [Borden et al. \[2012\]](#)

$$\sigma_t = \frac{9}{16} \sqrt{\frac{E \mathcal{G}_c}{3l}}. \quad (6.90)$$

Also, $\mathcal{G}_c$ and K_{Ic} in linear elastic materials are related as

$$\mathcal{G}_c = \frac{K_{Ic}^2}{E'}, \quad (6.91)$$

where $E' = E$ for plane stress and $E' = E / (1 - \nu^2)$ for plane strain. Substituting Eqs. (6.90) and (6.91) into Eq. (6.89) and solving for l yields

$$l = \frac{27}{256} \left(\frac{E}{E'} \right) \beta^2. \quad (6.92)$$

Here, the unit of l is meter because β is given by a square root meter. This equation allows us to estimate l from experimental data of σ_t and K_{Ic} . Once we have estimated l in this way and known the tensile strength and elasticity parameters, we can find the corresponding value of $\mathcal{G}_c$ from Eq. (6.90).

Interestingly, Eq. (6.92) suggests that the length parameter is related to the relationship between the tensile strength and the mode I fracture toughness, rather than the values of these properties. Given that the linear relationship (6.89) has been found for many types of geomaterials (though not necessary valid for all types of geomaterials, see [Lakshmikantha et al. \[2008\]](#)), Eq. (6.92) may serve as a useful guide to set a length parameter for phase-field modeling of fracture in geomechanical problems.

6.5 Discrete formulation

This section develops a discrete formulation for numerical solution of the chemo-hydro-mechanics and phase-field model described so far. We begin by stating the governing equations and the relevant initial-boundary value problem. For robust and accurate spatial discretization of the coupled problem, we use the finite element method for the deformation and fracture problem (momentum balance and phase-field equations), and the finite volume method for the flow and transport problem (mass balance equations). The resulting discrete system is solved by a staggered scheme that sequentially updates the chemo-hydro-mechanics problem and the phase-field problem. To facilitate monolithic solution of the coupled chemo-hydro-mechanics problem, we design a block-partitioned preconditioner which significantly speeds up iterative linear solvers.

6.5.1 Governing equations

The coupled problem at hand is furnished by five governing equations that follow. The first one is the balance equation for linear momentum in the mixture. For simplicity we shall neglect the effects of inertial forces and crystallization kinetics on the linear momentum. Substituting the expression for effective stress in Eq. (6.38) into Eq. (6.21), we express the balance of linear momentum

$$\nabla^x \cdot (\sigma' - B\bar{p}\mathbf{1}) + \rho g = 0. \quad (6.93)$$

The second governing equation is the mass balance for the liquid solution, which was originally given by Eq. (6.10). Since $\rho^l = (1 - \phi^s)S^l\rho_l$, we can expand this equation as

$$(1 - \phi^s)(\rho_l \dot{S}^l + S^l \dot{\rho}_l) - S^l \rho_l (\dot{\phi}^s + \phi^s \nabla^x \cdot \mathbf{v}) + S^l \rho_l \nabla^x \cdot \mathbf{v} + \nabla^x \cdot \mathbf{w}_l = m_l. \quad (6.94)$$

Here, the second product can be simplified using the Biot coefficient B . To wit, we can rewrite Eq. (6.33) as

$$\dot{\phi}^s + \phi^s \nabla^x \cdot \mathbf{v} = (1 - B) \nabla^x \cdot \mathbf{v}, \quad (6.95)$$

and insert this equation into Eq. (6.94). Then the liquid mass balance equation takes the form

$$(1 - \phi^s)(\rho_l \dot{S}^l + S^l \dot{\rho}_l) + S^l \rho_l B \nabla^x \cdot \mathbf{v} + \nabla^x \cdot \mathbf{w}_l = m_l. \quad (6.96)$$

In case $\dot{\rho}_l$ and $m_l = 0$, the above equation degenerates to the mass balance equation of unsaturated poromechanics for shallow water problems [Borja \[2006b\]](#), [Borja and White \[2010b\]](#).

Third, we consider the mass balance equation for the dissolved minerals. Because $\rho^d = c\rho^l$, the mass balance equation can be rewritten as

$$\rho^l \dot{c} + c(\dot{\rho}^l + \rho^l \nabla^x \cdot \mathbf{v}) + \nabla^x \cdot (c\mathbf{w}_l) + \nabla^x \cdot \mathbf{j} = m_d. \quad (6.97)$$

In the above we have shown that

$$\dot{\rho}^l + \rho^l \nabla^x \cdot \mathbf{v} = (1 - \phi^s)(\rho_l \dot{S}^l + S^l \dot{\rho}_l) + S^l \rho_l B \nabla^x \cdot \mathbf{v}. \quad (6.98)$$

Substituting the above equation and $m_d = m_l$ into Eq. (6.97) gives

$$\rho^l \dot{c} + c(1 - \phi^s)(\rho_l \dot{S}^l + S^l \dot{\rho}_l) + cS^l \rho_l B \nabla^x \cdot \mathbf{v} + \nabla^x \cdot (c\mathbf{w}_l) + \nabla^x \cdot \mathbf{j} = m_l, \quad (6.99)$$

which is a transient advection–diffusion equation for the mass fraction, c .

The fourth governing equation is the mass balance for the crystals in pores. Given that $\rho^c = (1 - \phi^s)S^c\rho_c$ and $\mathbf{w}_c$ has been assumed to be zero, we can rewrite Eq. (6.13) as

$$(1 - \phi^s)\rho_c \dot{S}^c - S^c \rho_c (\dot{\phi}^s + \phi^s \nabla^x \cdot \mathbf{v}) + S^c \rho_c \nabla^x \cdot \mathbf{v} = m_c, \quad (6.100)$$

Again, by appealing to Eq. (6.95), we get

$$(1 - \phi^s)\rho_c \dot{S}^c + S^c \rho_c B \nabla^x \cdot \mathbf{v} = m_c. \quad (6.101)$$

Last, specifying $g'(d) = -2(1 - d)$, we rewrite the phase-field governing equation as

$$2(1 - d)\mathcal{H} - \mathcal{G}_c \left(\frac{d}{l} - l \nabla^x \cdot (\nabla^x d) \right) = 0. \quad (6.102)$$

As for the primary variables of these governing equations, we select the following five fields: the displacement vector of the solid matrix $\mathbf{u}$, the pore pressure of the liquid solution $p := p_l$, the mass fraction of the dissolved minerals c , the saturation ratio of the crystals $s := S^c$, and the phase-field variable d .

We complete the statement of the problem by prescribing boundary and initial conditions as follows. Let Ω denote the domain of interest and $\partial\Omega$ denote its boundary. The boundary is suitably decomposed as: displacement and traction boundaries, $\partial\Omega_u$ and $\partial\Omega_t$; pressure and liquid flux boundaries, $\partial\Omega_p$ and $\partial\Omega_q$; and mass fraction and mineral flux boundaries, $\partial\Omega_c$ and $\partial\Omega_j$. These decomposed boundaries satisfy $\partial\Omega = \overline{\partial\Omega_u} \cup \overline{\partial\Omega_t} = \overline{\partial\Omega_p} \cup \overline{\partial\Omega_q} = \overline{\partial\Omega_c} \cup \overline{\partial\Omega_j}$ and $\emptyset = \partial\Omega_u \cap \partial\Omega_t = \partial\Omega_p \cap \partial\Omega_q = \partial\Omega_c \cap \partial\Omega_j$. The boundary conditions are given by

$$\mathbf{u} = \hat{\mathbf{u}} \quad \text{on} \quad \partial\Omega_u \quad (6.103)$$

$$\mathbf{n} \cdot \boldsymbol{\sigma} = \hat{\mathbf{t}} \quad \text{on} \quad \partial\Omega_t \quad (6.104)$$

$$p = \hat{p} \quad \text{on} \quad \partial\Omega_p \quad (6.105)$$

$$-\mathbf{n} \cdot \mathbf{w}_l = \hat{q} \quad \text{on} \quad \partial\Omega_q \quad (6.106)$$

$$c = \hat{c} \quad \text{on} \quad \partial\Omega_c \quad (6.107)$$

$$-\mathbf{n} \cdot (c\mathbf{w}_l + \mathbf{j}) = \hat{j} \quad \text{on} \quad \partial\Omega_j, \quad (6.108)$$

where $\mathbf{n}$ is the unit outward normal vector and the hats denote the prescribed boundary values. The initial conditions are given by

$$\mathbf{u} = \mathbf{u}_0(\mathbf{x}), \quad p = p_0(\mathbf{x}), \quad c = c_0(\mathbf{x}), \quad (6.109)$$

for all position vectors $\mathbf{x} \in \Omega$ at time $t = 0$. For s and d , we consider no flux boundary conditions and zero initial conditions throughout.

6.5.2 Time discretization

We begin the discretization process by approximating the time derivatives in the residuals of mass balance for liquid, dissolved mineral, and crystals. Given that the chemical, hydrological, and mechanical processes in our problem may involve a variety of time scales, we use an unconditionally stable, first-order backward Euler method. Consider a time increment Δt from t_n to t_{n+1} . We discretize the time derivatives of the primary variables as

$$\mathbf{v} = \dot{\mathbf{u}} = \frac{\mathbf{u}_{n+1} - \mathbf{u}_n}{\Delta t}, \quad \dot{p} = \frac{p_{n+1} - p_n}{\Delta t}, \quad \dot{c} = \frac{c_{n+1} - c_n}{\Delta t}, \quad \dot{S}^c = \frac{S_{n+1}^c - S_n^c}{\Delta t}, \quad (6.110)$$

and the time derivatives of liquid saturation and liquid density as $\dot{S}^l = (S_{n+1}^l - S_n^l) / \Delta t$ and $\dot{\rho}_l = (\rho_{l,n+1} - \rho_{l,n}) / \Delta t$. All other variables are evaluated at time $n + 1$. For notational simplicity, hereafter we drop the subscript $n + 1$ for quantities at time t_{n+1} .

6.5.3 Space discretization

For space discretization we use the finite element method for the deformation and fracture problem (momentum balance and phase-field equations) and the finite volume method for the flow and transport problem (mass balance equations). This combination is motivated by mathematical natures of the two problems. The deformation and fracture problem is described by elliptic equations, so its discretization by the finite element method

is an optimal choice. On the other hand, the flow and transport problem involves hyperbolic systems, for which the finite volume method is usually more appropriate than the (continuous) finite element method. This is mainly because finite volume discretization is robust in the presence of sharp gradients in the solution fields as well as locally mass conservative at the element level.

We use a single mesh for both finite element and finite volume discretization, as shown in Fig. 6.4. The degrees of freedom for the deformation and fracture unknowns—the displacement vector and the phase field—are located at the element nodes, whereas those for the flow and transport unknowns—the pressure, mass fraction, and crystal saturation—are located at the element center.

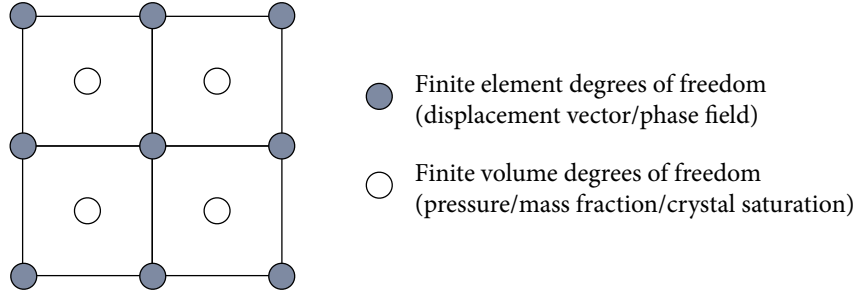


Fig. 6.4. An example mesh illustrating the locations of (linear) finite element and finite volume degrees of freedom.

To begin finite element discretization of the deformation and fracture problem, we define the spaces of the trial solutions for $\mathbf{u}$ and d as

$$\mathcal{S}_u = \{\mathbf{u} : \Omega \rightarrow \mathbb{R}^{\dim} \mid \mathbf{u} \in H^1, \mathbf{u} = \hat{\mathbf{u}} \text{ on } \partial\Omega_u\}, \quad (6.111)$$

$$\mathcal{S}_d = \{d : \Omega \rightarrow \mathbb{R} \mid d \in H^1\}, \quad (6.112)$$

where H^1 is the Sobolev space of order one. Accordingly, the spaces of the weighting functions are defined as

$$\mathcal{V}_u = \{\boldsymbol{\eta} : \Omega \rightarrow \mathbb{R}^{\dim} \mid \boldsymbol{\eta} \in H^1, \boldsymbol{\eta} = \mathbf{0} \text{ on } \partial\Omega_u\}, \quad (6.113)$$

$$\mathcal{V}_d = \{\varphi : \Omega \rightarrow \mathbb{R} \mid \varphi \in H^1\}. \quad (6.114)$$

Through the standard weighted residual procedure, we can readily develop the variational equations of the deformation and fracture problem as

$$R_{\text{mom}} = \int_{\Omega} \nabla^s \boldsymbol{\eta} : \boldsymbol{\sigma}' dV - \int_{\Omega} B\bar{p} \nabla^x \cdot \boldsymbol{\eta} dV - \int_{\Omega} \boldsymbol{\eta} \cdot \rho \mathbf{g} dV - \int_{\partial\Omega_t} \boldsymbol{\eta} \cdot \hat{\mathbf{t}} dA = 0, \quad (6.115)$$

$$R_{\text{frac}} = \int_{\Omega} \varphi [2(1-d)\mathcal{H}] dV - \int_{\Omega} \frac{g_c}{l} [\varphi + l^2(\nabla^x \varphi \cdot \nabla^x d)] dV = 0. \quad (6.116)$$

Here, we have presented the variational equations as residuals to solve these equations via Newton's method later. We then perform finite element discretization of Eqs. (6.115) and (6.116), and obtain the discrete residual vectors by assembling element contributions. The contributions of element e are given by

$$[R_{\text{mom}}]^e = \int_{\Omega^e} \nabla^s \boldsymbol{\eta}^i : \boldsymbol{\sigma}' dV - \int_{\Omega^e} B\bar{p} \nabla^x \cdot \boldsymbol{\eta}^i dV - \int_{\Omega^e} \boldsymbol{\eta}^i \cdot \rho \mathbf{g} dV - \int_{\partial\Omega_t^e} \boldsymbol{\eta}^i \cdot \hat{\mathbf{t}} dA = 0, \quad (6.117)$$

$$[R_{\text{frac}}]^e = \int_{\Omega^e} \varphi^i [2(1-d)\mathcal{H}] dV - \int_{\Omega^e} \frac{\mathcal{G}_c}{l} [\varphi^i + l^2(\nabla^x \varphi^i \cdot \nabla^x d)] dV = 0, \quad (6.118)$$

where i denotes a shape function index. We use standard linear shape functions in this work.

Next, we discretize the flow and transport problem via the finite volume method, on the same mesh used in the finite element discretization. We integrate the mass balance equations over each element, apply the divergence theorem to the fluid flux terms, and multiply the residuals by the time increment Δt . Element-wise contributions of the discrete mass residuals are then obtained as

$$[R_{\text{mass},l}]^e = \int_{\Omega^e} (1 - \phi^s) [\rho_l (S^l - S_n^l) + S^l (\rho_l - \rho_{l,n})] dV + \int_{\Omega^e} S^l \rho_l B \nabla^x \cdot (\mathbf{u} - \mathbf{u}_n) dV dV \\ - \Delta t \int_{\Omega^e} m_l dV + \Delta t \int_{\partial\Omega^e} \mathbf{w}_l \cdot \mathbf{n}^e dA - \Delta t \int_{\partial\Omega_q^e} \hat{q} dA = 0, \quad (6.119)$$

$$[R_{\text{mass},m}]^e = \int_{\Omega^e} \rho^l (c - c_n) dV + \int_{\Omega^e} c (1 - \phi^s) [\rho_l (S^l - S_n^l) + S^l (\rho_l - \rho_{l,n})] dV \\ + \int_{\Omega^e} c S^l \rho_l B \nabla^x \cdot (\mathbf{u} - \mathbf{u}_n) dV - \Delta t \int_{\Omega^e} m_l dV \\ + \Delta t \int_{\partial\Omega^e} (c \mathbf{w}_l) \cdot \mathbf{n}^e dA + \Delta t \int_{\partial\Omega^e} \mathbf{j} \cdot \mathbf{n}^e dA - \Delta t \int_{\partial\Omega_j^e} \hat{j} dA = 0, \quad (6.120)$$

$$[R_{\text{mass},c}]^e = \int_{\Omega^e} (1 - \phi^s) \rho_c (S^c - S_n^c) dV + \int_{\Omega^e} S^c \rho_c B \nabla^x \cdot (\mathbf{u} - \mathbf{u}_n) dV - \Delta t \int_{\Omega^e} m_c dV = 0, \quad (6.121)$$

where $\mathbf{n}^e$ is the outward normal to the boundary of element e . As depicted in Fig. 6.4, the interpolation functions for the pressure, mass fraction, and crystal saturation fields take a constant value of 1 over element e and 0 at all other elements. Thus the volume integrals can be evaluated much like integrating finite elements with a piecewise constant shape function.

The surface flux integrals are evaluated as a sum of interelement fluxes between element e and its neighboring elements f . Let n_{face} denote the number of faces of element e and $\mathbf{n}^{ef}$ denote the outward normal vector at the interface $\partial\Omega^{ef}$. The integral of the liquid mass flux is then expressed as

$$\int_{\partial\Omega^e} \mathbf{w}_l \cdot \mathbf{n}^e dA = \sum_{f=1}^{n_{\text{face}}} \int_{\partial\Omega^{ef}} \mathbf{w}_l \cdot \mathbf{n}^{ef} dA = \sum_{f=1}^{n_{\text{face}}} (w_l)^{ef}. \quad (6.122)$$

We apply a two-point flux approximation scheme for multiphase flow in porous media as described in Aziz et al. [2014], Jha and Juanes [2014]. Since $\mathbf{w}_l$ is governed by Darcy's law, $(w_l)^{ef}$ is given by

$$(w_l)^{ef} = \Gamma^{ef} (\Phi^e - \Phi^f). \quad (6.123)$$

Here, $\Phi^e = p^e + (\rho_l)^e g z^e$ and $\Phi^f = p^f + (\rho_l)^f g z^f$ are the flow potentials at elements e and f , respectively, where g denotes the gravitational acceleration and z denotes the elevations. Γ^{ef} is the transmissibility at the interface $\partial\Omega^{ef}$, which is multiplicatively decomposed into the geometric transmissibility T^{ef} and the fluid mobility $(\lambda_l)^{ef}$ as

$$\Gamma^{ef} = T^{ef} (\lambda_l)^{ef}. \quad (6.124)$$

The geometric transmissibility T^{ef} is estimated from the harmonic average of permeabilities of elements e and f

$$T^{ef} = \frac{A^{ef}}{l^e/k^e + l^f/k^f}, \quad (6.125)$$

where A^{ef} is the area of the interface $\partial\Omega^{ef}$, l^e and l^f are the distances between the center of the interface and the center of elements e and f , respectively, and k^e and k^f are permeabilities of elements e and f , respectively. Note that this term is independent of the flow direction. On the other hand, the fluid mobility $(\lambda_l)^{ef}$ is evaluated in an upstream weighting (upwinding) manner as follows:

$$(\lambda_l)^{ef} = \begin{cases} \rho_l^e k_r^e / \mu_l^e & \text{if } \Phi^e > \Phi^f, \\ \rho_l^f k_r^f / \mu_l^f & \text{otherwise.} \end{cases} \quad (6.126)$$

The remaining flux terms can be computed in a similar manner. For evaluating the surface integral of $c w_l \cdot \mathbf{n}^e$, c^e or c^f is multiplied to $(\lambda_l)^{ef}$ in Eq. (6.126) depending on the flow direction. Modification of the above equations to calculate the interelement fluxes of $j = -\rho_l D \nabla^x c$ is straightforward, and omitted for brevity.

6.5.4 Fully discrete form and staggered scheme for phase field

We now develop the matrix form of the discrete residuals as a linear system that needs to be solved in each Newton iteration. In doing so, we split the overall system into two parts—the phase-field part and the rest—and apply a staggered solution method developed for phase-field modeling of fracture. The primary motivation of using a staggered scheme is its computational robustness: it is far more convergent than a monolithic scheme during the evolution of phase-field fracture. Such robustness is particularly more desirable for phase-field modeling of fracture in strongly coupled multiphysics problems [Wilson et al. \[2013\]](#), [Zhang et al. \[2016\]](#). The phase-field system, which is uncoupled from other variables, is given by

$$J_{dd} \Delta d = -R_{\text{frac}}, \quad (6.127)$$

where J_{dd} is the Jacobian matrix of R_{frac} with respect to the phase-field variable d , and Δ denotes a Newton increment. Indeed the phase-field system is linear, so d can be updated by solving Eq. (6.127) once. Next, the rest part—the chemo-hydro-mechanics system—is given by the following block-partitioned system:

$$\begin{bmatrix} J_{uu} & J_{up} & J_{uc} & J_{us} \\ J_{pu} & J_{pp} & J_{pc} & J_{ps} \\ J_{cu} & J_{cp} & J_{cc} & J_{cs} \\ J_{su} & J_{sp} & J_{sc} & J_{ss} \end{bmatrix} \begin{bmatrix} \Delta u \\ \Delta p \\ \Delta c \\ \Delta s \end{bmatrix} = - \begin{bmatrix} R_{\text{mom}} \\ R_{\text{mass},l} \\ R_{\text{mass},d} \\ R_{\text{mass},c} \end{bmatrix}, \quad (6.128)$$

where $J_{(\cdot)(\cdot)}$ denotes a sub-matrix in the Jacobian matrix. Specific expressions for the sub-matrices are straightforward to obtain and omitted for brevity. This chemo-hydro-mechanics system is usually nonlinear due to water retention characteristics and crystallization kinetics, among other reasons. It is noted that $J_{s(\cdot)}$ and $J_{(\cdot)s}$ are nonzero only when the crystals are growing or dissolving in the pores.

Extending the staggered scheme proposed by Miehe et al. [Miehe et al. \[2010d\]](#), we proceed the numerical solution from time t_n to t_{n+1} through the following three sub-steps:

1. Determine $\mathcal{H}$ in the phase-field system with the displacement variable $\mathbf{u}$ at t_n .
2. With this $\mathcal{H}$, update the phase-field variable d at t_{n+1} , by solving Eq. (6.127).
3. With the updated d , update all other variables $\mathbf{u}/p/c/s$ at time t_{n+1} , by Newton's method solving Eq. (6.128) at each iteration.

In the last step, we solve the coupled chemo-hydro-mechanics system in a monolithic manner that updates the primary variables in Eq. (6.128) together in each iteration. However, other methods that sequentially solve the coupled system (e.g., Kim et al. [2015]) can also be used for the same purpose. Such sequentially-implicit methods may be preferred particularly when separate code is available for each physics, e.g., one for the deformation and fracture problem and another for the flow and transport problem. Yet, the analysis of White et al. [2016b] suggests that, with respect to computation cost *per se*, a sequential solution approach is suboptimal to a monolithic one employing a decent block-partitioned preconditioner. This motivates the following discussion on a block-partitioned preconditioner for our problem at hand.

6.5.5 Block-partitioned preconditioner

We now seek to design a quality preconditioner for the Jacobian matrix in the coupled chemo-hydro-mechanics system given by Eq. (6.128). To this end, we extend a block-partitioning strategy originally developed for poromechanics White et al. [2016b], White and Borja [2011b], Castelletto et al. [2016] to our problem which involves more coupled physics. For the purpose of preconditioning, we first consider a subset of the problem whereby crystal kinetics are insignificant (i.e., $J_s(\cdot) \rightarrow 0$ and $J_{(\cdot)s} \rightarrow 0$). In such cases, the Jacobian matrix reduces to

$$J = \begin{bmatrix} J_{uu} & J_{up} & J_{uc} \\ J_{pu} & J_{pp} & J_{pc} \\ J_{cu} & J_{cp} & J_{cc} \end{bmatrix}, \quad (6.129)$$

which is a 3 by 3 block-partitioned matrix. Our particular interest is in developing a block lower-triangular preconditioner of the form

$$P^{-1} = \begin{bmatrix} X & 0 & 0 \\ L_1 & Y & 0 \\ L_2 & L_3 & Z \end{bmatrix}, \quad (6.130)$$

which, when multiplied to the Jacobian matrix J , results in a matrix very close to the block upper-triangular matrix obtained from the LDU factorization of J . In other words, we want a block lower-triangular matrix P^{-1} such that

$$P^{-1}J \approx \begin{bmatrix} I & U_1 & U_2 \\ 0 & I & U_3 \\ 0 & 0 & I \end{bmatrix}, \quad (6.131)$$

irrespective of U_1 , U_2 , and U_3 . If this equation is satisfied exactly, the preconditioned Jacobian has a single eigenvalue of 1 and the Jacobian system can be solved by a couple of Krylov iterations. Such an exact preconditioner, however, is generally very expensive to construct.

Therefore, to find an effective yet inexpensive preconditioner, we take the following three-step approach:

1. Find the block preconditioner that exactly satisfies Eq. (6.131).
2. Approximate dense terms in the exact block preconditioner.
3. Replace inverse operations in the approximated block preconditioner with their own preconditioners.

This approach has been put forward by White and co-workers [White et al. \[2016b\]](#), [White and Borja \[2011b\]](#), [Castelletto et al. \[2016\]](#) for coupled poromechanics, and it has proven very useful for arriving at a scalable preconditioner. In the following we apply this approach to the coupled chemo-hydro-mechanics problem of our interest.

First, we solve for Eq. (6.131) to find a block lower-triangular matrix P^{-1} that exactly satisfies this equation. We then obtain six equations for the six sub-matrices of P^{-1} as follows:

$$XJ_{uu} = I, \quad (6.132)$$

$$L_1J_{uu} + YJ_{pu} = 0, \quad (6.133)$$

$$L_1J_{up} + YJ_{pp} = I, \quad (6.134)$$

$$L_2J_{uu} + L_3J_{pu} + ZJ_{cu} = 0, \quad (6.135)$$

$$L_2J_{up} + L_3J_{pp} + ZJ_{cp} = 0, \quad (6.136)$$

$$L_3J_{pc} + ZJ_{cc} = I, \quad (6.137)$$

The first equation obviously leads to $X = J_{uu}^{-1}$. From the second and third equations we get

$$L_1 = -YJ_{pu}J_{uu}^{-1}, \quad (6.138)$$

$$Y = (J_{pp} - J_{pu}J_{uu}^{-1}J_{up})^{-1}. \quad (6.139)$$

Here, $J_{pp} - J_{pu}J_{uu}^{-1}J_{up}$ is the Schur complement of the block J_{uu} of the upper left 2 by 2 block matrix, which will be denoted by S_u in the following. Solving the rest, we get

$$L_2 = -Z[J_{cu} - (J_{cp} - J_{cu}J_{uu}^{-1}J_{up})S_u^{-1}J_{pu}]J_{uu}^{-1}, \quad (6.140)$$

$$L_3 = -Z(J_{cp} - J_{cu}J_{uu}^{-1}J_{up})S_u^{-1}, \quad (6.141)$$

$$Z = [J_{cc} - J_{cu}J_{uu}^{-1}J_{uc} - (J_{cp} - J_{cu}J_{uu}^{-1}J_{up})S_u^{-1}(J_{pc} - J_{pu}J_{uu}^{-1}J_{uc})]^{-1}. \quad (6.142)$$

Similar to before, Z appears in L_2 and L_3 .

Second, we approximate dense terms in the exact preconditioner derived above. Of particular interest is in finding good approximations of those containing J_{uu}^{-1} multiplied by coupling matrices, e.g., $S_u = J_{pp} - J_{pu}J_{uu}^{-1}J_{up}$. These terms and their inverses appear all blocks of the exact P^{-1} except the X block, but their exact values are overly expensive to be used for a preconditioning purpose. Thus we will approximate these terms, drawing on the “fixed-stress” split scheme for poromechanics which has been widely used as a sequential solution method [Kim et al. \[2011\]](#), [Mikelić and Wheeler \[2013\]](#) and recently rephrased as a block-preconditioning approach [White et al. \[2016b\]](#). The split scheme goes as follows. If the mean volumetric stress is assumed to be fixed during fluid flow, the divergence of the solid velocity can be related to the change of pore pressure only, i.e.,

$$\dot{\sigma}_{\text{vol}} = K \nabla^x \cdot \boldsymbol{v} - B\dot{p} = 0 \quad \rightarrow \quad \nabla^x \cdot \boldsymbol{v} = \frac{B}{K}\dot{p} \quad (6.143)$$

where K is the bulk modulus of the solid matrix. Substituting this expression into the mass balance equations for the liquid and dissolved minerals, Eqs. (6.96) and (6.99), we get the fixed-stress approximations of these mass balance equations as

$$(1 - \phi^s)(\rho_l \dot{S}^l + S^l \dot{\rho}_l) + S^l \rho_l \frac{B^2}{K} \dot{p} + \nabla^x \cdot \mathbf{w}_l = m_l, \quad (6.144)$$

and

$$\rho^l \dot{c} + c(1 - \phi^s)(\rho_l \dot{S}^l + S^l \dot{\rho}_l) + c \left(S^l \rho_l \frac{B^2}{K} \dot{p} \right) + \nabla^x \cdot (c \mathbf{w}_l) + \nabla^x \cdot \mathbf{j} = m_l. \quad (6.145)$$

Observe that the displacement vector $\mathbf{u}$ is now eliminated from the original mass balance equations. Discretization of these equations gives sparse approximations of the coupling terms containing J_{uu}^{-1} . For example, the approximated Schur complement, denoted by $\tilde{S}_U$, is given by

$$S_U \approx J_{pp} + S^l \rho_l \frac{B^2}{K} V^e := \tilde{S}_U, \quad (6.146)$$

where V^e is the volume of elements assembled in an element-wise manner (corresponds to the mass matrix of the pressure shape functions in finite elements). Here, the original dense term $J_{pu} J_{uu}^{-1} J_{up}$ is approximated by a sparse term (the second term). This approximation reduces to that derived by White et al. [White et al. \[2016b\]](#) in case the material is fully saturated ($S^l = 1$) and ρ_l is constant. Similarly, we approximate other terms as

$$J_{cp} - J_{cu} J_{uu}^{-1} J_{up} \approx J_{cp} + c S^l \rho_l \frac{B^2}{K} V^e := \tilde{J}_{cp}, \quad (6.147)$$

$$J_{pc} - J_{pu} J_{uu}^{-1} J_{uc} \approx J_{pc} + S^l \frac{\partial \rho_l}{\partial c} \frac{B^2}{K} (p - p_n) V^e := \tilde{J}_{pc}, \quad (6.148)$$

$$J_{cc} - J_{cu} J_{uu}^{-1} J_{uc} \approx J_{cc} + S^l \frac{\partial \rho_l}{\partial c} \frac{B^2}{K} (p - p_n) V^e := \tilde{J}_{cc}, \quad (6.149)$$

Accordingly, the Z block is also approximated as

$$Z \approx (\tilde{J}_{cc} - \tilde{J}_{cp} \tilde{S}_U^{-1} \tilde{J}_{pc})^{-1}. \quad (6.150)$$

Third, we replace inverse operations in the approximated block preconditioner—which are usually expensive to compute—with their own preconditioners. Specifically,

$$J_{uu}^{-1} \approx P_{J_{uu}}^{-1}, \quad \tilde{S}_U^{-1} \approx P_{\tilde{S}_U}^{-1}, \quad \tilde{J}_{cc}^{-1} \approx P_{\tilde{J}_{cc}}^{-1}, \quad (6.151)$$

where $P_{(\cdot)}^{-1}$ is a preconditioner for $(\cdot)^{-1}$. We use algebraic multigrid preconditioners in this work, but a number of other combinations of sub-preconditioners can be efficient as well. Discussions on the choice of sub-preconditioners are presented in [White et al. \[2016b\]](#), [White and Borja \[2011b\]](#), [Castelletto et al. \[2016\]](#) in the context of poromechanics problems.

Following the three-step procedure described above, we finally arrive at a specific expression for Eq. (6.130) as

$$P^{-1} = \begin{bmatrix} P_{J_{uu}}^{-1} & 0 & 0 \\ -P_{\tilde{S}_U}^{-1} J_{pu} P_{J_{uu}}^{-1} & P_{\tilde{S}_U}^{-1} & 0 \\ -P_{\tilde{J}_{cc}}^{-1} (J_{cu} - \tilde{J}_{cp} P_{\tilde{S}_U}^{-1} \tilde{J}_{pu}) P_{J_{uu}}^{-1} & -P_{\tilde{J}_{cc}}^{-1} \tilde{J}_{cp} P_{\tilde{S}_U}^{-1} & P_{\tilde{J}_{cc}}^{-1} \end{bmatrix}. \quad (6.152)$$

It would be illuminating to demonstrate how this block-preconditioner operates in real problems. Indeed, what is needed by an iterative solution method is the matrix-vector product of the preconditioner and an input vector $\mathbf{x}$, say $P^{-1}\mathbf{x} = \mathbf{y}$. This operation can be written as

$$\begin{bmatrix} P_{J_{uu}}^{-1} & 0 & 0 \\ -P_{\tilde{S}_u}^{-1}J_{pu}P_{J_{uu}}^{-1} & P_{\tilde{S}_u}^{-1} & 0 \\ -P_{\tilde{J}_{cc}}^{-1}(J_{cu} - \tilde{J}_{cp}P_{\tilde{S}_u}^{-1}\tilde{J}_{pu})P_{J_{uu}}^{-1} & -P_{\tilde{J}_{cc}}^{-1}\tilde{J}_{cp}P_{\tilde{S}_u}^{-1} & P_{\tilde{J}_{cc}}^{-1} \end{bmatrix} \begin{Bmatrix} x_u \\ x_p \\ x_c \end{Bmatrix} = \begin{Bmatrix} y_u \\ y_p \\ y_c \end{Bmatrix}. \quad (6.153)$$

The vector $\mathbf{y}$ can be efficiently updated via the following three steps:

1. $y_u = P_{J_{uu}}^{-1}x_u$
2. $y_p = P_{\tilde{S}_u}^{-1}(x_p - J_{pu}y_u)$
3. $y_c = P_{\tilde{J}_{cc}}^{-1}(x_c - J_{cu}y_u - \tilde{J}_{cp}y_p)$

Notice that, from the second step, we use the block(s) of $\mathbf{y}$ that we updated in previous step(s)— y_u in the second step, and y_u and y_p in the third step. In this sense, this block-preconditioning approach can also be interpreted as a sequential solution method. See White et al. [White et al. \[2016b\]](#) for a thorough discussion on this interpretation. The repeated pattern in this sequence can be further applied to update the additional block related to the crystal kinetics, which we have observed satisfactory results in the numerical examples that follow.

The proposed block-preconditioner has shown decent performance (usually less than 20 Krylov iterations for a tolerance of 10^{-6}) for a number of chemo-hydro-mechanical problems we have tested, and it is believed to be a good choice for other problems sharing a similar mathematical structure. However, much more extensive studies are needed to verify and improve the performance of the current preconditioner—especially with respect to its scalability in large computing platforms, as done by [White et al. \[2016b\]](#), [White and Borja \[2011b\]](#), [Castelletto et al. \[2016\]](#) for poromechanical problems. To better focus on the scope of the current work, we leave this topic as a future research direction.

6.6 Numerical examples

In this section, two numerical examples are presented to validate and demonstrate the capability of the developed computational model for simulating complex interactions among unsaturated flow, solid deformation, fracturing, and crystallization in pores. The first example serves as a benchmark problem for verifying the numerical implementation and validating the mathematical model. We simulate a validation problem in unsaturated poromechanics, because a benchmark setting for chemo-hydro-mechanics involving in-pore crystallization is yet to emerge to the best of our knowledge. Subsequently, in the second example we incorporate the flow, transport, and crystallization of minerals and the resulting fracturing process in the host material. We particularly consider the problem of capillary infiltration of saturated solution into a porous medium, which has been commonly used by a number of experimental and numerical studies on crystallization of salts in geomaterials (e.g., [Castellazzi et al. \[2013\]](#), [Noiriel et al. \[2010\]](#), [Dai et al. \[2016\]](#)). The main purpose of this example is to showcase the performance of the computational model for simulating the development of fully cracked regions ensuing crystallization in pores.

The numerical examples have been prepared using `Geocentric`, a massively parallel finite element code for geomechanics that has been used in a number of previous studies (e.g., Choo et al. [2016b], Borja and Choo [2016b], Borja et al. [2016], Choo and Sun [2018a], Choo [2016], White et al. [2016b], White and Borja [2011b], Choo and Borja [2015b]). This code is built upon the `deal.II` finite element library Bangerth et al. [2007b], `p4est` mesh handling library Burstedde et al. [2011b], and the `Trilinos` project Heroux and Willenbring [2012b].

6.6.1 Drainage of freshwater in a porous column

Our first example has two purposes: (1) to verify the numerical implementation of combined finite element and finite volume discretization in the current code, and (2) to validate the mathematical model's capability for capturing coupled hydro-mechanical responses that can lead to crystallization of materials. For the latter purpose, we particularly focus on a drainage (saturation decreasing) process whereby supersaturation and crystallization can take place.

To achieve these two purposes simultaneously, we consider a benchmark problem of unsaturated poromechanics that simulates Liakopoulos' drainage experiment of a sand column Liakopoulos [1964]. This problem has been used by a number of previous studies to validate their own combinations of governing equations, effective stress, water retention, relative permeability, and others (e.g., Schrefler and Xiaoyong [1993], Kluber-tanz et al. [1997], Schrefler and Scotta [2001], Ehlers et al. [2004], Liu et al. [2014]). Here we use this problem in the same context. To verify the implementation, we simulate the drainage experiment with the current code using the finite element method for the solid deformation problem and the finite volume method for the fluid flow problem, and compare the results with those obtained by unsaturated poromechanics code using mixed finite elements for both the solid deformation and fluid flow problems (see Borja and Koliji [2009], Borja et al. [2016], Choo [2016], White and Borja [2011b] for details of the mixed finite element discretization). It is noted that, for problems like this one that do not involve a strong advection phenomenon, finite volume and finite element results should converge to the same solution. The numerical results are also compared with the experimental data of Liakopoulos Liakopoulos [1964] to validate the mathematical model.

Figure 6.5 shows a schematic illustration of the problem setup. A 0.1 m wide and 1.0 m tall column is prepared such that it is initially under equilibrium with zero pore pressure. The test begins by making the top boundary undrained, which leads to drainage of the pore fluid through the bottom boundary. During the test the bottom boundary is kept zero pressure and fixed, whereas no flux and no lateral displacement are allowed along the side boundaries. Both of the fluid flow and solid deformation in this problem are one-dimensional, and the column is discretized by 80 quadrilateral elements along the height. The same mesh is used for both finite element/finite volume and mixed finite elements simulations. With a uniform time increment $\Delta t = 1$ minute, we simulate the experiment until the drainage time reaches 600 minutes.

Material parameters are assigned close to the properties of the Del Monte sand tested by Liakopoulos Liakopoulos [1964]. The material is composed of sand grains and fresh pore water whose intrinsic densities are $\rho_s = 2700 \text{ kg/m}^3$ and $\rho_w = 1000 \text{ kg/m}^3$, respectively, with porosity $\phi = 1 - \phi^s = 0.2975$. As for hydraulic parameters, we use the absolute permeability $k = 4.5 \times 10^{-13} \text{ m}^2$ (isotropic), the dynamic viscosity $\mu_l = 10^{-6} \text{ kPa}\cdot\text{s}$, and the van Genuchten model parameters $S_1 = 0$, $S_2 = 1$, $\alpha_{ca} = 15 \text{ kPa}$, and $n = 5$. Because no mechanical data is available for the tested sand, we assume Young's modulus

$E = 800 \text{ kPa}$ and Poisson's ratio $\nu = 0.25$.

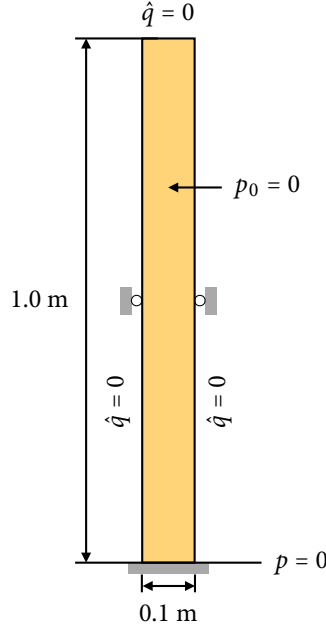


Fig. 6.5. Schematic illustration of the column drainage example that simulates Liakopoulos' experiment [Liakopoulos \[1964\]](#). Initially, the domain is fully saturated with zero pore pressure ($p_0 = 0$). Once the simulation begins, the pore fluid is drained through the bottom outlet filter by gravitational force.

In Fig. 6.6 we present simulation results obtained by the finite element/finite volume and mixed finite element schemes, along with the experimental data of Liakopoulos [Liakopoulos \[1964\]](#), in terms of time evolutions of pore pressure variation along the height and fluid velocity at the bottom outlet filter. First, we find that the two numerical schemes yield virtually identical results, thus verify our implementation of combined finite element/finite volume discretization. Second, we see that the mathematical model well reproduces the spatio-temporal evolution of pore pressure measured in the experiment. In Fig. 6.6 the agreement between the simulation and experiments is rather qualitative in the beginning, but it becomes increasingly quantitative as time proceeds. Figure 6.6 shows that the calculated outlet velocities are in an excellent agreement with the measured data throughout the test. These results demonstrate that, even when the solid behavior is grossly simplified, the computational model can capture salient physics of coupled solid deformation and unsaturated flow in real geomaterials.

6.6.2 Capillary infiltration of salt water and crystallization-induced cracking

We now proceed to the simulation of chemo-hydro-mechanical processes involving crystallization in pores and resulting damage in the solid matrix. Specifically, we simulate a laboratory-scale problem analogous to a typical experiment studying growth of salt crystals in geomaterials [Noiriel et al. \[2010\]](#), [Dai et al. \[2016\]](#), in which a specimen is infiltrated by salt water by capillarity and then damaged by crystallization of salt minerals in pores.

Figure 6.7 illustrates the setup of this problem. We consider a porous rock column of width 0.025 m and height 0.1 m. The column is initially filled with freshwater and

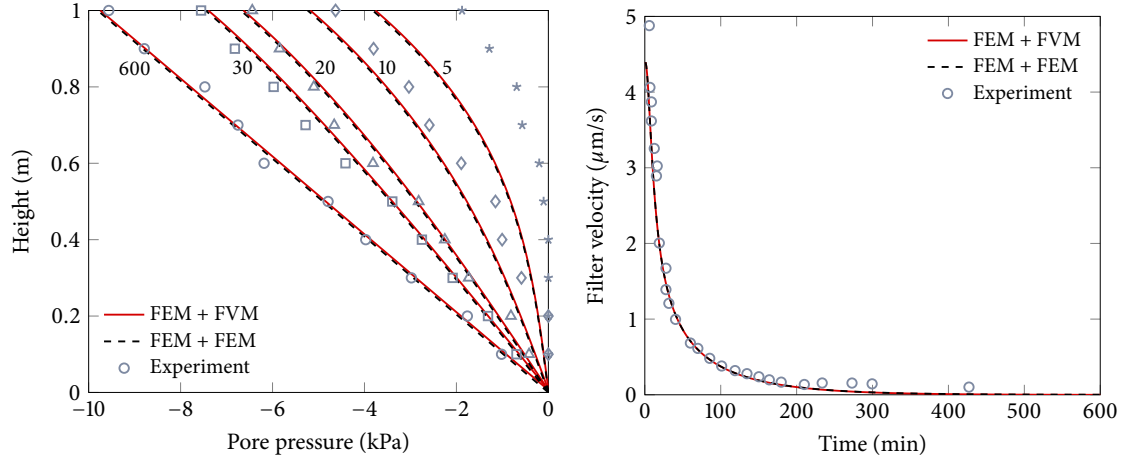


Fig. 6.6. Drainage simulation results of the current code using finite elements for the solid deformation problem and finite volume for the fluid flow problem (FEM + FVM), and an unsaturated poromechanics code using finite elements for both the solid deformation and fluid flow problems (FEM + FEM). The numerical results are compared with experimental data of Liakopoulos [Liakopoulos \[1964\]](#). (a) Pore pressure variation along the height (numbers inside the figure denote elapsed time in minutes). (b) Fluid velocity at the bottom outlet filter.

air, manifesting a uniform capillary pressure of 20 kPa. To emulate capillary rise of a mineral solution, we prescribe the pressure and concentration at the bottom boundary as 1 kPa and c_{eq} (equilibrium solute mass fraction), respectively. The liquid solution is not allowed to flow outside through the lower half of the lateral boundaries as well as the top boundary, like in the experiment of Noiriél et al. [Noiriél et al. \[2010\]](#). In contrast, the solution is subject to a constant outward flux boundary condition of $\hat{q} = -2 \times 10^{-5}$ kg/m²/s in the upper half of the lateral boundaries. Throughout the domain the dissolved minerals are subject to no flux boundary conditions such that the minerals stay in the pores during an outflow of the liquid solution. These boundary conditions are intended to drive supersaturation of the solution—and ultimately crystallization in pores—in the upper half of the specimen. Exploiting the symmetry of the problem, we model the left half of the domain and reflect the result in the post-processing stage. The half domain is discretized by 32,768 quadrilateral elements of uniform size (mesh diameter $h = 0.025$ cm), which leads to 198,339 degrees of freedom for the five primary variables.

The material is assumed to be similar to the porous rock studied in Rasmussen et al. [Rasmussen et al. \[1993\]](#). We assign the material's properties as follows: solid density $\rho_s = 2550$ kg/m³, initial porosity is $\phi_0 = 1 - \phi^s = 0.172$, absolute permeability $k = 4.3 \times 10^{-16}$ m², and van Genuchten parameters $S_1 = 0.4$, $S_2 = 1.0$, $\alpha_{ca} = 50$ kPa, and $n = 1.65$. The linear elasticity parameters of the solid matrix are assumed to be $E = 10$ GPa and $\nu = 0.25$. The properties of the pore water remain unchanged from the previous example. As for the mineral, we consider salt (sodium chloride), and adopt its parameters mainly from Castellazzi et al. [Castellazzi et al. \[2013\]](#). They are: the mineral density, $\rho_m = 2160$ kg/m³, the molar volume, $V_m = 27$ cm³/mol, the equilibrium solute mass fraction, $c_{eq} = 0.264$ kg/kg, the diffusion coefficient over tortuosity, $D_m/\tau = 0.5 \times 10^{-9}$ m²/s, and the kinetic parameters, $K_c = 4 \times 10^{-4}$ kg/m³·s, $g_c = 1.0$, and $U_{thr} = 1.6$. The temperature is set as $T = 293.15$ K (20°C). Lastly, using Eqs. (6.90) to (6.92) with

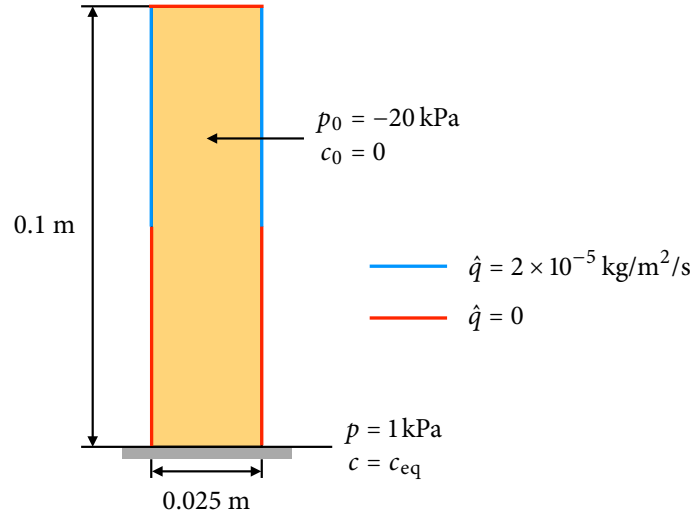


Fig. 6.7. Schematic illustration of the capillary rise example. The domain is initially filled with freshwater and air such that they manifest a capillary pressure of 20 kPa. The simulation begins by prescribing the pressure and mass fraction at the bottom boundary as 1 kPa and c_{eq} , respectively. The lower half of the boundaries are no flux boundaries, whereas the upper half domain is subject to a constant liquid flux of $-2 \times 10^{-5} \text{ kg/m}^2/\text{s}$. Mineral flux $\hat{j}$ is zero throughout the boundaries.

$\beta = 0.1453$ Zhang [2002] and $\sigma_t = 5 \text{ MPa}$, we get the phase-field model parameters as $\mathcal{G}_c = 49.5 \text{ J/m}^2$ and $l = 2.1 \times 10^{-3} \text{ m}$. It is noted that the ratio of the length parameter to the mesh diameter l/h is greater than 8, which has been shown to give sufficient accuracy Borden et al. [2012].

Once the simulation begins, the salt water infiltrates the specimen from the bottom boundary. Figure 6.8 shows the evolution of the supersaturation ratio U until 2760 minutes. At 30 minutes and 720 minutes the maximum supersaturation ratio equals unity, since no outflow is allowed through the lower half boundaries for both the solution and dissolved minerals. However, when the solution reaches the upper half boundaries, the water in the solution flows outside through the boundaries, whereas the dissolved minerals remain inside the specimen. This type of outflow makes the supersaturation ratio higher than unity. As a result, the contour at 2760 minutes show supersaturated zones in which $U > 1$. We can also find that the supersaturation ratio exceeds $U_{thr} = 1.6$ in some regions, which means that some minerals have been crystallized.

Figures 6.9 and 6.10 present the crystal saturation ratio, S^c , and the crystallization pressure, p_{cr} , at three time instances from 2760 minutes to 4092 minutes. Figure 6.9 shows that during this time crystals grow continuously at the upper half of the lateral boundaries, which may be viewed as salt efflorescence. We can also see from Fig. 6.10 that this crystal growth leads to an increase in crystallization pressure. Recall that the crystallization pressure enters the mean pore pressure in effective stress. As such, this increase in the crystallization pressure can give rise to tensile effective stress that drives the evolution of the phase-field variable.

In Fig. 6.11 we plot the evolution of the phase-field variable d from 4092 minutes, with a uniform time interval of 0.1 minute. Until 4092.1 minutes the phase-field value does not reach 1.0, so the material may be regarded as partially damaged by microcracks. After 0.1 minute, the phase-field value evolves to 1 at some locations, which means that the

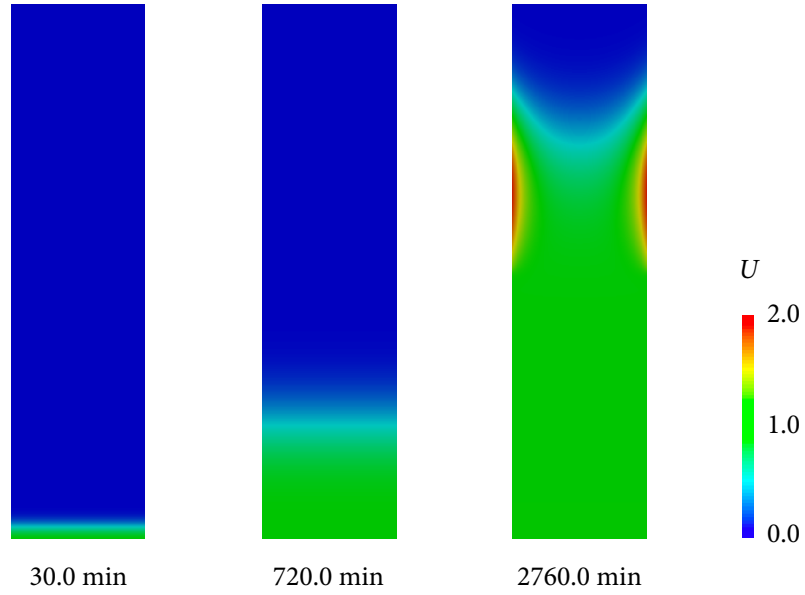


Fig. 6.8. Evolution of the supersaturation ratio U until 2760 minutes.

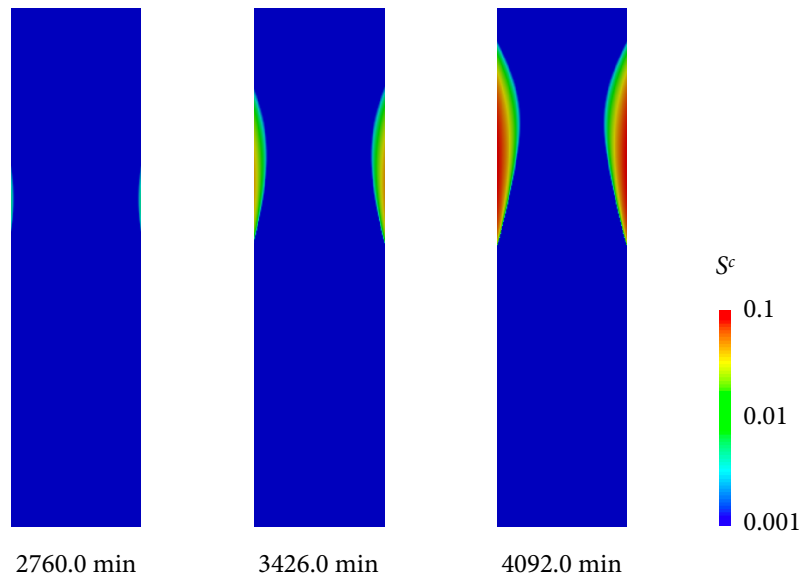


Fig. 6.9. Evolution of the crystal saturation S^c from 2760 minutes to 4092 minutes. Color bar in log scale.

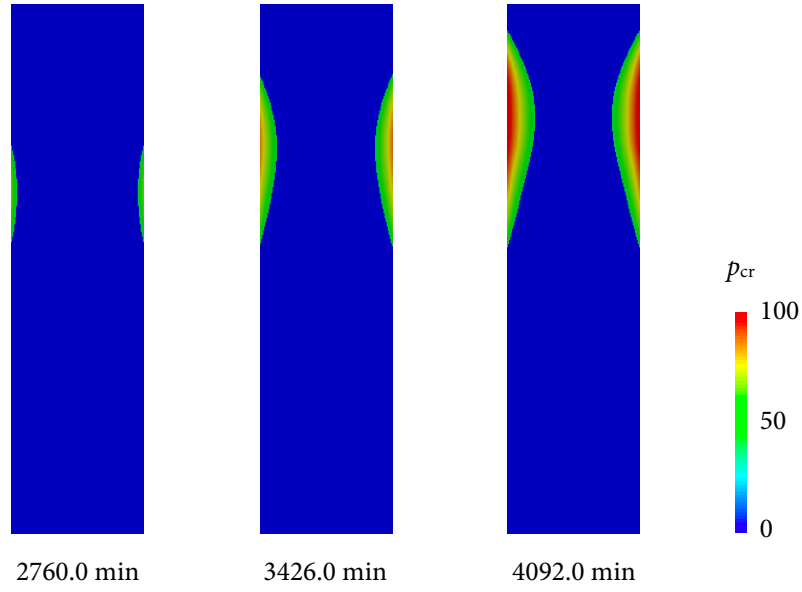


Fig. 6.10. Evolution of the crystallization pressure p_{cr} from 2760 minutes to 4092 minutes. Color bar in MPa.

material is fully cracked therein. The cracked zones enlarge further in the next 0.1 minute. It is noted that the shape of the cracked regions at 4092.3 minutes resembles characteristic cave-like damage zones in salt weathering. To the best of our knowledge, this is the first numerical simulation of the evolution of damage and fully-cracked zones by in-pore crystallization of minerals.

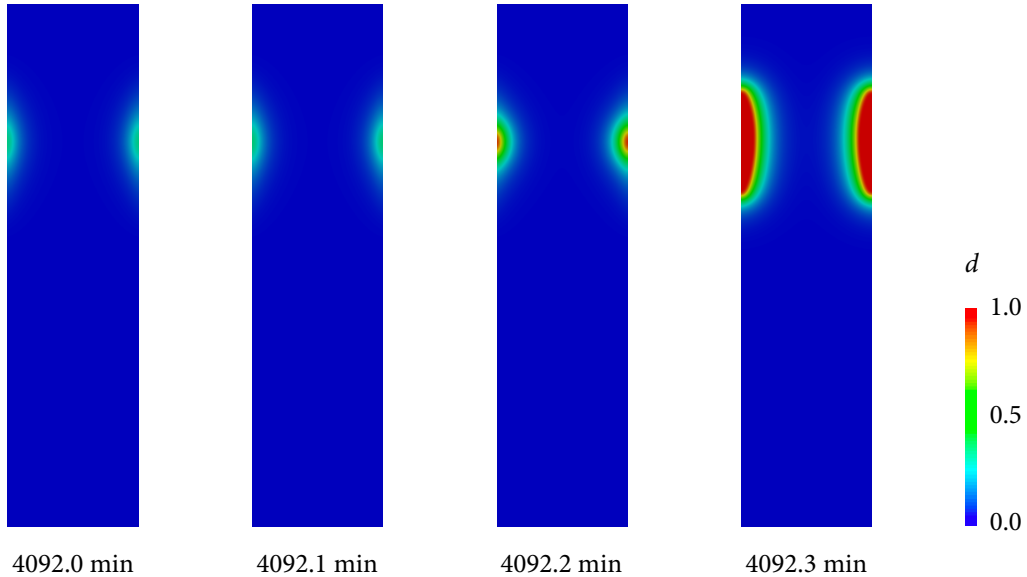


Fig. 6.11. Evolution of the phase-field variable d from 4092.0 minutes to 4092.3 minutes.

The simulation is terminated at this point, because many assumptions in the constitutive models break down upon fracturing of the material. For example, the crystallization pressure equations in the literature have been proposed for a crystal confined in small pore space such that a solution film exists between the crystal and the solid matrix. Therefore,

when cracks develop significantly as 4092.3 minutes in Fig. 6.11, these equations might become invalid or the crystallization pressure might disappear. Also, we have assumed that the kinetic parameter K_c is constant, but it is expected to evolve by the fracturing process. Note, however, that all these limitations stem from the lack of constitutive relations for crystals within fractures, not from the modeling framework. Once constitutive models that address the aforementioned issue become available, they can be cast into the current modeling framework.

Lastly, we would like to discuss computational aspects of the problem. As shown in Fig. 6.11, the phase-field variable has evolved from partial damage to full cracks dramatically within the last 0.2 minute. To capture such an accelerated cracking process, we have used an adaptive time stepping algorithm proposed by Borden et al. [Borden et al. \[2016b\]](#). As a result, during the cracking process the time increments have been reduced to very small numbers (orders of magnitude smaller than a minute), giving rise to undrained deformations in which the relative flow of the pore fluid is negligible. This aspect further justifies our choice of finite volume discretization for the flow problem, because mixed finite elements for coupled poromechanics are subject to an inf-sup stability condition in undrained conditions [Choo and Borja \[2015b\]](#), [White and Borja \[2008b\]](#), [Sun et al. \[2013b\]](#), [Sun \[2015b\]](#), [Krischok and Linder \[2016b\]](#). We also would like to mention that solving a large number of steps in the crack development stage has been made affordable thanks to the block-partitioned preconditioner described in the previous section. This preconditioner has allowed us to solve each linear system via a couple of dozens Krylov iterations until large cracked zones emerge.

6.7 Closure

This paper has presented a theoretical and computational framework for modeling cracking and damage in porous materials by in-pore crystallization of minerals. The framework combines a chemo-hydro-mechanics approach with a phase-field description of fracture. Particular contributions of this work include: (1) derivation of a thermodynamically consistent effective stress tensor in porous materials containing growing crystals, (2) identification of state variables that must be linked via constitutive laws, and (3) a block-preconditioned iterative solver that facilitates numerical solution of the fully coupled chemo-hydro-mechanics equations. The computational model has been demonstrated to be capable of simulating the onset and evolution of cracking and damage from in-pore crystallization of minerals.

In this work we have proposed a general modeling framework and then constructed its particular class by employing relatively simple constitutive models of crystallization kinetics and solid deformation. We would like to note that use of more advanced constitutive models for these physics may lead to other classes that have better predictive capabilities. Particularly, if quantifiable, pore-scale characteristics need to be incorporated into the crystallization kinetics model. An important example is the pore size distribution because crystallization in real geomaterials begins from smaller pores. The orientation of the mineral crystal may also exert control on this problem [Lecampion \[2010\]](#). As for the solid model, incorporating pressure-dependent plasticity will allow us to accommodate the effect of confining pressure on fracturing [Choo and Sun \[2018a\]](#), which is important for deep subsurface problems. Beyond constitutive models, the temperature field must be augmented to the formulation when heat flow is significant. For problems involving complex drying phenomena, one needs to introduce additional phases for vapors and

models of moisture transport [Scherer \[1990\]](#), [Kowalski and Rybicki \[1996\]](#), [Kowalski \[2003\]](#). The framework developed in this work is amenable to incorporating these more complicated physics.

Chapter 7

Modeling polycrystalline rock salt

We present a new thermal-mechanical-chemical-phase field model that captures the multi-physical coupling effects of precipitation creeping, crystal plasticity, anisotropic fracture, and crack healing in polycrystalline rock at various temperature and strain-rate regimes. This model is solved via a fast Fourier transfer solver with an operator-split algorithm to update displacement, temperature and phase field, and chemical concentration incrementally. In nuclear waste disposal in salt formation, brine inside the crystal salt may migrate along the grain boundary and cracks due to the gradient of interfacial energy and pressure. This migration has a significant implication on the permeability evolution, creep deformation, and crack healing within rock salt but is difficult to incorporate implicitly via computational homogenization of effective medium. As such, we introduce a thermodynamic framework and a corresponding computational implementation that explicitly captures the brine diffusion along the grain boundary and crack at the grain scale. Meanwhile, the anisotropic fracture and healing are captured via a high-order phase field that represents the regularized crack region in which a newly derived non-monotonic driving force is used to capture the fracture and healing due to the solution precipitation. Numerical examples are presented to demonstrate the capacity of the thermodynamic framework to capture the multiphysics material behaviors of rock salt.

7.1 Introduction

Rock salt formation has been widely considered as one of the potential repositories for nuclear waste disposal for decades. The design of these salt repositories often involves re-consolidated crushed salt as buffer or backfill materials to reduce excavation void space and the time required for the salt to close in around the nuclear waste. Due to the high thermal conductivity, the low permeability, the self-healing properties, and the ready availability of crushed salt in a repository, the re-consolidated crushed salt has attracted a significant amount of interest and become a major focus point in many studies by the US Department of Energy for heat-generating waste (e.g. [Kuhlman \[2013\]](#), [Martin et al. \[2015\]](#)). However, during the excavation process, micro-cracks may form in the salt materials within the excavation damaged zone (EDZ) near the repository surface. Furthermore, the reconsolidation process of the crushed rock salt as the backfill material will also introduce defects and impurities such as brine, micro-cracks, pores, and a small amount of clay. These imperfections will evolve under thermal-mechanical loadings via different mechanisms, such as the deformation-induced perlocation [[Ghanbarzadeh et al.](#),

2015], microcrack propagation [Zhu et al., 2015], and crack healing [Houben et al., 2013, Koelemeijer et al., 2012, Franssen and Spiers, 1990, Heard, 1972]. To prevent leakage of the radioactive materials, it is necessary to understand how effective permeability of rock salt evolves under different temperatures and in situ stress.

This article is Part II of the paper series *Computational thermomechanics of crystalline rock*, preceded by Na and Sun [2018], which focuses on the modeling of single-crystal salt. Our objective in this new contribution is to propose a computational framework for polycrystalline rock salt that explicitly captures the rate-dependent multi-physical coupling mechanisms that lead to a variety of anisotropic creeping, fracture, healing and plasticity under different temperature and pressure ranges at the mesoscale level. Due to the coupling nature of the healing, fracture, solution precipitation creep, and heat transfer across length scales, it is difficult to derive macroscopic predictive models that capture the interactions of those complex mechanisms via phenomenological or even microstructure-inspired path-dependent material laws. As a result, our goal is to propose a model that directly simulate those multiphysics phenomena occurred at the polycrystalline microstructures, rather than introducing phenomenological constitutive laws for a homogenized effective medium [Bryant and Sun, 2019, Pouya et al., 2016].

7.1.1 Fracture and healing in rock salt: experimental evidence

The path-dependent deformation of polycrystalline rock salt is primarily dominated by three mechanisms that across different length and time scales – dislocation creeping, solution precipitation, and micro-cracking dilatancy [Li and Urai, 2016]. The dislocation creep typically refers to the dislocation sliding along the slip planes or climbing perpendicular to the slip planes. Dislocation creep is the major deformation mechanism when the strain rate is larger than $1.0 \times 10^{-7} \text{ s}^{-1}$ [Li and Urai, 2016]. When the strain rate is below $1.0 \times 10^{-7} \text{ s}^{-1}$, solution precipitation becomes the major deformation mechanism due to the existence of intergranular brine. Figure 7.1 shows the typical microstructure of a polycrystalline rock salt specimen with “island-channel” type grain boundaries. For a rock salt in a natural environment, the grain boundary regions are usually filled with saturated brine, which assists the solution precipitation process. Meanwhile, the creeping due to intergranular solution precipitation is the results of three sequential physical processes (cf. Kruzhanov and Stöckhert [1998]): (1) the dissolution of solid phase across solid/liquid interface at high-pressure region; (2) the solute diffusion within intergranular brine due to the concentration gradient; and (3) the solute precipitation from brine to the solid phase at the low-pressure region. The efficiency of the solution precipitation is pre-dominated by the slowest of these three sequential processes, which is diffusion rather than dissolution or precipitation for wet rock salt [Hickman and Evans, 1995]. Therefore, solution precipitation is largely influenced by pressure gradient [Schott et al., 2009], grain boundary structure [van Noort et al., 2008], grain boundary misorientation [Van Noort et al., 2007], and concentration of aqueous trace metals [Alkattan et al., 1997].

The long-term creep behavior is also affected by the grain boundary or crack healing process [Desbois et al., 2012c], which not only reduces the permeability but also restore the stiffness [Houben et al., 2013]. Three major mechanisms account for crack healing [Houben et al., 2013, Koelemeijer et al., 2012]: (1) instantaneous mechanical closure due to increased grain boundary normal pressure; (2) diffusive crack-healing driven by surface energy reduction; and (3) crack healing by recrystallization. Among these healing mechanisms, the diffusive crack healing driven by surface energy reduction is the most important in the long term crack healing of natural rock salt. Along with the pressure-

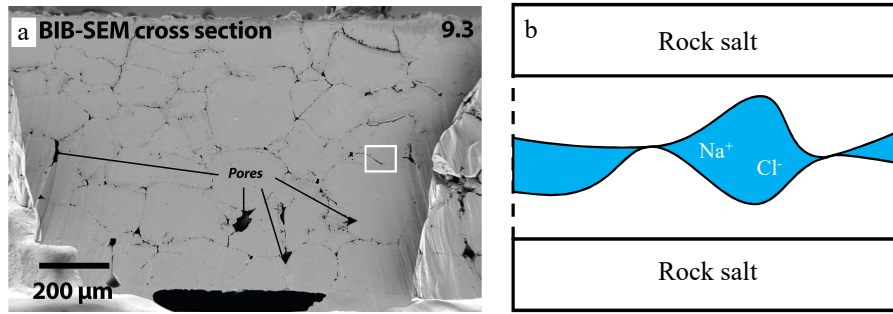


Fig. 7.1. Microstructure of rock salt with pores and intergranular brine: (a) SEM image [Desbois et al., 2012c]; (b) schematic illustration.

induced solution precipitation, the crack tip curvature also reduces the chemical potential of the solid phase, providing an additional driving force for precipitation [Houben et al., 2013, Koelemeijer et al., 2012, van Noort et al., 2008]. The crack healing rate is controlled by solute diffusion instead of solution and precipitation [Houben et al., 2013], and the diffusivity within the thin film is measured [Koelemeijer et al., 2012]. Recently, a detailed observation of the brine distribution in grain boundaries has revealed that the healed grain boundaries provide a threshold for the solution precipitation process [Desbois et al., 2012c].

The deformation mechanism within the polycrystalline rock is so complicated that it is difficult for phenomenological based models to quantitatively describe dislocation creep, solution precipitation, microcrack dilatancy, and crack healing simultaneously. Therefore, physics-based models and explicit representation of polycrystalline structure are necessary for a fundamental understanding and quantitative prediction of the deformation and permeability evolution within rock salt.

7.1.2 Fracture and healing in rock salt: material modeling

Numerous researches have been conducted toward a unified field formulation to predict multi-physical behaviors of rock salt and other crystalline rock. For instance, a self-consistent homogenization method is used such that the crystal plasticity simulations were upscaled to predict the anisotropic plastic deformation of polycrystalline rock salt [Wenk et al., 1989, Lebensohn et al., 2003]. A general kinematic framework was formulated to describe the diffusion and convection of brine and air inclusions within polycrystalline rock salt [Olivella et al., 1994b]. Kruzhanov and Stöckhert [1998] modeled the solution-precipitation creep by introducing an inelastic displacement field that maximizes the creep deformation potential defined along the grain boundary. Front tracking technique and adaptive mesh technique are combined at the expense of numerical accuracy and efficiency to simulate grain boundary migration and diffusion in a Lagrangian framework by Bower and Wininger [2004].

Predicting crack initiation, crack propagation, and crack healing within damaged rock salt through numerical simulations has received significant attention due to the ever-increasing demand for evaluating the permeability of crushed salt after re-consolidation. Crack healing has traditionally been considered within the framework of continuum damage mechanics [Chan et al., 1996, 2000]. A phenomenological model is proposed to describe the competitive effect between mechanical damage and healing in the excavation damaged zone (EDZ) through continuum damage mechanics [Hou, 2003]. More recently,

this model is extended to capture the thermo-mechanical crack healing model to simulate the competitions between mechanical damage and crack healing in rock salt [Zhu and Arson, 2015b]. Furthermore, the effect of solution-precipitation on the micropore healing process within polycrystalline rock salt is simulated by a diffusion-based homogenization model in Shen and Arson [2019].

While those phenomenological models can replicate some aspects of the constitutive behaviors of rock salt, the number of material parameters required for curve-fitting is large and those material parameters often lack significant physical underpinnings and hence over-fitting may occur when those models are used for blind predictions [Wang et al., 2016, Liu et al., 2016b, Wang et al., 2019]. To circumvent this situation, multiscale DEM-FEM or FEM² approaches are sometimes used to upscale the simplified microscopic behaviors from the grain scale to the macroscopic scale [Pouya et al., 2016, Wei and Anand, 2008, Kang et al., 2015, Tjioe and Borja, 2016, Liu et al., 2019]. However, this upscaling procedure is only meaningful if the interplay of the coupled mechanisms such as microcracking dilatancy, solution precipitation creeping, crack healing, crystal plasticity, and heat transfer can be sufficiently replicated at the grain scale. The objective of this paper is to provide this important theoretical framework and an FFT solver that *explicitly* captures these multi-physical coupling mechanisms, and as a result, enables us to explain, understand and upscaling these responses for macroscopic predictions.

7.1.3 Outlines, major contributions and notations

In this paper, a thermodynamic framework is proposed which explicitly incorporates crystal plasticity, solution-precipitation creep, strongly anisotropic cracking, and crack healing starting from the previous work [Na and Sun, 2018]. The ductile plastic deformation and dislocation creeping of each crystal grain under different temperature and confining pressure is captured via crystal plasticity, whereas a high-order phase-field model is introduced to predict the anisotropic crack propagation in polycrystalline rock salt with a non-convex cleavage energy determined by the preferential fracture plane of rock salt. Meanwhile, the solution-precipitation creep is replicated by simulating the transport of chemical species along the grain boundary. The chemical concentration then induces deformation. This approach is inspired by recent investigations that capture both Herring and Coble creeps at high homologous temperature [Garikipati et al., 2001, Villani et al., 2014, 2015]. In this work, our new contribution is that we incorporate this diffusion problem into a unified framework to predict how the solution-precipitation creep affects both the fracture and the healing process. Since the healing often starts at the crack tip, we introduce a measure of the curvature of the phase-field into our derivation of chemical potential such that the healing mechanism is depending on the surface areas. [Houben et al., 2013]. Finally, considering the high computational cost of the coupled equations and the global C_1 continuity requirement of the strongly anisotropic phase-field model, an FFT-based method is adopted to solve the coupled equations in an operator split manner.

This paper will proceed as follows. Section 7.2 discusses the balance law for mass, linear momentum, microforce, and energy, respectively. Section 7.3 presents the constitutive relation for small strain crystal plasticity, solution precipitation, crack healing, and high-order phase-field. In Section 7.4, three examples are presented to demonstrate the capability of the proposed framework to represent the multiphysics behavior of rock salt in mesoscale. Section 7.5 summarizes the major results and concluding remarks.

As for notations and symbols, bold-faced letters denote tensors (including vectors which are rank-one tensors); the symbol $\cdot$ denotes a single contraction of adjacent indices

of two tensors (e.g. $\mathbf{a} \cdot \mathbf{b} = a_i b_i$ or $\mathbf{c} \cdot \mathbf{d} = c_{ij} d_{jk}$); the symbol ‘:’ denotes a double contraction of adjacent indices of tensor of rank two or higher (e.g. $\mathbf{C} : \boldsymbol{\varepsilon}^e = C_{ijkl} \varepsilon_{kl}^e$); the symbol ‘::’ denotes a fourth contraction of adjacent indices of tensor of rank four or higher (e.g. $\mathbf{C} :: \mathbf{D} = C_{ijkl} D_{ijkl}$); the symbol ‘ $\otimes$ ’ denotes a juxtaposition of two vectors (e.g. $\mathbf{a} \otimes \mathbf{b} = a_i b_j$) or two symmetric second order tensors (e.g. $(\boldsymbol{\alpha} \otimes \boldsymbol{\beta})_{ijkl} = \alpha_{ij} \beta_{kl}$); the symbol ‘ $\mathcal{F}$ ’ and ‘ $\mathcal{F}^{-1}$ ’ represent forward and backward Fourier transformation, respectively. Materials are assumed to be simple cubic symmetry throughout this paper unless specified.

7.2 Balance laws

In this section, the governing equations of the thermal-mechanical-chemical-phase field framework are introduced. These equations constitute the boundary value problem that replicates the multiphysical material behaviors of polycrystalline rock salt in the geometrically linear regime. We first introduce an interfacial indicator function that defines the location of the regularized grain boundary and crack region. Then the balance principle for mass, linear momentum, and microforce (material force) are derived by extending the model in [Na and Sun, 2018]. Finally, the energy balance equation and dissipation inequality are derived for the multiphysics problem.

7.2.1 Definition of interface region and indicator function

Cryogenic experiments have revealed the island–channel structure of the rock salt grain boundary region, where the saturated brine exists and solution–precipitation occurs [Desbois et al., 2012c]. The solution–precipitation in return will also influence the porosity of the grain boundary, and crack healing is enabled by the thin brine films within the crack region [Renard et al., 2004]. However, the morphology of the grain boundary region and the crack region is highly irregular and therefore difficult to capture via conformal meshes or embedded discontinuities [Wang and Sun, 2019, Liu and Sun, 2019]. To overcome this problem, we introduce regularized interfacial regions represented by phase fields to capture the multiphysical coupling process occurring along the grain boundary and the crack [Garikipati et al., 2001, Sharma et al., 2018]. First, the indicator for the grain boundary region is introduced as

$$d_{\text{GB}}(\mathbf{X}) = \begin{cases} 0, & \mathbf{X} \text{ in the lattice region} \\ 1, & \mathbf{X} \text{ in the grain boundary region,} \end{cases} \quad (7.1)$$

where $\mathbf{X}$ denotes the coordinate of the material point. A similar concept is also proposed in van Noort et al. [2008] to derive a theoretical model for solution precipitation within rock salt. The indicator for the interfacial region (including grain boundary region and crack region) is defined as:

$$d_i(d_c, d_{\text{GB}}) = 1 - [1 - d_c^2(\mathbf{X})] [1 - d_{\text{GB}}^2(\mathbf{X})], \quad (7.2)$$

where $d_c(\mathbf{X}) \in [0, 1]$ is the phase field of the crack region.

Consider a periodic domain Ω with n_{sd} the spatial dimensions, this domain Ω can be divided into the bulk region Ω_{bulk} and the interfacial region Ω_i through the interfacial indicator:

$$\Omega_{\text{bulk}} = \{\mathbf{X} | d_i(\mathbf{X}) < \text{tol}, \mathbf{X} \in \Omega\}, \quad \Omega_i = \{\mathbf{X} | d_i(\mathbf{X}) \geq \text{tol}, \mathbf{X} \in \Omega\}, \quad (7.3)$$

where $\text{tol} \in (0, 1)$ is a small enough positive number. The interfacial region contains the physical interface between the solid and liquid phase, and also involves plastic deformation at the solid-solid contact region [van Noort et al., 2008].

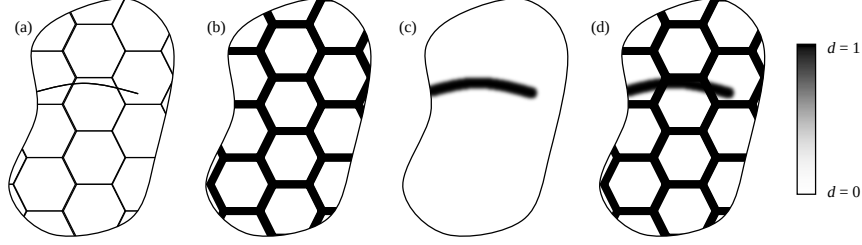


Fig. 7.2. A schematic representation of (a) A solid body Ω with crack discontinuity Γ_c and grain boundary discontinuity Γ_{GB} ; (b) A regularized grain boundary region Ω_{GB} by grain boundary indicator d_{GB} ; (c) A regularized crack region Ω_c by fracture phase field d_c ; (d) A regularized interfacial region $\Omega_i = \Omega_{GB} \cup \Omega_c$ by interfacial indicator d_i .

7.2.2 Balance of mass

Let ρ_s denotes the intrinsic density of solid halite, ρ_l denotes the intrinsic density of brine, c denotes the intrinsic molar concentration of brine, and ϕ represents the local porosity of the interfacial domain. According to the poromechanics theory [Choo and Sun, 2018b], the mass balance equations can be written as:

$$\begin{cases} \frac{D}{Dt}[(1-\phi)\rho_s] + (1-\phi)\rho_s \nabla \cdot \mathbf{v} = -\phi r_c M, & \text{in } \Omega_i \\ \frac{D}{Dt}(\phi\rho_l) + \phi\rho_l \nabla \cdot \mathbf{v} + \nabla \cdot [\phi\rho_l(\mathbf{v}_l - \mathbf{v})] = \phi r_c M, & \text{in } \Omega_i \\ \frac{D}{Dt}(\phi c) + \phi c \nabla \cdot \mathbf{v} + \nabla \cdot [\phi\rho_l(\mathbf{v}_l - \mathbf{v}) + \mathbf{J}] = \phi r_c, & \text{in } \Omega_i. \end{cases} \quad (7.4)$$

Here, $\mathbf{v}$ is the velocity of the solid phase, $\mathbf{v}_l$ is the velocity of the liquid phase, $\mathbf{J}$ is the diffusion flux, r_c is the molar dissolution rate, and M is the molar mass of halite.

We made several assumptions to simplify the mathematical model. First, we assume that the bulk porosity ϕ is constant in time scale and homogeneous in spatial scale. Furthermore, the material parameters are measured from the effective medium which may contain voids. However, the diffusivity of the crystalline material is mainly attributed to the interconnected interface domain, i.e. the grain boundaries and the cracks, which can be identified from the interfacial indicator d_i [Wang and Sun, 2018]. Second, it is assumed that the fluid infiltration inside the bulk material is negligible such that the fluid velocity $\mathbf{v}_l$ equals to the solid velocity $\mathbf{v}$. As a result, the mass balance equation (7.4) can be simplified as:

$$\begin{cases} \dot{\epsilon}^c = -r_c \Omega_v \mathbf{n} \otimes \mathbf{n}, & \text{in } \Omega_i \\ \dot{\rho}_l = r_c M, & \text{in } \Omega_i \\ \dot{c} + \nabla \cdot \mathbf{J} = r_c, & \text{in } \Omega_i. \end{cases} \quad (7.5)$$

In this equation, $\dot{\epsilon}^c$ is the precipitation creep rate, Ω_v is the molar volume of solid rock salt, and $\mathbf{n}$ is the normal direction of the interfacial region. The diffusion creep rate $\dot{\epsilon}^c$ is also used to model the Coble creep deformation within the grain boundary region [Garikipati et al., 2001, Villani et al., 2014, 2015]. The mass flux $\mathbf{J}$ is a function of the chemical potential gradient. The source term r_c is proportional to the difference between the equilibrium concentration c_{eq} and the current concentration c , as illustrated in Section 7.3.2.

7.2.3 Balance of linear momentum and microforce

To complete the field theory, the balance law of linear momentum and microforce (material force) are briefly summarized. While the balance of linear momentum constrains the stress field, the microforce balance provides an additional governing equations for the degradation evolution within the crystal grains and grain boundaries (cf. [Na and Sun \[2018\]](#)). Moreover, microforce corresponding to the second-order gradient of phase-field is introduced to incorporate the strongly anisotropic fracture observed in rock salt.

First, it is assumed throughout this paper that all the governing equations and corresponding physical quantities are defined in a cubic shape representative volume element (RVE) with periodic boundary conditions, and no body force or inertia force are considered. As a result, the balance of linear momentum requires that the divergence of the Cauchy stress σ vanishes everywhere:

$$\nabla \cdot \sigma = 0. \quad (7.6)$$

The Cauchy stress σ is, under small strain assumption, work conjugate with the strain rate $\dot{\epsilon}$.

The strain energy equivalence principle is adopted here to simplify the coupling relationship between the phase field and the Cauchy stress, where we assume that a fictitious undamaged body exists with possibly unbalanced linear momentum corresponding to the damaged counterpart. The total stress σ within the damaged body and the effective stress $\hat{\sigma}$ within the undamaged body are assumed to be co-axial and can be related by introducing a scalar degradation function $g(d_c)$, defined as

$$\sigma = g(d_c)\hat{\sigma}, \quad g(d_c) = (1 - k)(1 - d_c)^2 + k \quad (7.7)$$

where $0 < k \ll 1$ represents the residual portion of stiffness within the damaged region to retain the well-posedness of the problem.

The existence of microforce power conjugate to the phase-field is postulated together with the balance law for the microforce, such that the phase-field theory can be incorporated into the coupling equations with thermodynamic consistency [[Gurtin, 1996c](#)]. Supposing that π , ξ , and η are the microforces power conjugate to the phase field d_c and its first ∇d_c and second order gradient $\nabla^2 d_c$ respectively, then the balance law of the microforces requires that

$$\pi - \nabla \cdot \xi + (\nabla \otimes \nabla) : \zeta = 0. \quad (7.8)$$

It is also assumed that the body force of the microforces vanishes.

The anisotropic microfracture can be incorporated into the crystal plasticity model to predict the brittle-ductile transition in rocks [[Tjioe and Borja, 2015](#)]. Here, we try to model the microcrack propagation explicitly in order to provide a deep understanding of the multi-physics material process within rock salt. Note that in our previous work [[Na and Sun, 2018](#)], multi-phase-field is adopted for strongly anisotropic fracture. While this approach is feasible to replicate strongly anisotropic fracture, the introduction of multiple phase fields may significantly increase the computational resources required to solve the problems numerically. To improve efficiency, we have adopted the higher-order phase field fracture model (cf. [Li et al. \[2015\]](#), [Li and Maurini \[2019\]](#) where only one phase field is required to replicate the strong anisotropy. The global continuity of the interpolated phase field required to resolve the higher-order terms is fulfilled by the trigonometric function basis of the FFT model [[Ma and Sun, 2020](#)].

7.2.4 Balance of energy and dissipation inequality

In this section, the thermodynamic laws are presented in terms of the mechanical work, structural heating, crack surface energy, and chemical potential. Our starting point is the derivation from [Na and Sun \[2018\]](#) with the following improvements and modifications: (1) High-order phase-field is introduced instead of multi-phase-field to improve computational efficiency and reduce the number of material parameters; (2) The dissipation due to fluid diffusion and those due to dissolution and precipitation are incorporated in the formulation to model the solution-precipitation creeping and crack healing.

The energy balance law can be written in terms of the internal energy e , which is a function of the elastic strain ϵ^e , internal variables $\tilde{\tau}$, phase field d_c , solute concentration c , and entropy η . The first law of thermodynamics requires that the internal energy changing rate $\dot{e}$ equals to the total energy input rate:

$$\dot{e} = \sigma : \dot{\epsilon} - \nabla \cdot \mathbf{q} + r_\theta + \pi \dot{d}_c + \xi : \nabla \dot{d}_c + \zeta : (\nabla \otimes \nabla \dot{d}_c) + \mu \dot{c}, \quad (7.9)$$

where $\mathbf{q}$ is the thermal flux, r_θ is the heat source.

The second law of thermodynamics can be written in the form of the Clausius–Duhem inequality:

$$\mathcal{D} = \mathcal{D}_{\text{int}} + \mathcal{D}_{\text{diff}} + \mathcal{D}_{\text{tran}} \geq 0, \quad (7.10)$$

where $\mathcal{D}$ is the total dissipation, $\mathcal{D}_{\text{int}}$ is the internal dissipation due to mechanical work and thermal diffusion, $\mathcal{D}_{\text{diff}}$ is the diffusion dissipation, and $\mathcal{D}_{\text{tran}}$ represents the dissipation due to dissolution and precipitation.

A sufficient condition for the total dissipation $\mathcal{D}$ to be non-negative is that all the dissipation components are non-negative individually. The diffusion and solution-precipitation dissipation are given by

$$\begin{cases} \mathcal{D}_{\text{diff}} = -\nabla \mu \cdot \mathbf{J} \geq 0 \\ \mathcal{D}_{\text{tran}} = \sigma : \dot{\epsilon}^c + r_c (\mu_{se} - \mu) \geq 0. \end{cases} \quad (7.11)$$

Here, μ is the chemical potential of the interfacial brine, and μ_{se} is the equilibrium potential of the solute in the interfacial brine which equals to the potential of the solid phase. These two inequality conditions pose restrictions to the admissible mass diffusion and solution precipitation constitutive relations as shown in Section 7.3.2. Note that the second term in the transformation dissipation $\mathcal{D}_{\text{tran}}$ is always non-negative, but the first term can be either positive or negative. Therefore, the dissipation inequality poses restrictions to the admissible states of the constitutive relation.

The internal dissipation $\mathcal{D}_{\text{int}}$ is usually written as

$$\mathcal{D}_{\text{int}} = \mathcal{D}_{\text{loc}} + \mathcal{D}_{\text{con}} = \dot{\eta} + \nabla \cdot \left(\frac{\mathbf{q}}{\theta} \right) - \frac{r_\theta}{\theta} \geq 0, \quad (7.12)$$

where η is the specific entropy per unit volume and θ is the absolute temperature. Equation (7.12) is usually referred to as the Clausius–Planck inequality, which requires that the real entropy increment should surpass the entropy input from thermal diffusion and heat source. In order to ensure that the internal dissipation $\mathcal{D}_{\text{int}}$ is non-negative, the mechanical dissipation ($\mathcal{D}_{\text{loc}}$) inequality and thermal conduction dissipation ($\mathcal{D}_{\text{con}}$) inequality are usually satisfied separately [[Truesdell and Noll, 2004](#)]:

$$\begin{cases} \mathcal{D}_{\text{loc}} = \dot{\eta} - \frac{r_\theta}{\theta} + \frac{1}{\theta} \nabla \cdot \mathbf{q} \geq 0 \\ \mathcal{D}_{\text{con}} = -\frac{1}{r_\theta \theta^2} \mathbf{q} \cdot \nabla \theta \geq 0. \end{cases} \quad (7.13)$$

A specific form of free energy ψ is proposed such that the mechanical dissipation $\mathcal{D}_{\text{loc}}$ is always non-negative. The thermal conduction dissipation $\mathcal{D}_{\text{con}}$ is ensured by the Fourier's law and a positive thermal conductivity κ . The Fourier's law is used to correlate the heat flux $\mathbf{q}$ and the temperature θ :

$$\mathbf{q} = -\kappa \nabla \theta \quad (7.14)$$

where the scalar variable κ is the isotropic thermal conductivity.

Since the internal energy e is a convex function of the entropy η , the thermodynamics laws can be described using the Helmholtz free energy ψ through Legendre transformation without losing any information:

$$\psi = e - \theta \eta. \quad (7.15)$$

Substituting equation (7.15) into the dissipation inequality (7.13) and considering the specific form of the Helmholtz free energy (7.20), the dissipation inequality of the local entropy production $\mathcal{D}_{\text{loc}}$ can be written as:

$$\begin{aligned} \mathcal{D}_{\text{loc}} = & \left(\sigma - \frac{\partial \psi}{\partial \epsilon^e} \right) : \dot{\epsilon}^e + \left(\sigma : \dot{\epsilon}^p - \frac{\partial \psi}{\partial \tilde{\tau}} \dot{\tilde{\tau}} \right) + \left(\mu - \frac{\partial \psi}{\partial c} \right) \dot{c} + \left(\pi - \frac{\partial \psi}{\partial d_c} \right) \dot{d}_c \\ & + \left(\xi - \frac{\partial \psi}{\partial \nabla d_c} \right) \cdot \nabla \dot{d}_c + \left(\zeta - \frac{\partial \psi}{\partial (\nabla \otimes \nabla d_c)} \right) : (\nabla \otimes \nabla \dot{d}_c) - \left(\eta + \frac{\partial \psi}{\partial \theta} \right) \dot{\theta} \geq 0 \end{aligned} \quad (7.16)$$

Note that the precipitation induced mechanical dissipation $\sigma : \dot{\epsilon}^c$ is included in the solution precipitation dissipation $\mathcal{D}_{\text{diff}}$ in Equation (7.11). According to the Coleman–Noll argument, the arbitrary changing rate of the state variables $\dot{\epsilon}^e$, $\dot{\theta}$, and $\dot{c}$ requires that:

$$\sigma = \frac{\partial \psi}{\partial \epsilon^e}, \quad \eta = -\frac{\partial \psi}{\partial \theta}, \quad \mu = \frac{\partial \psi}{\partial c} \quad (7.17)$$

Furthermore, the arbitrariness of the phase field $\dot{d}_c$ and its gradient $\nabla \dot{d}_c$ and $(\nabla \otimes \nabla \dot{d}_c)$ requires that:

$$\pi = \frac{\partial \psi}{\partial d_c}, \quad \xi = \frac{\partial \psi}{\partial \nabla d_c}, \quad \zeta = \frac{\partial \psi}{\partial (\nabla \otimes \nabla d_c)}. \quad (7.18)$$

Therefore, the dissipation inequality requires that the plastic dissipation be positive for any admissible set of state variables:

$$\mathcal{D}_{\text{loc}} = \sigma : \dot{\epsilon}^p - \frac{\partial \psi}{\partial \tilde{\tau}} \dot{\tilde{\tau}} = \chi \sigma : \dot{\epsilon}^p \geq 0, \quad (7.19)$$

where $\tilde{\tau}$ is the internal hardening variable, and the Taylor–Quinney coefficient χ is utilized to simplify the plastic dissipation.

The following explicit expression is adopted for the total free energy ψ , which is split into the elastic free energy ψ^e , the plastic stored free energy ψ^p , the crack surface energy ψ^d , the chemical free energy ψ^c , and the thermal contribution of the stored energy ψ^θ :

$$\psi = \psi^e(\epsilon^e, \theta, d_c) + \psi^p(\tilde{\tau}, d_c) + \psi^d(d_c, \nabla d_c, \nabla \otimes \nabla d_c) + \psi^c(c, \theta) + \psi^\theta(\theta). \quad (7.20)$$

A specific form of free energy is proposed based on our previous work [Na and Sun \[2018\]](#) with the following modifications: (1) Chemical free energy ψ^c is included for solute diffusion within interfacial brine; (2) High-order phase-field cleavage energy is used

instead of multi-phase-field cleavage energy; (3) An initial phase-field penalizing term is included to avoid the sharp material contrast between the crack region and the intact region. The five parts of the Helmholtz free energy ψ take the following form:

$$\begin{cases} \psi^e = g(d_c)w_+^e(\varepsilon^e, \theta) + w_-^e(\varepsilon^e, \theta) \\ \psi^p = g(d_c)\hat{\psi}^p(\tilde{\tau}) \\ \psi^d = G_c \left[\frac{1}{2l_0} d_c^2 + \frac{l_0}{4} \nabla d_c \cdot \nabla d_c + \frac{l_0^3}{32} (\nabla \otimes \nabla d_c) : \mathbb{A} : (\nabla \otimes \nabla d_c) + \frac{1}{2} \beta_{ipf} (d_c - 1)^2 \right] \\ \psi^c = \mu_0 c + R\theta c \left(\ln \frac{c}{c_0} - 1 \right) \\ \psi^\theta = c_v [(\theta - \theta_0) - \theta \ln(\theta/\theta_0)] \end{cases} \quad (7.21)$$

Here, G_c is the cleavage energy per unit mass, l_0 is the character length, β_{ipf} is the penalty coefficient for the initial phase field, θ_0 is the initial temperature, and c_v is the specific heat coefficient per unit mass. Note that in the mesoscale Coble creep models, either statistical thermodynamics based chemical free energy density ψ^c [Garikipati et al., 2001] or classical lattice-void free energy density ψ^c [Villani et al., 2014, 2015] is utilized. For the interfacial brine, the chemical free energy density ψ^c for ideal fluid is used, although other choices remain applicable.

To further simplify the balance of energy equation, we assume that the entropy change due to chemical diffusion and crack propagation is negligible compared with plastic dissipation, such that:

$$\theta \frac{\partial^2 \psi}{\partial \theta \partial c} \dot{c} = R\theta \ln \frac{c}{c_0} \dot{c} \ll \mathcal{D}_{loc}, \quad \theta \frac{\partial^2 \psi}{\partial \theta \partial d_c} \dot{d}_c = -3\alpha\theta K g'(d_c) \langle \varepsilon_v^e \rangle_+ \dot{d}_c \ll \mathcal{D}_{loc}. \quad (7.22)$$

Substitute the specific form of free energy ψ (7.20) into the energy balance equation (7.9), and considering the Coleman–Noll arguments (7.17) and (7.18) and the above equation, the energy balance equation can be written as:

$$c_v \dot{\theta} = \theta \frac{\partial^2 \psi}{\partial \theta \partial \varepsilon^e} : \dot{\varepsilon}^e + \sigma : \dot{\varepsilon}^p - \frac{\partial \psi}{\partial \tilde{\tau}} \dot{\tilde{\tau}} - \nabla \cdot \mathbf{q} + r_\theta = -3\alpha K \theta \mathbf{I} : \dot{\varepsilon}^e + (1 - \chi) \sigma : \dot{\varepsilon}^p - \nabla \cdot \mathbf{q} + r_\theta. \quad (7.23)$$

Considering that the phase field d_c is non-conserved and brittle fracture (instead of creep damage) is rate-independent, Ginzburg-Landau type phase-field equations are derived based on the specific form of free energy ψ (7.20). Substitute the Coleman–Noll relation (7.18) into the microscopic force balance equation (7.8), and assuming that the fourth order anisotropic tensor $\mathbb{A}$ is piecewise constant:

$$\frac{G_c}{l_0} d_c - \frac{G_c l_0}{2} \nabla \cdot \nabla d_c + \frac{G_c l_0^3}{16} \mathbb{A} :: \nabla^4 d_c + \beta_{ipf} (d_c - 1) = 2(1 - d_c) \mathcal{H}, \quad d_c \in H_{\#}^4. \quad (7.24)$$

Here, the phase field driving force $\mathcal{H}$ is a function of the fictitious stored strain energy without degradation:

$$\mathcal{H} = \max_{\tau \in [0, t]} [w_+^e + \hat{\psi}^p(\tilde{\tau})] = \max_{\tau \in [0, t]} [w_+^e + (1 - \chi) \langle w^p - w_0^p \rangle], \quad (7.25)$$

where the positive part of the elastic stored energy w_+^e and the accumulated plastic work w^p will be introduced in the following section. The plastic deformation threshold w_0^p is introduced to control the contribution from accumulated plastic work w^p to ductile fracture. Note that the phase-field driving force $\mathcal{H}$ in equation (7.25) is monotonically increasing, which will be modified in Section 7.3.3 to enable diffusion controlled crack healing.

7.3 Constitutive relations

This section introduces the constitutive relations used in this paper. First, a small strain crystal plasticity model is revisited with a Voce type hardening relation. Then, a diffusion model is introduced which allows the solution, diffusion, and precipitation of NaCl solute along the grain boundary region and the crack region. Chemical potential depending on pressure and solid/liquid interface curvature is considered which enables pressure gradient driven and crack tip driven solution and precipitation. Finally, a fourth-order anisotropic tensor representing the preferential cleavage direction is presented for the high-order phase-field model, together with a modified phase-field driving force to enable diffusion-controlled crack healing.

7.3.1 Small strain crystal plasticity

The current small strain crystal plasticity model is re-formulated based on the finite strain counterpart [Messner et al., 2015, Ma and Truster, 2019]. Linear elasticity is assumed for the elastic response:

$$\hat{\sigma} = \hat{C} : \varepsilon^e, \quad \varepsilon^e = \varepsilon - \varepsilon^p - \varepsilon^c \quad (7.26)$$

where σ is the Cauchy stress, ε is the total strain rate, ε^θ is the thermal expansion, ε^c is the chemical deformation, and C is the 4th order elastic stiffness tensor. The chemical deformation is an explicit function of the solution-precipitation rate r_c , as defined in equation (7.5).

In polycrystalline material, the plastic strain is achieved by dislocation slide on each slip system. Let $\mathbf{n}^{(s)}$ and $\mathbf{b}^{(s)}$ represents the normal and slip direction of the (s) th slip system. Then the total plastic strain is the tensorial summation of the shear strain on each slip system:

$$\dot{\varepsilon}^p = \sum_{s=1}^{n_{slip}} \dot{\gamma}^{(s)} \mathbf{m}^{(s)}, \quad \mathbf{m}^{(s)} = \text{sym} \left(\mathbf{b}^{(s)} \otimes \mathbf{n}^{(s)} \right). \quad (7.27)$$

For small strain problem, the anti-symmetric part of the slip system does not contribute to the plastic strain.

In this investigation, the Voce model is adopted as the constitutive relation considering that rock salt is strain rate sensitive. Note that the Voce model is temperature independent, and temperature dependence can be introduced by replacing the Voce model by other crystal plasticity models such as the mechanical threshold (MTS) model. The relationship between the shear strain rate of the (s) th slip system $\dot{\gamma}^{(s)}$ and the resolved shear stress $\tau^{(s)}$ is assumed to follow the power law:

$$\dot{\gamma}^{(s)} = \frac{\dot{\gamma}_0}{\tilde{\tau}} \left| \frac{\tau^{(s)}}{\tilde{\tau}} \right|^{n-1} \tau^{(s)}. \quad (7.28)$$

Here, $\dot{\gamma}_0$ is the reference slip rate, and $\tilde{\tau}$ is the isotropic hardening variable. In this model, Taylor hardening is assumed where all slip systems contribute equally to the hardening variable $\tilde{\tau}$. The slip system resistance $\tilde{\tau}$ is decomposed into the intrinsic (yield) resistance τ_y and the extrinsic (hardening) resistance τ_w

$$\tilde{\tau} = \tau_y + \tau_w. \quad (7.29)$$

The extrinsic resistance τ_w evolves as a function of the slip system activity $\dot{\gamma}^{(s)}$

$$\dot{\tau}_w = h_0 \left(1 - \frac{\tau_w}{\tau_v} \right)^m \sum_{s=1}^{n_{slip}} \left| \dot{\gamma}^{(s)} \right| \quad (7.30)$$

where θ_0 is the initial hardening rate, and the work hardening saturation strength τ_v sets the upper bound of τ_w . The exponents m and n are separate parameters. Note that the hardening variable $\tilde{\tau}$ is monotonically increasing, so the current crystal plasticity model is not suitable for creep loading. Dislocation creep deformation can be modeled by introducing a dislocation annihilation model [Barton et al., 2013].

The elastic stored energy and plastic stored energy are also defined, which serves as the phase-field driving force in equation (7.25). The positive part of the elastic strain energy w_+^e is slitted into the volumetric and deviatoric part to avoid crack propagation under volumetric compression:

$$\begin{cases} w_+^e = \frac{1}{2} K \langle \epsilon_v^e \rangle_+^2 + \mu (\epsilon_d^e : \epsilon_d^e) - 3\alpha K (\theta - \theta_0) \langle \epsilon_v^e \rangle_+ \\ w_-^e = \frac{1}{2} K \langle \epsilon_v^e \rangle_-^2 - 3\alpha K (\theta - \theta_0) \langle \epsilon_v^e \rangle_-, \end{cases} \quad (7.31)$$

where ϵ_v^e is the volumetric elastic strain, ϵ_d^e is the deviatoric elastic strain, and K and μ are Lamé constants. The accumulated plastic work w^p is also computed in each Newton iteration to incorporate the contribution of plastic strain on ductile fracture utilizing the Taylor-Quinney coefficient χ :

$$\dot{w}^p = \hat{\sigma} : \dot{\epsilon}^p, \quad \hat{\psi}^p(\tilde{\tau}) = (1 - \chi) \int \dot{w}^p dt \quad (7.32)$$

7.3.2 Constitutive relation for interfacial diffusion

This section provides the derivation of the constitutive relation for the diffusion problem. To capture the fully coupled chemical-mechanical effect, the constitutive relation for the chemical potential μ is derived from a corresponding Gibbs free energy [Garikipati et al., 2001]. We assume that the chemical potential of the solute in brine does not explicitly depend on stress. Hence, the chemical potential μ [unit: J mol⁻¹] for ideal solution is adopted here:

$$\mu = \mu_0 + R\theta \ln \frac{c}{c_0}, \quad \nabla \mu = \frac{R\theta}{c} \nabla c, \quad (7.33)$$

where μ_0 and c_0 are the reference chemical potential and reference concentration, respectively.

The diffusion of brine along grain boundaries and cracks contributes to the solution-precipitation creep and crack healing. It is assumed that the diffusion coefficient D_0 is independent of the grain boundary normal pressure, so that the diffusion equation remains linear. The flux of the salt solution J along the grain boundary is proportional to the gradient of the chemical potential μ , i.e.,

$$J = - \frac{d_i(d_c, d_{GB}) D_0 c}{R\theta} \nabla \mu = -d_i(d_c, d_{GB}) D_0 \nabla c \quad (7.34)$$

where D_0 [unit: mm² s⁻¹] is the diffusion coefficient of saturated salt solution. This diffusion coefficient D_0 is a positive scalar such that the diffusion dissipation $\mathcal{D}_{diff}$ is non-negative. The interface indicator $d_i(d_c, d_{GB})$ ensures that the diffusion flux outside the interfacial region Ω_i approximately vanishes [Sharma et al., 2018].

Assuming that the brine concentration is close to the halite equilibrium concentration c_{eq} , the solution-precipitation rate r_c is proportional to the difference between the equilibrium concentration c_{eq} and current concentration c based on the experimental observations on mineral solution-precipitation [Schott et al., 2009, Alkattan et al., 1997]:

$$r_c = d_i (d_c, d_{GB}) \alpha_s k_s (c_{eq} - c), \quad (7.35)$$

where α_s [unit: mm^{-1}] is a material coefficient which is inversely proportional to the grain boundary thickness, and k_s [unit: mm s^{-1}] is the solution-precipitation coefficient which denotes the speed of salt migrating across the solid/liquid interface. Note that the solution coefficient k_s is positive such that the second part of $\mathcal{D}_{\text{tran}} \geq 0$. In equation (7.35), the equilibrium concentration c_{eq} depends on the pressure, temperature, and solid/liquid interface curvature [Driesner and Heinrich, 2007]. If the current concentration c is lower than the equilibrium concentration c_{eq} , the solution rate is larger than the precipitation rate and the source term r_c is positive; otherwise, the source term r_c is negative. Note that the new crack region is assumed to be filled promptly with saturated salt solution.

Remark 4. The material coefficient α_s is introduced to fix the difference of the source term between Equation (7.35) and Alkattan et al. [1997]. In equation (7.35), the source term r_c has the unit $\text{mol m}^{-3} \text{s}^{-1}$, while in Alkattan et al. [1997] the source term r_c has the unit $\text{mol m}^{-2} \text{s}^{-1}$ representing the amount of salt migrating across the solid/liquid interface. In this paper, the material coefficient α_s [unit: mm^{-1}], which is inversely proportional to the grain boundary thickness, converts the surface source term to the volumetric source term by assuming that the mass migration across the interface region is evenly distributed across the regularized grain boundary region. This coefficient also includes the ratio between the actual grain boundary area and the homogenized grain boundary area.

Furthermore, the crack tip curvature can also influence the chemical potential of the solid phase and provide the driving force for diffusion-controlled crack healing. At the room temperature, the chemical potential of the solid phase μ_{se} is a function of the pressure p and principle curvature $1/r$ (only one component for 2D):

$$\mu_{se} = f + p\Omega_v + \frac{\Omega_v \gamma_{sl}}{r} = \mu_0 + 2R\theta \ln \frac{c_{eq}}{C_0}, \quad (7.36)$$

where f is the free energy of the solid phase under atmospheric pressure, Ω_v is the molecular volume of the solid, γ_{sl} is the interfacial energy between solid and liquid phase, and μ_0 and C_0 are the reference chemical potential and concentration. The crack tip radius is positive for convex a solid interface. With the expression in equation (7.33) for chemical potential, the equilibrium concentration c_{eq} is defined such that the chemical potential of the solute in brine equals to that of the adjacent solid-phase μ_{se} . Therefore, the equilibrium concentration (c_{eq}) at crack tip can be written as an explicit function of the pressure p and principle curvature $1/r$ (2D):

$$c_{eq} = c_{eq}^0 \exp \left(\frac{p\Omega_v}{2RT} + \frac{\gamma_{sl}\Omega_v}{2rRT} \right) \approx c_{eq}^0 \left(1 + \frac{p\Omega_v}{2RT} + \frac{\gamma_{sl}\Omega_v}{2rRT} \right), \quad (7.37)$$

where c_{eq}^0 is the equilibrium concentration at atmosphere pressure and room temperature with a straight solid-liquid interface.

When using the phase-field model to predict the fracture behavior, it is usually assumed that the crack tip is sharp and the crack tip radius is zero when the length-scale

parameter l_0 approaches zero. Therefore, it is impossible to compute the crack tip curvature numerically. To overcome this problem, a user-input crack tip radius is assigned to the crack tip region with the help of the Heaviside function $H(x)$ for 2-dimensional case:

$$r = \begin{cases} r_0, & l_0^3 \|(\nabla \otimes \nabla d_c - \nabla^2 d_c \mathbf{I}) \cdot \nabla d_c\| \geq \text{tol.} \\ \infty, & \text{otherwise} \end{cases} \quad (7.38)$$

where tol is the tolerance to differentiate the crack tip region from the crack region. Note that the second-order gradient of the phase-field is difficult to compute in finite element, since the gradient of the polynomial shape function is not continuous across the element edge. The effect of equation (7.38) in capturing the crack tip region is shown in Figure 7.3, where the phase field d_c is computed by the penalty term with $\mathcal{H} = 0$ in Equation (7.24). Figure 7.3 (a) and (b) show the phase field and crack tip indicator of a 'C' shape crack, while Figure 7.3 (c) and (d) show the phase field and crack tip indicator of two crossed cracks. The proposed criterion is effective in capturing the crack tip region even for a curved crack. Note that equation (7.38) can be only applied for high-order phase-field model. For second-order phase-field model, the second-order gradient has singular values, which will cause numerical issues when determining the crack tip region.

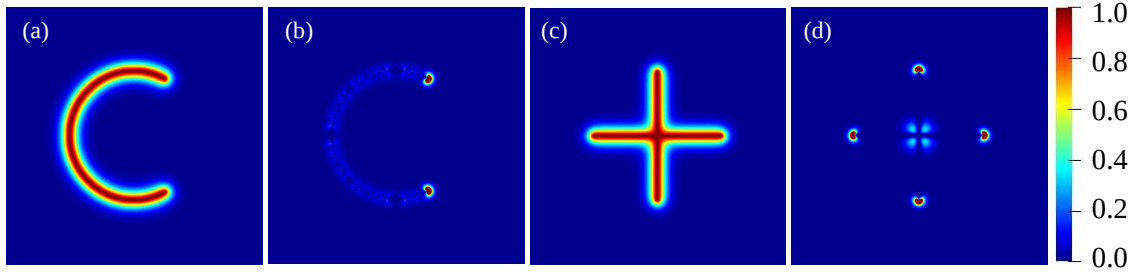


Fig. 7.3. Effectiveness of using equation (7.38) to detect the crack tip region. The crack region detection criteria takes the value $l_0^3 \|(\nabla \otimes \nabla d_c - \nabla^2 d_c \mathbf{I}) \cdot \nabla d_c\|$. (a) Initial phase field distribution of curved crack using high-order phase-field model; (b) Crack tip determination criteria of curved crack; (c) Initial phase field distribution of intersecting crack using high-order phase-field model; (d) Crack tip determination criteria of intersecting crack.

Combining equation (7.5), (7.34), and (7.35), the final form of the diffusion equation can be written as:

$$\dot{c} - D_0 \nabla \cdot (d_i \nabla c) = d_i (d_c, d_{GB}) \alpha_s k_s (c_{eq} - c), \quad (7.39)$$

where the equilibrium concentration c_{eq} depends on local pressure and solid-liquid interface curvature.

7.3.3 Constitutive relations for anisotropic phase field and crack healing

In this section, the anisotropic tensor $\mathbb{A}$ for the high-order phase field problem (7.24) is introduced. Furthermore, a crack propagation driving force $\mathcal{H}$ taking account of both cracking and healing is proposed.

In the high-order phase-field problem (7.24), the fourth-order anisotropic tensor $\mathbb{A}$ forms non-convex cleavage energy in the polar plot. For material with cubic symmetry,

the anisotropic tensor $\mathbb{A}$ adopts the general form [Teichtmeister et al., 2017b]:

$$\mathbb{A} = \mathbb{I} + \alpha_{apf}(\mathbf{A}_1 \otimes \mathbf{A}_1 + \mathbf{A}_2 \otimes \mathbf{A}_2) + \beta_{apf} \text{sym}(\mathbf{A}_1 \otimes \mathbf{A}_2), \quad (7.40)$$

where $\mathbb{I}$ is the fourth order identity tensor, α_{apf} and β_{apf} are material parameters penalizing the anisotropy, and $\mathbf{A}_1$ and $\mathbf{A}_2$ are second order anisotropic tensors determined by the preferential cleavage plane of rock salt. The phase-field problem reduces to an isotropic case if the coefficient α_{apf} and β_{apf} vanish. In order for the anisotropic tensor $\mathbb{A}$ to be positive definite, the following criteria shall be met:

$$\alpha_{apf} > -1, \quad |\beta_{apf}| < 2|1 + \alpha_{apf}| \quad (7.41)$$

Let $\mathbf{a}_1$ and $\mathbf{a}_2$ represent the normal directions of two perpendicular cleavage planes of single crystal rock salt which is determined by the initial orientation, then the second order anisotropic tensor $\mathbf{A}_1$ and $\mathbf{A}_2$ can be defined as:

$$\mathbf{A}_1 = \mathbf{a}_1 \otimes \mathbf{a}_1, \quad \mathbf{A}_2 = \mathbf{a}_2 \otimes \mathbf{a}_2. \quad (7.42)$$

In the typical phase-field based brittle fracture model, the driving force $\mathcal{H}$ is forced to be monotonically increasing by keeping its maximum historical value, since the crack healing process is not prevented by the thermodynamic laws. Herein, it is assumed that the healing process is activated when the total strain is volumetric compression by allowing the phase-field driving force to decrease. The phenomenological model for halite cleavage plane healing based on solution-precipitation kinetics is proposed in this paper, considering that the stiffness recovery rate becomes slower along with the healing process [Shen and Arson, 2019]:

$$\mathcal{H}_{n+1} = \max [\mathcal{H}_n - \alpha_h \mathcal{H}_n \langle -r_c \rangle V_m \Delta t, w_+^e + (1 - \chi) \langle w^p - w_0^p \rangle]. \quad (7.43)$$

Here, w_0^p is the reference plastic work controlling the contribution of accumulated plastic work w^p on phase field evolution, and α_h is a non-dimensional coefficient indicating the percentage of contribution from precipitation to stiffness recovery.

Note that the crack healing is also possible when the crack is opening [Koelmeijer et al., 2012]. In this case, the healing process is accomplished through solution precipitation in a thin water film coated on the crack wall. This process is much slower than the crack closure case when the crack region is filled with brine, and therefore is not considered in this model.

7.4 Numerical aspect on FFT-based method

The boundary value problem consists the following governing equations: balance of linear momentum equation (7.6), the energy balance equation (7.23), the phase field problem (7.24), and the diffusion equation (7.39). These coupled equations are solved in an operator-split manner by a collocation FFT-based solver to take advantage of its globally C^∞ continuous basis functions and more efficient computational cost.

The mechanical equation is solved using the matrix-free Newton-Krylov method [Zeman et al., 2010]. Balance of linear momentum (7.6) can be re-formulated in a periodic domain with the help of the Green's operator $\mathbb{G}$:

$$\begin{cases} \mathbb{G} * \boldsymbol{\sigma} = \mathbf{0} & \text{in spatial domain} \\ \mathcal{F}^{-1} [\hat{\mathbb{G}} : \mathcal{F}(\boldsymbol{\sigma})] = \mathbf{0} & \text{in frequency domain.} \end{cases} \quad (7.44)$$

Here, the operator $*$ denotes convolution, which can be computed conveniently in the frequency domain. The Green's operator G used in this paper is independent of the reference material, which projects an arbitrary strain field to its compatible part. The major idea of the Newton-Krylov method is to use an iterative linear solver to solve the linearized form of equation (7.44), but instead of assembling the stiffness matrix, the convolution operation is performed utilizing FFT. In this paper, the conjugate gradient solver is used [Ma and Truster, 2019, de Geus et al., 2017]. Compare with the traditional fixed-point scheme, accelerated scheme, and the augmented Lagrangian scheme, the Newton-Krylov method generally exhibits better numerical efficiency. However, a major trade-off is that the Newton-Krylov method fails to converge when the spatial domain contains jump conditions such as a sharp contract of material properties. This issue can be alleviated numerically by replacing the sharp material contract with regularized interfacial representation via implicit function and introduce a residual stiffness for a completely damaged zone. Note that due to the periodic nature of the trigonometric basis functions and the numerical efficiency of the FFT-based method, the FFT-based method is frequently used for homogenization and concurrent multiscale modeling [Kochmann et al., 2018]. The numerical efficiency of the microscale simulation within the multiscale modeling can be improved by reducing the number of basis functions in FFT-based method [Kochmann et al., 2019], or using discrete harmonics based homogenization [Barton et al., 2015].

The temperature field is updated in a semi-implicit fashion using the FFT-based method [Zhu et al., 1999]. To simplify the formulation, we assume that the thermal conductivity κ and specific heat C_v are homogeneous and temperature independent. By taking the strain energy dissipation from the last converged step, the temperature field update at step $n + 1$ can be performed in the frequency domain as:

$$\hat{\theta}_{n+1} = \frac{C_v \hat{\theta}_n + \hat{r}_\theta + \mathcal{F} [\chi \sigma_n : \dot{\epsilon}_n^p - 3\alpha K \theta_n I : \dot{\epsilon}_n^e]}{C_v + \kappa \mathbf{k} \cdot \mathbf{k} \Delta t} \quad (7.45)$$

where $\mathbf{k}$ is the frequency vector, Δt is the time step increment, and $\hat{\cdot}$ denotes quantities in the frequency domain.

The continuum form of the high-order phase-field (7.24) defined in the periodic RVE can also be solved by the FFT-based spectral method. The gradient operator and Laplacian operator can be conveniently computed in the frequency domain, and the Gibbs effect can be alleviated utilizing the finite-difference based frequency vector. Assuming that the anisotropic tensor $\mathbb{A}$, the cleavage energy G_c , and length scale l_0 are piecewise constant and periodic, the continuous linear equation (7.24) can be discretized as:

$$\left(2\mathcal{H} + \frac{G_c}{l_0} + \frac{G_c l_0}{2} \mathcal{F}^{-1} \mathbf{k} \cdot \mathbf{k} \mathcal{F} + \frac{G_c l_0^3}{16} \mathbb{A} :: \mathcal{F}^{-1} \mathbf{k} \otimes \mathbf{k} \otimes \mathbf{k} \otimes \mathbf{k} \mathcal{F} + \beta_{ipf} \right) d_c = 2\mathcal{H} + \beta_{ipf}. \quad (7.46)$$

This equation can be solved by generalized minimal residual (GMRES) method. A modified driving force $\mathcal{H}$ based on equation (7.43) instead of a typical monotonically increasing driving force (7.25) is used to allow diffusion-controlled crack healing. Also, the initial defect region is enforced by the penalty method to avoid sharp material contrast, which leads to deteriorated convergence behavior of the mechanical equation (7.44).

The diffusion equation (7.39) can be discretized by estimating the gradient operator in

the frequency domain:

$$\begin{cases} [1 + \alpha_s k_s \Delta t d_i - \bar{\lambda} \mathcal{F}^{-1}(ik) \cdot (ik) \mathcal{F} - \mathcal{F}^{-1}(ik) \cdot (\mathcal{F} \tilde{\lambda}(x) \mathcal{F}^{-1}(ik) \mathcal{F})] c_{n+1} = c_n + \alpha_s k_s \Delta t d_i c_{eq} \\ \lambda(x) = \bar{\lambda} + \tilde{\lambda}(x) = d_i D_0 \Delta t. \end{cases} \quad (7.47)$$

The discretized form is solved by the conjugate gradient (CG) method. The equilibrium concentration c_{eq} depends on the pressure field and phase-field of last converged time step. The diffusion equation is more involved to be solved by the FFT-based spectral method in two aspects. First, the material parameters are continuously varying making it impossible to update the concentration field semi-implicitly. Although the varying diffusivity can be split into a volume average part and a perturbation part [Zhu et al., 1999], a fully implicit scheme is adopted considering that the solution-diffusion-precipitation process requires a long time step [Sharma et al., 2018]. Second, the large diffusivity contrast between the interfacial region and the bulk region is highly heterogeneous and will introduce numerical issues. This problem can be alleviated by selecting a proper diffusivity residual for the bulk region.

The coupled thermo-chemo-mechanical-phase field equations are solved in a staggered iterative scheme. The mechanical problem is first solved within each time step, which involves two Newton iterations: one for average stress boundary condition, and the other for average strain boundary condition. Then, the thermal equation, the diffusion equation, and the phase-field equation are solved successively. The internal variables and the equilibrium concentration c_{eq} are updated at the end of each time step.

7.5 Examples

In this section, numerical examples are presented to demonstrate the capability of the proposed numerical framework in capturing the coupled physical process in rock salt. In particular, the interplay among including crystal plasticity, strongly anisotropic cracking, solution precipitation, and crack healing are, for the first time, replicated explicitly in numerical simulations. We first calibrate the material model for crystal plasticity with data available from the literature. Then we introduce physics-based material parameters for the phase-field fracture, thermal diffusion and chemical transport. Then, a polycrystalline RVE creep simulation with constant stress boundary condition is performed to illustrate the solution precipitation creep within the interfacial region. A polycrystalline RVE simulation with monotonically increasing loading is performed to capture the competition between intergranular and intragranular fracture in polycrystalline rock salt. Finally, a cyclic loading numerical example is used to demonstrate the capability of the proposed crack healing model in capturing the diffusion-controlled crack healing.

7.5.1 Simulation setup and material model calibrations

A salt crystal exhibits a face-centered-cubic (FCC) structure. Along with the typical $\{111\}\langle\bar{1}\bar{1}0\rangle$ slip systems, two other slip systems are also observed including $\{110\}\langle\bar{1}\bar{1}0\rangle$ and $\{100\}\langle\bar{1}10\rangle$. In this paper, only $\{111\}\langle\bar{1}\bar{1}0\rangle$ slip systems are considered, since it has the lowest critical resolved stress at room temperature [Carter and Heard, 1970].

Note that the $\{110\}\langle\bar{1}\bar{1}0\rangle$ slip systems alone do not provide sufficient dimensionality to span the second-order tensor in 3D space and hence there are some kinematic modes of plastic deformation cannot be triggered by these two slip systems.

Table 7.1. Material properties for crystal plasticity

Parameters	Description	Value	Unit	Reference
E	Elastic modulus	38.0	GPa	Na and Sun [2018]
ν	Poisson's ratio	0.25	-	Na and Sun [2018]
n	Rate sensitivity exponent	15.0	-	-
$\bar{\gamma}$	Rate normalization factor	1.0×10^{-10}	s^{-1}	-
τ_y	Initial yield stress	0.5	MPa	-
τ_v	Saturation stress	10.0	MPa	-
m	Hardening exponent	1.0	-	-
h_0	Initial hardening rate	30.0	MPa	-

In our case, five independent variables are required to describe an arbitrary plastic strain in the three-dimensional space, but the $\{110\}\langle 1\bar{1}0\rangle$ slip systems can only provide two. Similarly, two independent variables are required to describe an arbitrary plastic strain in 2-dimension, but the simplified slip systems can only provide one. But the influence is limited since the other two slip systems have 6 times larger critical resolved shear stress than $\{110\}\langle 1\bar{1}0\rangle$ [Wenk et al., 1989, Lebensohn et al., 2003].

The single crystal uniaxial compression experiment from Carter and Heard [1970] are used to calibrate the crystal plasticity parameters. The elastic constants of single crystal rock salt are from Carter and Norton [2007]. Note that although rock salt has a cubic symmetry crystal structure, its elastic anisotropy factor is almost one, so isotropic elastic constants are used. Since the Voce model (7.29) is independent of temperature, only room temperature stress-strain curves are used with three different strain rates. A single crystal RVE is loaded in the $[001]$ direction with uniaxial compression boundary conditions. The calibrated material parameters for crystal plasticity are shown in Table 7.1. The comparisons between the experiment and the simulation are shown in Figure 7.4.

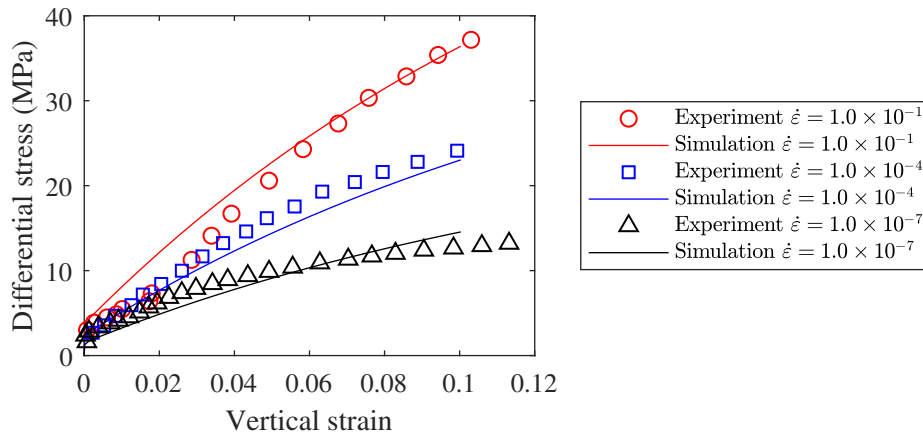


Fig. 7.4. Confined compression stress-strain response with different strain rates at room temperature [Carter and Heard, 1970].

The material parameters for the phase-field fracture model are shown in Table 7.2. The cleavage energy G_c is taken from our previous paper [Na and Sun, 2018], and other parameters are adjusted such that the strongly anisotropic cleavage behaviors observed experimentally are satisfied. Typical crack tip radius and crack opening angle are taken from Koelemeijer et al. [2012]. The preferential cleavage plane is $\{100\}$.

Table 7.2. Material properties for strongly anisotropic phase field

Parameters	Description	Value	Unit	Reference
G_c	Cleavage energy	1.15	J m ²	Na and Sun [2018]
l_0	Length scale	1.0×10^{-5}	m	Na and Sun [2018]
β_{ipf}	Initial phase field penalty	1000.0	-	-
α_{apf}	Anisotropy factor	1.2	-	-
r_0	Crack tip radius	0.5-5	μm	Koelemeijer et al. [2012]
β_{apf}	Anisotropy factor	1000.0	-	-

Table 7.3. Material properties for thermal problem

Parameters	Description	Value	Unit	Reference
α	Thermal expansion coefficient	11.0×10^{-6}	K ⁻¹	Na and Sun [2018]
c_v	Specific heat	2.0×10^6	J m ⁻³ K ⁻¹	Na and Sun [2018]
κ	Thermal conductivity	2.0	W m ⁻¹ K ⁻¹	Na and Sun [2018]

Table 7.4. Material properties for diffusion

Parameters	Description	Value	Unit	Reference
D_0	Diffusion coefficient	2.2×10^{-3}	m ² s ⁻¹	Alkattan et al. [1997]
k_s	Solution-precipitation coefficient	5.0×10^{-1}	m s ⁻¹	Alkattan et al. [1997]
γ_{sl}	Solid-liquid interfacial energy	0.129	J m ⁻²	Houben et al. [2013]
Ω_v	Molecular volume of solid NaCl	2.7×10^{-5}	m ³ mol ⁻¹	Koelemeijer et al. [2012]
R	Ideal gas constant	8.314	J mol ⁻¹ K ⁻¹	Koelemeijer et al. [2012]
α_s	Grain boundary thickness coefficient	1000.0	m ⁻¹	-
α_h	Crack healing coefficient	10.0	-	-
c_0^{eq}	Equilibrium concentration	5416.0	mol m ⁻³	Alkattan et al. [1997]

The thermal parameters [[Na and Sun, 2018](#)] and the diffusional parameters [[Alkattan et al., 1997](#)] are shown in Table 7.3 and 7.4, respectively. The crack healing coefficient α_h is manually adjusted to qualitatively meet the experimental crack healing rate [[Houben et al., 2013](#)], although the experiment is designed for opening crack adsorbed with thin brine film which makes quantitative comparison impossible. Note that simplifications are adopted that the interfacial region has the same thermal conductivity and thermal expansion as the grain bulk region. Also, it is assumed that the thickness of the brine film is much larger than several hundred nanometers such that the diffusion coefficient D_0 and solubility c_{eq} of the macroscale brine are representative.

7.5.2 Long-term creeping due to solution precipitation

In this numerical example, a series of creep simulations are performed to illustrate the solution precipitation creep observed in polycrystalline rock salt [[Schott et al., 2009](#)]. The numerical set-up is shown in Figure 7.5 (a). A four-grain RVE is constructed with 1 mm edge length. The initial orientations of the grains are denoted in Figure 7.5 (a), with [001] axis perpendicular to the xy plane. Constant average stress rate is enforced during the loading period until the destinate stress is reached, and then constant stress boundary condition is enforced during the creep deformation period.

Grain boundary layer with 0.05 mm thickness is introduced between each pair of grains, and initial grain boundary indicator d_{GB} is assigned to the grain boundary region.

Crystal plasticity constitutive relation is assigned to the grain boundary region, and the initial orientation inherits from the neighboring grains. In the current model, grain boundary thickness is also a key material parameter. The solution-precipitation creep rate increases as the grain boundary thickness increases. A constant grain boundary thickness should be used for different RVEs to reveal size effect instead of using an arbitrary fraction of the RVE edge length.

The triple junctions of the grain boundary region also require proper treatment regarding the grain boundary normal and the solution precipitation strain mode. Here, the solution precipitation creep deformation $\dot{\epsilon}$ equals to zero at the triple junctions, but the initial phase field is enforced as 1 for compensation. Otherwise, the triple junctions become rigid inclusions as creep deformation increases.

In this paper, only solution precipitation creep is considered instead of the competition between dislocation creep and solution precipitation creep, since the hardening variable $\tilde{\tau}$ is monotonically increasing in the Voce type crystal plasticity model (7.29). The dislocation creep can be incorporated by introducing a dislocation annihilation mechanism to allow decreasing hardening variables.

Figure 7.5 (b) shows the creep rate evolution during the creep loading under different stress level. Three constant stress levels are tested: 0.1 MPa, 0.5 MPa, 1.0 MPa. During the constant stress-rate loading period, a monotonically increasing strain rate is observed which represents the transition from the elastic region to the plastic region. In the constant stress period, the strain rate gradually decreases due to the monotonically increasing hardening variable. Then, the creep strain rate reaches a constant value which depends on the stress level. It is also observed that the strain rate constant $n = 1$, which is consistent with the solution precipitation creep experiment [Schott et al., 2009].

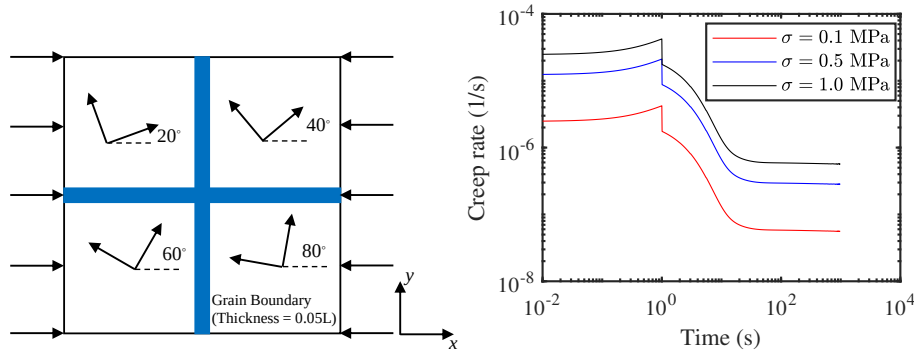


Fig. 7.5. Analysis of creep loading effect on solution-precipitation creep rate: (a) The numerical set-up for the creep simulation; (b) Creep rate evolution for each creep loading condition.

Figure 7.6 shows the evolution of grain boundary brine concentration (a-c), longitudinal strain (d-f), and transverse strain (g-i) for the $\sigma = 1.0$ MPa case. It is observed that as time increases, the grain boundary brine concentration almost remains constant. A concentration gradient exists along the grain boundary, where the high concentration region corresponds to the grain boundary region with higher grain boundary normal compression. Concentration flux normal to the grain boundary is almost negligible, although slight concentration increasing is observed at the grain bulk adjacent to the grain boundary. On the other hand, creep deformation is observed in the grain boundary region as time increases. A compressive grain-boundary-normal strain ϵ_{xx} is observed in

the vertical grain boundary since the grain boundary pressure p is much higher than the horizontal grain boundary where tensile grain-boundary-normal strain ε_{yy} is observed. It is also observed that the compressive strain ε_{xx} in the vertical grain boundary region approximately equals the tensile strain ε_{yy} in the horizontal grain boundary region, indicating that the solution precipitation model is mass conservative assuming that density is constant. Note that the strain values at the triple junctions have large perturbations, since four grains with different orientations interact with each other. The stiffness of the triple junction is reduced by imposing unit initial phase field d_c in this region.

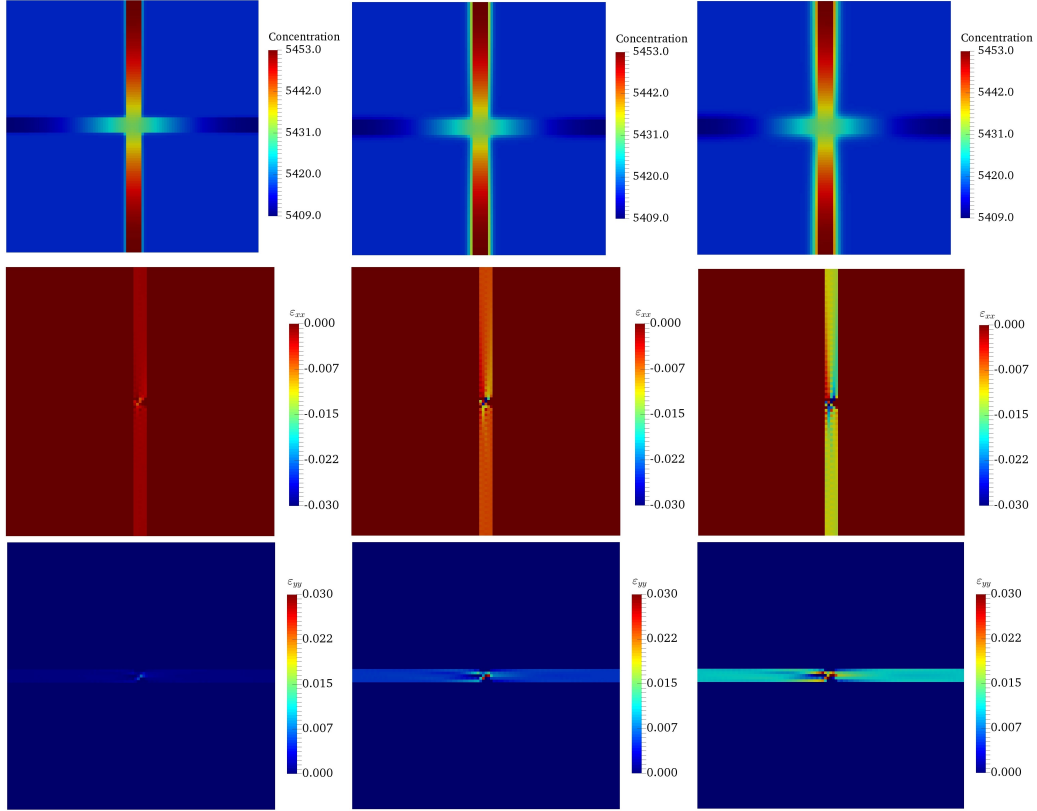


Fig. 7.6. Brine concentration and strain distribution at different creep stages for the $\sigma = 1$ MPa simulation in Figure 7.5: (a-c) Evolution of grain boundary brine concentration [unit: mol m^{-3}]; (d-f) Evolution of strain in the loading direction ε_{xx} ; (g-i) Evolution of strain in the transverse direction ε_{yy} .

7.5.3 Anisotropic cracking

The third example is designed to illustrate the anisotropic crack initiation and propagation in polycrystalline rock salt and the competition among intergranular and intragranular fracture, heat transfer and plastic deformation.

A 2D polycrystalline RVE with 40 grains is generated by Neper [Quey et al., 2011], and the RVE is equally divided into 399×399 grid points, as shown in Figure 7.7 (a). The RVE edge length is 1 mm, such that the average grain size is 0.2 mm. Random initial orientations are assigned to each grain with the [001] axis perpendicular to the xy plane, such that the cleavage planes are also perpendicular to the xy plane. The fracture energy

G_c of the material point within the bulk region is 1.15 J m^2 , and the preferential fracture plane is $\{001\}$. Reduced fracture energy (1.0 J m^2) is assigned to the grain boundary region, and the grain boundary is assumed to be isotropic in crack propagation. Small strain crystal plasticity model is applied to both the grain interior region and the grain boundary region. The grain boundary layer thickness approximately equals to $0.014L$, where L represents the RVE edge length. The deformation process is assumed to be isothermal at room temperature, which is reasonable considering that the specimen size is relatively small.

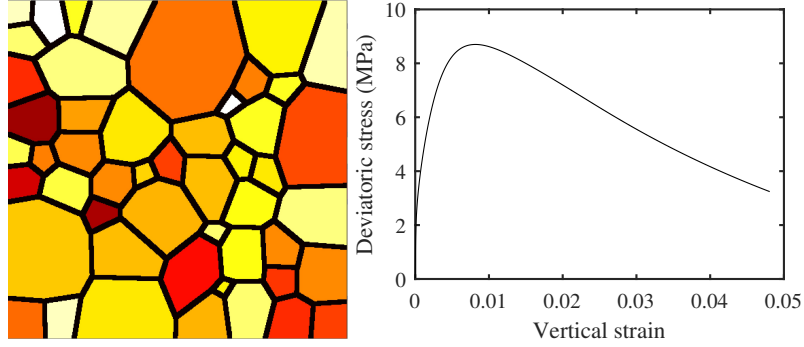


Fig. 7.7. Analysis of crack initiation and propagation in polycrystalline rock salt: (a) Initial configuration of polycrystal RVE and grain boundary. The RVE edge length L is 1 mm, and the grain boundary thickness is $l = 0.014L$, the initial orientation is random with $[001]$ axis perpendicular to the xy plane. (b) Homogenized stress-strain curve.

Pure shear average strain is enforced as the boundary condition, with a constant average strain rate $\dot{\gamma} = 1.0 \times 10^{-4} \text{ s}^{-1}$:

$$\boldsymbol{\varepsilon} = \begin{bmatrix} \gamma & 0.0 \\ 0.0 & -\gamma \end{bmatrix}. \quad (7.48)$$

Figure 7.7 (b) shows the homogenized stress-strain response in the axial direction. It is observed that the fracture process of polycrystal rock salt is more ductile compared with corresponding single crystal results [Na and Sun, 2018]. One major reason is that the grain boundary region and intergranular anisotropy prevent the crack from propagating through the specimen, as shown in Figure 7.8 (a-c). It is observed that the as external loading increases, both intragranular crack and intergranular crack initiate and gradually forms a network within the specimen.

The Von-Mises stress distributions at different loading stages are shown in Figure 7.8 (d-f). As expected, stress concentration is observed at the crack tip and the grain boundary regions. The Gibbs effect is also observed mainly for two reasons: (1) regular frequency vector is used instead of the finite difference based frequency vector for solving the mechanical problem; (2) large material stiffness contrast exists within the interfacial region.

7.5.4 Chemical-diffusion-controlled crack healing

In the last example, diffusion-controlled crack healing is simulated through a prescribed loading-unloading-reloading strain path. A two-dimensional single-crystal RVE is divided into 399^2 grid points, and the edge length of the specimen is 1 mm by 1 mm. A

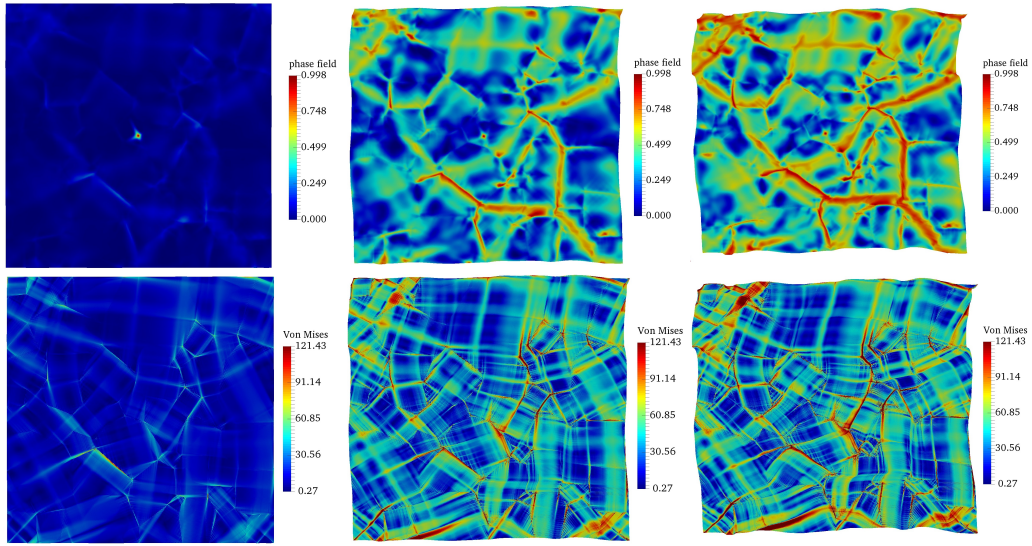


Fig. 7.8. Phase field and Von Mises stress distribution within the polycrystal specimen at different load stages: (a-c) Phase field; (d-f) Von Mises stress ([unit: MPa]).

circular flaw with 0.1 mm radius is introduced in the center of the specimen for crack initiation. The initial Euler angle is $(0^\circ, 0^\circ, 0^\circ)$ in Bunge notation, such that the $[100]$ axis is parallel to the loading direction. The loading-unloading-reloading strain path is shown in Figure 7.9 (a). A uniaxial tension boundary condition is conducted with a constant strain rate.

In this section, the linear elastic constitutive relation is used instead of crystal plasticity for several reasons. First, creeping deformation during the healing process due to the residual stress can be avoided, such that the crack healing simulation could be more comparable to the experiment [Houben et al., 2013]. Also, the focus of this section is to demonstrate the effectiveness of the precipitation, diffusion, and crack healing model, and to determine whether crack healing in rock salt is diffusion-controlled or precipitation controlled. Note that the usage of elastic constitutive relation is solely for illustration convenience without losing generality.

The homogenized stress-strain response of the loading-unloading-reloading process is shown in Figure 7.9 (b). Reduced stress due to crack propagation is observed after the external loading reaches a critical point, and unloading boundary condition is initiated before the crack propagates through the specimen. This is possible for the staggered coupling scheme, where crack initiation and propagation are delayed compared with the corresponding monolithic coupling scheme. After the unloading process, the specimen is held at stress-free status for 300 seconds which is long enough for diffusion induced crack healing, and stiffness recovery is observed during the reloading process.

The phase-field evolution during the loading-unloading-reloading process is shown in Figure 7.10. The initial circular flaw with 0.1 mm radius is prescribed by the initial phase field, as shown in Figure 7.10 (a), to provide crack initiation spot. The fracture phase-field distribution after the loading process, the stress-free holding process, and the reloading process are shown in Figure 7.10 (b), (c), and (d), respectively. The crack did not penetrate through the specimen after the unloading process, as shown in Figure 7.10 (b). Crack healing is observed after the stress-free holding process as shown in Figure

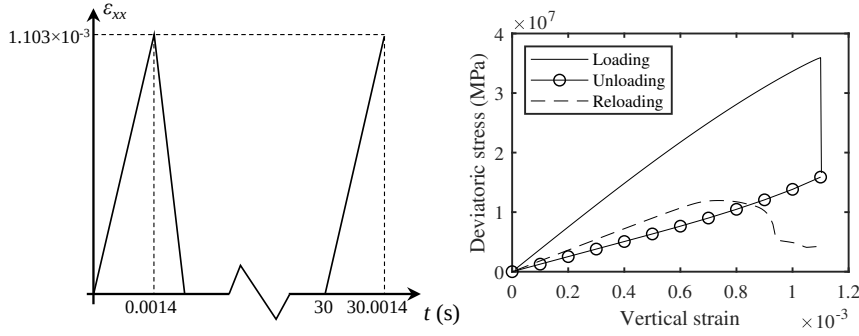


Fig. 7.9. Strain path and stress strain curve of the crack healing simulation. (a) Strain path of the loading-unloading-reloading process. (b) Uniaxial stress-strain curve during the loading-unloading-reloading process. The RVE holds for a while with no external loading between the unloading process and the reloading process to provide enough time for crack healing.

7.10 (c), which explains the stiffness recovery observed in the homogenized stress-strain response in Figure 7.9 (b). During the reloading process, the crack continues to propagate along the original path when the external loading reaches a critical point.

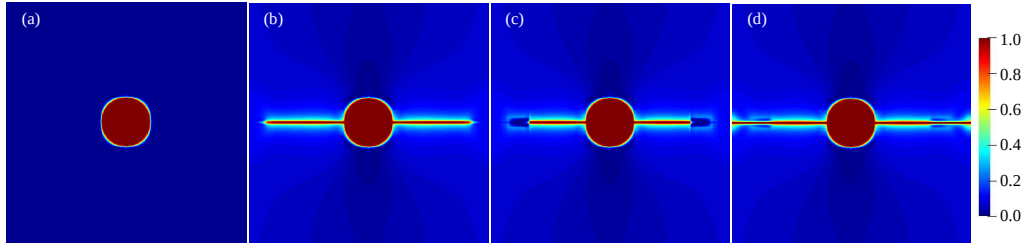


Fig. 7.10. Phase field evolution during the loading-unloading-reloading process. (a) Initial phase-field representing the initial circular flaw with 0.1 mm radius; (b) Fracture phase-field distribution after the loading and unloading process; (c) Crack healing after the stress-free holding process; (d) Crack propagation continues to penetrate the specimen after the reloading process.

The chemical concentration distribution corresponding to the time step described in Figure 7.10 (a-d) are shown in Figure 7.11 (a-d), respectively. The initial chemical concentration (Figure 7.10 (a)) is homogeneous and equals to the equilibrium chemical concentration at stress-free states. After the loading and unloading process, the chemical concentration remains unchanged, since the time is very short and both solution-precipitation and diffusion are time dependent. After holding the sample at stress-free states for 300 seconds which is long enough for solution-precipitation and diffusion to occur, the solid-liquid interface curvature induced precipitation is observed within the crack tip region and the initial flaw region, as shown in Figure 7.11 (c). This precipitation causes the crack healing observed in Figure 7.10 (c) and the stiffness recovery observed in Figure 7.9 (b).

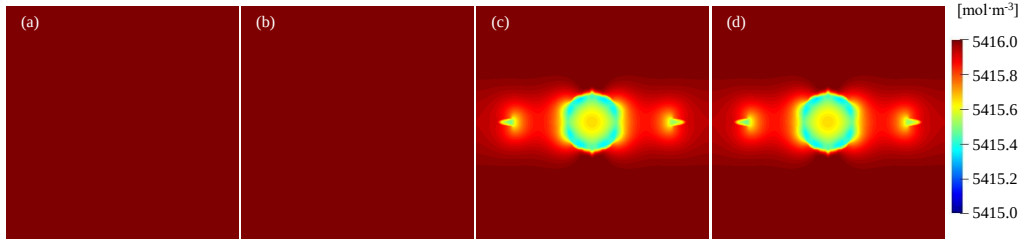


Fig. 7.11. Chemical concentration distribution during the loading-unloading-reloading process. (a) Homogeneous initial chemical concentration equals to the equilibrium concentration c_{eq} at stress free status; (b) Chemical concentration after the loading and unloading process corresponding to Figure 7.10 (b); (c) Chemical concentration distribution after the stress-free holding process corresponding to Figure 7.10 (c); (d) Chemical concentration after the reloading process corresponding to Figure 7.10 (d).

7.6 Conclusions

A mathematics framework is proposed to simulate the long-term creep, fracture and healing coupling process in rock salt under a variety of thermal, mechanical and chemical conditions. An FFT-based method is employed to solve the coupled equations in a staggered scheme. By leveraging the numerical efficiency and globally C_∞ continuous basis function, the strongly anisotropic crack growth and healing are simulated. Our numerical examples demonstrate that the proposed model is capable of capturing the physical behavior observed in rock salt, including solution-precipitation creep, strongly anisotropic cracking, and diffusion controlled crack healing with stiffness restoration.

Chapter 8

Three-dimensional benchmark simulations

We model thermo-mechanical interaction between the nucleate waste disposal and its surrounding medium by writing the balance of linear momentum for quasi-static deformation of continua and the balance of energy as

$$\nabla^X \cdot \sigma + \rho b = 0, \quad (8.1)$$

$$\rho c_p \dot{\theta} + \nabla^X \cdot q = G_\theta - G_u, \quad (8.2)$$

where σ , b , q , θ , G_θ , G_u , ρ , and c_p are Cauchy stress, body force, heat flux, temperature, rate of heat energy provided externally, rate of heat energy generated mechanically, material density, and specific heat at constant pressure, respectively.

Using infinitesimal kinematics and neglecting convective and radiation heat transfer the governing constitutive equations can be derived by the Duhamel-Neumann generalization of Hooke's and Fourier's heat conduction laws:

$$\sigma = C : (\varepsilon - \varepsilon_\theta); \quad C = \lambda I \otimes I + 2\mu I^{\text{sym}}, \quad (8.3)$$

$$q = -\kappa \nabla^X \theta, \quad (8.4)$$

Total and thermal strain tensors are defined as $\varepsilon = \nabla^{X^{\text{sym}}} u$ and $\varepsilon_\theta = \alpha(\theta - \theta_0) I$, respectively. $\theta_0(x, t = 0)$ is temperature distribution at rest. $u(x, t)$ is the displacement field at spatial coordinate x and time t . The heat energy generated through the mechanical deformation can be expressed by $G_u = \alpha(3\lambda + 2\mu)\theta_0 \text{tr}(\dot{\varepsilon})$. From now on, we neglect the effect of this term comparing to other terms due to the low value of thermal expansion, α , in the waste repository medium and mechanical loading rate [?]. $\nabla^X \cdot \Xi$, $\nabla^X \Xi$, $\nabla^{X^{\text{sym}}} \Xi$, $\dot{\Xi}$, $\text{tr}(\Xi)$, and $\Xi \otimes \Xi$ are spatial divergence, gradient, symmetric gradient, material time derivative, trace, and dyad operators acting on general tensor fields Ξ and Ξ , respectively. In this study, Lamé parameters, λ and μ , and thermal expansion coefficient are constants while thermal conductivity $\kappa(\theta, \phi)$ and density $\rho(\phi)$ depend on the temperature and displacement fields via porosity variable $\phi = \phi_0(1 + \text{tr}(\dot{\varepsilon}))$.

The Euler-Lagrange equation (??) are supplemented with the boundary and initial conditions:

$$\sigma(x, t) \cdot n(x) = \bar{\sigma}_n(x, t) \quad x \in \Gamma_\sigma, t \in \mathbb{R}^+, \quad (8.5)$$

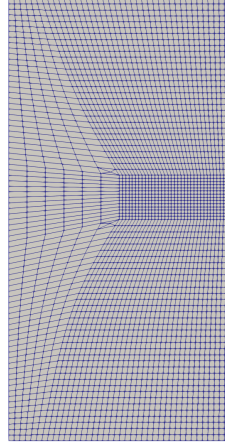


Fig. 8.1. Planar view of the 3-D computational domain.

$$\mathbf{u}(\mathbf{x}, t) = \bar{\mathbf{u}}(\mathbf{x}, t) \quad \mathbf{x} \in \Gamma_u, t \in \mathbb{R}^+ \cup \{0\}, \quad (8.6)$$

$$\mathbf{q}(\mathbf{x}, t) \cdot \mathbf{n}(\mathbf{x}) = \bar{q}_n(\mathbf{x}, t) \quad \mathbf{x} \in \Gamma_q, t \in \mathbb{R}^+, \quad (8.7)$$

$$\theta(\mathbf{x}, t) = \bar{\theta}(\mathbf{x}, t) \quad \mathbf{x} \in \Gamma_\theta, t \in \mathbb{R}^+ \cup \{0\}, \quad (8.8)$$

where $\Gamma_\sigma \cup \Gamma_u = \Gamma_q \cup \Gamma_\theta = \Gamma$, $\Gamma_\sigma \cap \Gamma_u = \Gamma_q \cap \Gamma_\theta = \emptyset$, and $\mathbf{n}$ is the unit normal vector to the boundary surface Γ . $\bar{\sigma}_n$, $\bar{\mathbf{u}}$, $\bar{q}_n$, and $\bar{\theta}$ are prescribed traction, displacement, net heat flux, and temperature over the boundary, respectively.

8.1 Numerical treatment

The Galerkin-Ritz approach is used to derive the weak statement of the above mentioned Euler-Largange equations. Subsequently, we employ the Bubnov-Galerkin method to obtain semi-discrete equations. The resultant nonlinear coupled equations are linearized by the Newton-Raphson method. Lastly, we find the final system of discrete equations by the implicit Euler time marching method, and the linear sytem is solved in a staggered fashion.

8.2 Numerical Benchmarks

The benchmark problem is chosen from [?] where a three dimensional waste repository is numerically solved. We consider the numerical domain contained the intact salt and alcove, and subscripts i and a indicate the corresponding properties for intact salt and alcove, respectively.

Material properties used in this study are $B_i = 1.2, B_a = 40\text{GPa}$, $\nu_i = \nu_a = 0.252$, $\rho_{0_i} = \rho_{0_a} = 2160 \frac{\text{Kg}}{\text{m}^3}$, $c_{p_i} = c_{p_a} = 931 \frac{\text{J}}{\text{Kg}\cdot\text{K}}$, $\alpha_i = 4.4 \times 10^{-5}$, $\alpha_a = 3.52 \times 10^{-5} \frac{1}{\text{K}}$, and $\phi_{0_i} = 0$, $\phi_{0_a} = 0.2$ where B and ν are Bulk modulus and Poisson's ratio. According to [?] the following empirical equations is used for thermal conductivity:

$$\kappa(\theta, \phi) = \kappa_{cs}(\phi) \left(\frac{300}{\theta} \right)^\gamma, \quad (8.9)$$

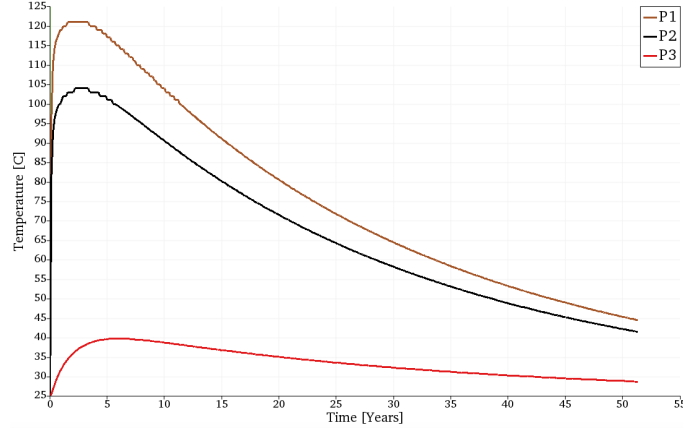


Fig. 8.2. Temperature history at different observation points: P1 = (0, 1.17, 1.4)m; P2 = (0, 0.59, 0.57)m; P3 = (0, 10, 1.42)m.

$$\kappa_{cs}(\phi) = \bar{\kappa}_{cs} \left(-270\phi^4 + 370\phi^3 - 136\phi^2 + 1.5\phi + 5 \right), \quad (8.10)$$

where $\gamma = 1.14$ and $\bar{\kappa}_{cs} = 1.08$ are model constants. Also, the density function is defined by $\rho(\phi) = \phi(1 - \phi_0)$.

The geometry of the boundary value problem is as follows. By applying symmetry, half the alcove is simulated. The width, height, and depth of the modeled half-alcove are $8 \times 4 \times 10$ m. The width, height, and depth of the half-domain are $20 \times 40 \times 20$ m. The boundaries of the half-alcove are conformally meshed, with the symmetry plane applied along the right-lateral boundary in Fig. 8.1. Mesh parameters used in this study are $h_i = 0.5$, $h_a = 0.1$ m, where h_i is the characteristic element length of the computational mesh towards the far-field, and h_a is the element length in the alcove.

Boundary and initial conditions are as follows. Symmetry conditions are imposed on all far-field boundaries, with respect to the displacement field. Zero Neumann conditions are imposed on all far-field boundaries, with respect to the temperature field, with the exception of the top and bottom which are Dirichlet and fixed to the initial temperature. The domain is initialized to static equilibrium, subject to overburden stress anisotropy of 5.9kPa. The initial temperature in the domain is 25°C. The heat source within the alcove is modeled as a time-varying contribution to G_θ . The decline curve characterizing this source term's time variance is calibrated to Fig. 3 in [?], i.e. the curve exponentially decays.

Deformation and temperature are modeled over a 50yr period. Due to the exponential decay of the source term, the temperature within the alcove rapidly increases but maximizes within 5yrs, Fig. 8.2 and Fig. 8.3(a-d). The temperature across the lateral cross-section of the domain is in 8.4(a-d).

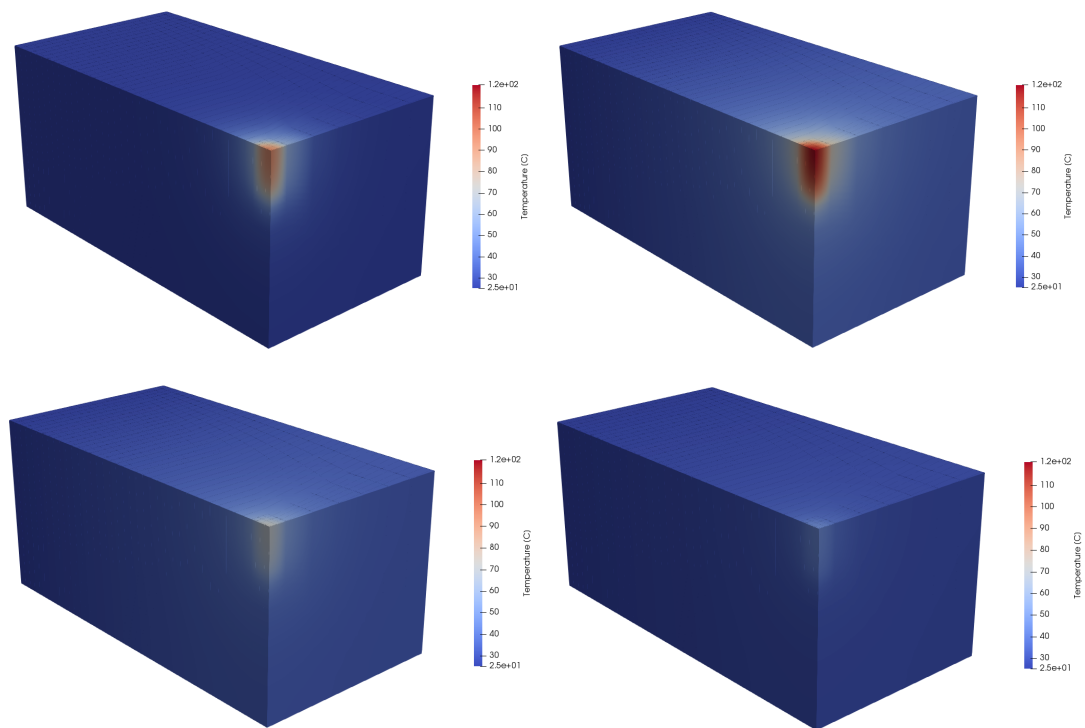


Fig. 8.3. Temperature field in near-alcove region after 0.125, 2, 25 and 50 years.

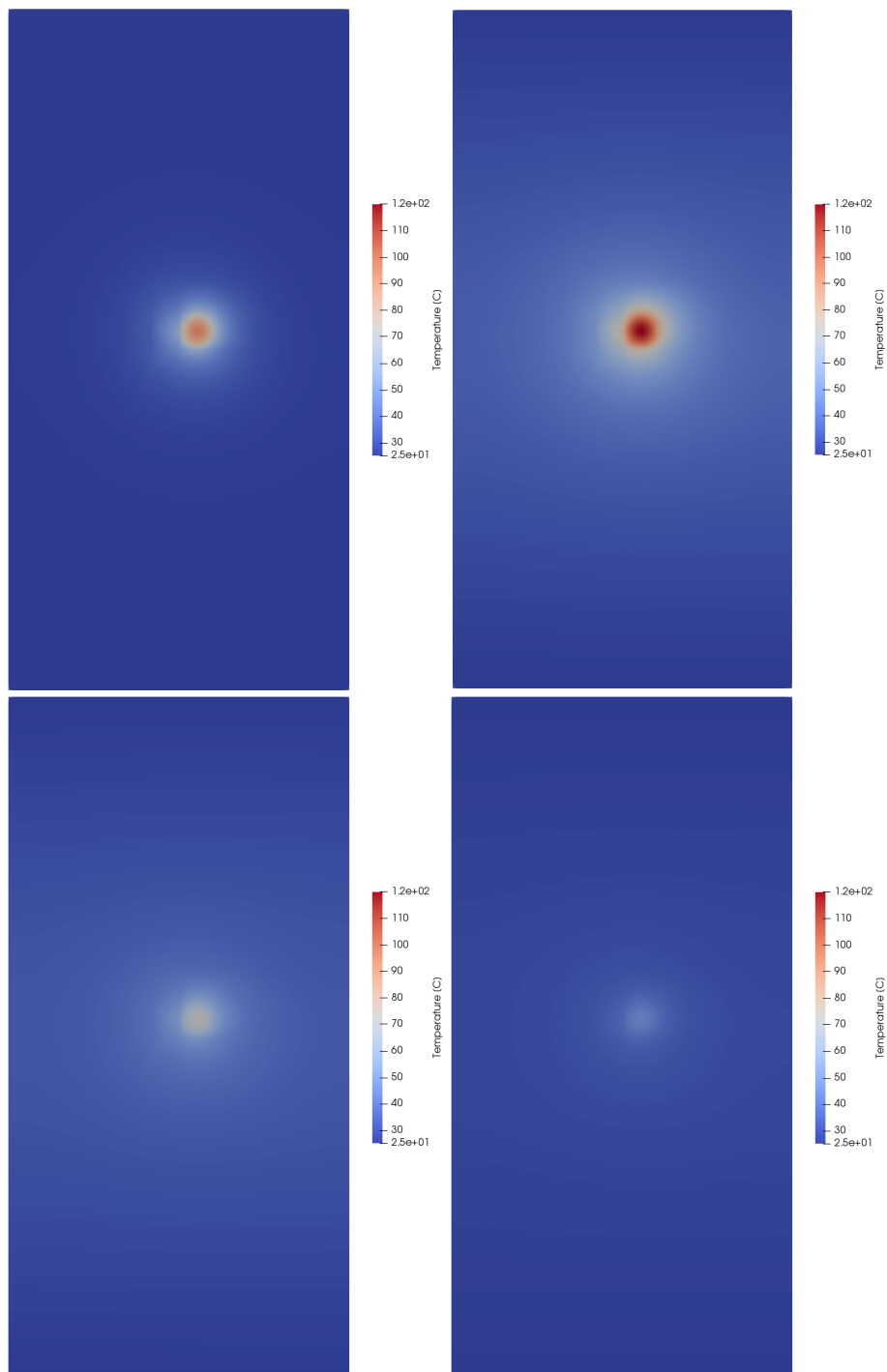


Fig. 8.4. Temperature field across the domain after 0.125, 2, 25 and 50 years.

Chapter 9

Final Quad Chart

The final quad chart is listed below.

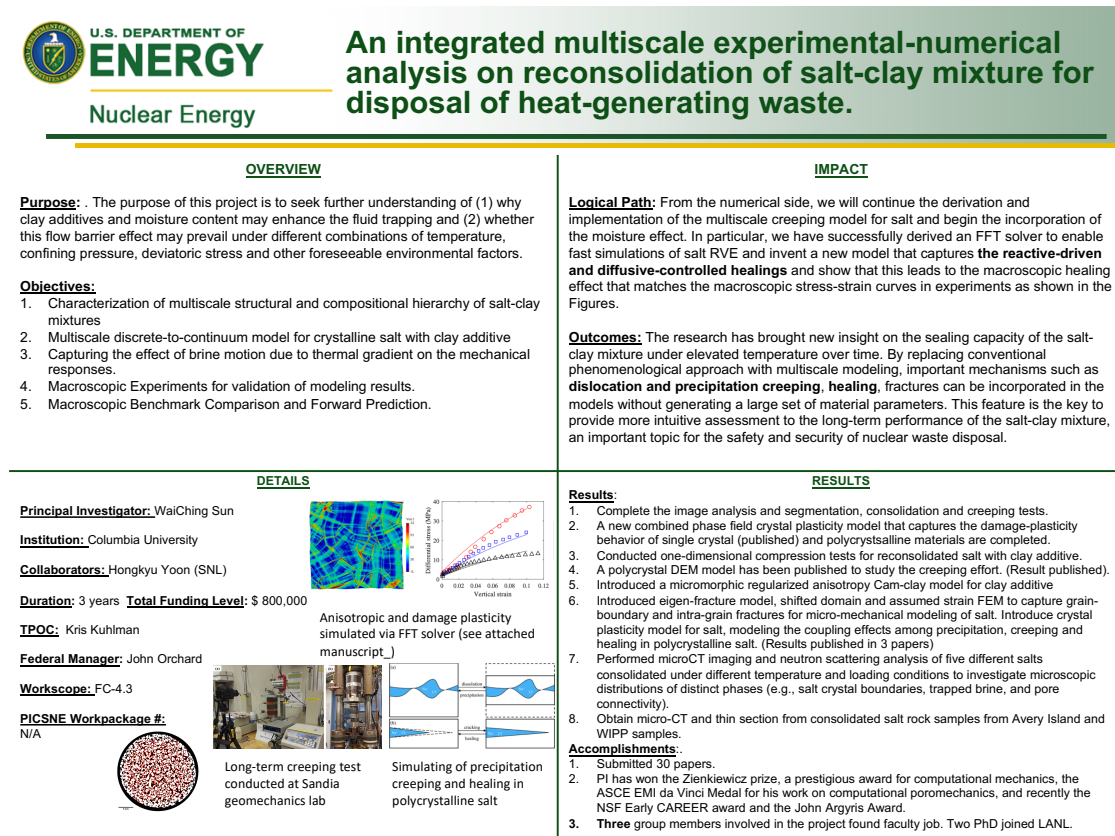


Fig. 9.1. Final Quad Chart.

Bibliography

- Frank D Hansen and Christi D Leigh. *Salt disposal of heat-generating nuclear waste*. Sandia National Laboratories Albuquerque, NM, 2011.
- William D Callister Jr and David G Rethwisch. *Fundamentals of materials science and engineering: an integrated approach*. John Wiley & Sons, 2012.
- William G Moffat, George W Pearsall, and John Wulff. *The structure and properties of materials. volume 1, structure*, volume 1. Wiley New York, 1964.
- R.I. Borja. *Plasticity: modeling & computation*. Springer Science & Business Media, 2013.
- Benjamin Reedlunn. Reinvestigation into closure predictions of room d at the waste isolation pilot plant (sand2016-9961). Technical report, Sandia National Laboratories (SNL-NM), Albuquerque, NM (United States), 2016.
- DE Munson, RL Jones, JR Ball, RM Clancy, DL Hoag, and SV Petney. Overtest for simulated defense high-level waste (room b): In situ data report (may 1984–february 1988): Waste isolation pilot plant (wipp) thermal/structural interactions program. Technical report, Sandia National Labs., Albuquerque, NM (USA), 1990.
- Kento Oshima, Tomohiro Takaki, and Mayu Muramatsu. Development of multi-phase-field crack model for crack propagation in polycrystal. *International Journal of Computational Materials Science and Engineering*, 3(02):1450009, 2014.
- Neville L Carter and Hugh C Heard. Temperature and rate dependent deformation of halite. *American Journal of Science*, 269(3):193–249, 1970.
- Thanh Tung Nguyen, Julien Réthoré, and Marie-Christine Baietto. Phase field modelling of anisotropic crack propagation. *European Journal of Mechanics-A/Solids*, 2017a.
- Thanh-Tung Nguyen, Julien Rethore, Julien Yvonnet, and Marie-Christine Baietto. Multi-phase-field modeling of anisotropic crack propagation for polycrystalline materials. *Computational Mechanics*, pages 1–26, 2017b.
- Kane C Bennett, Lucas A Berla, William D Nix, and Ronaldo I Borja. Instrumented nanoindentation and 3d mechanistic modeling of a shale at multiple scales. *Acta Geotechnica*, 10(1):1–14, 2015.
- M Ortiz and A Pandolfi. A variational Cam-clay theory of plasticity. *Computer Methods in Applied Mechanics and Engineering*, 193(27-29):2645–2666, 2004.
- S.J. Semnani, J.A. White, and R.I. Borja. Thermoplasticity and strain localization in transversely isotropic materials based on anisotropic critical state plasticity. *International*

- Journal for Numerical and Analytical Methods in Geomechanics*, 40(18):2423–2449, 2016. ISSN 1096-9853.
- K. Ikeda, Y. Yamakawa, and S. Tsutsumi. Simulation and interpretation of diffuse mode bifurcation of elastoplastic solids. *Journal of the Mechanics and Physics of Solids*, 51(9): 1649–1673, 2003.
- George W. Scherer. Stress from crystallization of salt. *Cement and Concrete Research*, 34(9): 1613–1624, 2004.
- Michael Steiger. Crystal growth in porous materials—I: The crystallization pressure of large crystals. *Journal of Crystal Growth*, 282(3-4):455–469, 2005a.
- Hannelore Derluyn, Peter Moonen, and Jan Carmeliet. Deformation and damage due to drying-induced salt crystallization in porous limestone. *Journal of the Mechanics and Physics of Solids*, 63(1):242–255, 2014.
- A. C. Liakopoulos. *Transient flow through unsaturated porous media*. PhD thesis, University of California, Berkeley, 1964.
- Guillaume Desbois, Janos L Urai, and Johannes HP de Bresser. Fluid distribution in grain boundaries of natural fine-grained rock salt deformed at low differential stress (qom kuh salt fountain, central iran): Implications for rheology and transport properties. *Journal of Structural Geology*, 43:128–143, 2012a.
- F Hansen, T Popp, K Wieczorek, and D Stührenberg. Salt reconsolidation applied to repository seals. In *The Mechanical Behaviour of Salt VIII, Rapid City, South Dakota*, pages 179–189. Taylor & Francis Group London, 2015.
- Frank Hansen, Till Popp, Klaus Wieczorek, and Dieter Stuehrenberg. Granular salt summary: reconsolidation principles and applications. Technical report, Sandia National Lab.(SNL-NM), Albuquerque, NM (United States), 2014a.
- ST Broome, SJ Bauer, FD Hansen, et al. Reconsolidation of crushed salt to 250 c under hydrostatic and shear stress conditions. In *48th US rock mechanics/geomechanics symposium*. American Rock Mechanics Association, 2014.
- S Olivella and A Gens. A constitutive model for crushed salt. *International journal for numerical and analytical methods in geomechanics*, 26(7):719–746, 2002.
- CJ Spiers, PMTM Schutjens, RH Brzesowsky, CJ Peach, JL Liezenberg, and HJ Zwart. Experimental determination of constitutive parameters governing creep of rocksalt by pressure solution. *Geological Society, London, Special Publications*, 54(1):215–227, 1990.
- A Hampel. Joint projects on the comparison of constitutive models for the mechanical behavior of rock salt ii. overview of the models and results of 3-d benchmark calculations: A. hampel k. salzer, r.-m. günther & w. minkley. In *Mechanical Behaviour of Salt VII*, pages 245–254. CRC Press, 2012.
- SeonHong Na and WaiChing Sun. Computational thermomechanics of crystalline rock, part i: A combined multi-phase-field/crystal plasticity approach for single crystal simulations. *Computer Methods in Applied Mechanics and Engineering*, 338:657–691, 2018.

- Ran Ma and WaiChing Sun. Fft-based solver for higher-order and multi-phase-field fracture models applied to strongly anisotropic brittle materials and poly-crystals. *Computer methods in applied Mechanics and engineering*, 2020.
- Hongkyu Yoon, Albert J Valocchi, Charles J Werth, and Thomas Dewers. Pore-scale simulation of mixing-induced calcium carbonate precipitation and dissolution in a microfluidic pore network. *Water Resources Research*, 48(2), 2012.
- Hongkyu Yoon, Qinjun Kang, and Albert J Valocchi. Lattice boltzmann-based approaches for pore-scale reactive transport. *Reviews in Mineralogy and Geochemistry*, 80(1):393–431, 2015.
- Kun Wang and WaiChing Sun. An updated lagrangian lbm–dem–fem coupling model for dual-permeability fissured porous media with embedded discontinuities. *Computer Methods in Applied Mechanics and Engineering*, 344:276–305, 2019.
- Yang Liu, WaiChing Sun, Zifeng Yuan, and Jacob Fish. A nonlocal multiscale discrete-continuum model for predicting mechanical behavior of granular materials. *International Journal for Numerical Methods in Engineering*, 106(2):129–160, 2016a.
- Kun Wang and WaiChing Sun. A semi-implicit discrete-continuum coupling method for porous media based on the effective stress principle at finite strain. *Computer Methods in Applied Mechanics and Engineering*, 304:546–583, 2016a.
- Eric C Bryant and WaiChing Sun. A micromorphically regularized cam-clay model for capturing size-dependent anisotropy of geomaterials. *Computer Methods in Applied Mechanics and Engineering*, 354:56–95, 2019.
- KP Kröhn, CL Zhang, J Wolf, D Stührenberg, M Jobmann, L von Borstel, and C Lerch. The compaction behaviour of salt backfill at low porosities. In *Mechanical Behaviour of Salt VII*, pages 175–182. CRC Press, 2012.
- Guillaume Desbois, Janos L Urai, Peter A Kukla, Uwe Wollenberg, Fabián Pérez-Willard, Zsolt Radí, and Sandor Riholm. Distribution of brine in grain boundaries during static recrystallization in wet, synthetic halite: Insight from broad ion beam sectioning and sem observation at cryogenic temperature. *Contributions to Mineralogy and Petrology*, 163(1):19–31, 2012b.
- Stephen J Bauer, Francis D Hansen, Scott Thomas Broome, Melissa Marie Mills, Brandon UNM Lampe, and John UNM Stormont. Gas flow measurements through consolidating crushed salt. Technical report, Sandia National Lab.(SNL-NM), Albuquerque, NM (United States), 2015.
- Kristopher L Kuhlman and Bwalya Malama. Brine flow in heated geologic salt (sand2013-1944). Technical report, Sandia National Labs., Albuquerque, NM (USA), 2013.
- Thilo von Berlepsch and Bernt Haverkamp. Salt as a host rock for the geological repository for nuclear waste. *Elements*, 12(4):257–262, 2016.
- Alexander Urquhart and Stephen Bauer. Experimental determination of single-crystal halite thermal conductivity, diffusivity and specific heat from- 75 c to 300 c. *International Journal of Rock Mechanics and Mining Sciences*, 78:350–352, 2015.

- W Bechthold, E Smailos, S Heusermann, W Bollingerfehr, B Bazargan Sabet, T Rothfuchs, P Kamlot, J Grupa, S Olivella, and FD Hansen. Backfilling and sealing of underground repositories for radioactive waste in salt (bambus-ii project). *Commission of the European Communities, EUR*, 20621, 2004.
- KS Chan, SR Bodner, and DE Munson. Recovery and healing of damage in wipp salt. *International Journal of Damage Mechanics*, 7(2):143–166, 1998.
- RL Bradshaw and Florentino Sanchez. Migration of brine cavities in rock salt. *Journal of Geophysical Research*, 74(17):4209–4212, 1969.
- EJ Nowak and DF McTigue. Interim results of brine transport studies in the waste isolation pilot plant (wipp). Technical report, Sandia National Labs., Albuquerque, NM (USA), 1987.
- RL Beauheim, SM Howarth, P Vaughn, SW Webb, and KW Larson. Integrated modeling and experimental programs to predict brine and gas flow at the waste isolation pilot plant (no. sand-94-0599c; conf-941053-1). Technical report, Sandia National Labs., Albuquerque, NM (United States), 1995.
- Zsolt Schlöder, János L Urai, Sofie Nollet, and Christoph Hilgers. Solution-precipitation creep and fluid flow in halite: a case study of zechstein (z1) rocksalt from neuhof salt mine (germany). *International Journal of Earth Sciences*, 97(5):1045–1056, 2008.
- FD Hansen, SJ Bauer, ST Broome, and GD Callahan. Coupled thermal-hydrological-mechanical processes in salt: Hot granular salt consolidation, constitutive model and micromechanics (sand2012-9893p). Technical report, Sandia National Labs., Albuquerque, NM (United States), 2012.
- M Guillope and JP Poirier. Dynamic recrystallization during creep of single-crystalline halite: An experimental study. *Journal of Geophysical Research: Solid Earth*, 84(B10): 5557–5567, 1979.
- Neville L Carter, Francis D Hansen, and Paul E Senseny. Stress magnitudes in natural rock salt. *Journal of Geophysical Research: Solid Earth*, 87(B11):9289–9300, 1982.
- Neville L Carter and Francis D Hansen. Creep of rocksalt. *Tectonophysics*, 92(4):275–333, 1983.
- Janos L Urai, Christopher J Spiers, Hendrik J Zwart, and Gordon S Lister. Weakening of rock salt by water during long-term creep. *Nature*, 324(6097):554–557, 1986.
- NL Carter, ST Horseman, JE Russell, and J Handin. Rheology of rocksalt. *Journal of Structural Geology*, 15(9-10):1257–1271, 1993.
- PE Senseny, FD Hansen, JE Russell, NL Carter, and JW Handin. Mechanical behaviour of rock salt: phenomenology and micromechanisms. In *International journal of rock mechanics and mining sciences & geomechanics abstracts*, volume 29, pages 363–378. Elsevier, 1992.
- JL Urai, Z Schlöder, CJ Spiers, and PA Kukla. Flow and transport properties of salt rocks. *Dynamics of complex intracontinental basins: The central European basin system*, pages 277–290, 2008.

- Frank D Hansen et al. Micromechanics of isochoric salt deformation. In *48th US Rock Mechanics/Geomechanics Symposium*. American Rock Mechanics Association, 2014b.
- Darrell E Munson and PR Dawson. Constitutive model for the low temperature creep of salt (with application to wipp). Technical report, Sandia National Labs., Albuquerque, NM (USA), 1979.
- KS Chan, AF Fossum, SR Bodner, DE Munson, et al. Constitutive representation of damage healing in wlp salt. In *The 35th US Symposium on Rock Mechanics (USRMS)*. American Rock Mechanics Association, 1995.
- Shuke Miao, Ming L Wang, and Howard L Schreyer. Constitutive models for healing of materials with application to compaction of crushed rock salt. *Journal of Engineering Mechanics*, 121(10):1122–1129, 1995.
- DE Munson. Constitutive model of creep in rock salt applied to underground room closure. *International Journal of Rock Mechanics and Mining Sciences*, 34(2):233–247, 1997.
- Michel Aubertin, Michel R Julien, Stéphane Servant, and Denis E Gill. A rate-dependent model for the ductile behavior of salt rocks. *Canadian Geotechnical Journal*, 36(4):660–674, 1999.
- Cheng Zhu and Chloé Arson. A model of damage and healing coupling halite thermo-mechanical behavior to microstructure evolution. *Geotechnical and Geological Engineering*, 33(2):389–410, 2015a.
- ST Broome, SJ Bauer, FD Hansen, and MM Mills. Mechanical response and microprocesses of reconsolidating crushed salt at elevated temperature. *Rock Mechanics and Rock Engineering*, 48(6):2615–2629, 2015.
- Xianda Shen, C Zhu, C Arson, et al. Chemo-mechanical damage and healing of granular salt: micro-macro modeling. In *50th US Rock Mechanics/Geomechanics Symposium*. American Rock Mechanics Association, 2016.
- S Olivella, J Carrera, A Gens, and EE Alonso. Nonisothermal multiphase flow of brine and gas through saline media. *Transport in porous media*, 15(3):271–293, 1994a.
- S Olivella, J Carrera, A Gens, and EE Alonso. Porosity variations in saline media caused by temperature gradients coupled to multiphase flow and dissolution/precipitation. *Transport in Porous Media*, 25(1):1–25, 1996a.
- Sebastià Olivella, A Gens, J Carrera, and EE Alonso. Numerical formulation for a simulator (code.bright) for the coupled analysis of saline media. *Engineering computations*, 13(7): 87–112, 1996b.
- S Olivella and A Gens. Vapour transport in low permeability unsaturated soils with capillary effects. *Transport in Porous Media*, 40(2):219–241, 2000.
- L Anand and M Kothari. A computational procedure for rate-independent crystal plasticity. *Journal of the Mechanics and Physics of Solids*, 44(4):525–558, 1996.
- James R Rice. Inelastic constitutive relations for solids: an internal-variable theory and its application to metal plasticity. *Journal of the Mechanics and Physics of Solids*, 19(6): 433–455, 1971.

- D Peirce, RJ Asaro, and A Needleman. An analysis of nonuniform and localized deformation in ductile single crystals. *Acta metallurgica*, 30(6):1087–1119, 1982.
- KS Havner. *Finite plastic deformation of crystalline solids*. Cambridge University Press, 1992.
- Morton E Gurtin, Eliot Fried, and Lallit Anand. *The mechanics and thermodynamics of continua*. Cambridge University Press, 2010.
- E.A. de Souza Neto, D. Peric, and D.R.J. Owen. *Computational methods for plasticity: theory and applications*. John Wiley & Sons, 2011.
- Alberto M Cuitino and Michael Ortiz. Computational modelling of single crystals. *Modelling and Simulation in Materials Science and Engineering*, 1(3):225, 1993.
- Ronaldo I Borja and Jon R Wren. Discrete micromechanics of elastoplastic crystals. *International Journal for Numerical Methods in Engineering*, 36(22):3815–3840, 1993.
- Christian Miehe and Jörg Schröder. A comparative study of stress update algorithms for rate-independent and rate-dependent crystal plasticity. *International Journal for Numerical Methods in Engineering*, 50(2):273–298, 2001.
- Ronaldo I Borja and Helia Rahmani. Computational aspects of elasto-plastic deformation in polycrystalline solids. *Journal of Applied Mechanics*, 79(3):031024, 2012.
- Martin Tjioe and Ronaldo I Borja. On the pore-scale mechanisms leading to brittle and ductile deformation behavior of crystalline rocks. *International Journal for Numerical and Analytical Methods in Geomechanics*, 39(11):1165–1187, 2015.
- C Miehe, F Welschinger, and M Hofacker. Thermodynamically consistent phase-field models of fracture: Variational principles and multi-field fe implementations. *International Journal for Numerical Methods in Engineering*, 83(10):1273–1311, 2010a.
- Christian Miehe, Martina Hofacker, and Fabian Welschinger. A phase field model for rate-independent crack propagation: Robust algorithmic implementation based on operator splits. *Computer Methods in Applied Mechanics and Engineering*, 199(45):2765–2778, 2010b.
- JD Clayton and J Knap. Phase field modeling of twinning in indentation of transparent crystals. *Modelling and Simulation in Materials Science and Engineering*, 19(8):085005, 2011.
- JD Clayton and J Knap. Phase field modeling of directional fracture in anisotropic polycrystals. *Computational Materials Science*, 98:158–169, 2015a.
- Seon Hong Na, Wai Ching Sun, Mathew D Ingraham, and Hongkyu Yoon. Effects of spatial heterogeneity and material anisotropy on the fracture pattern and macroscopic effective toughness of mancoshale in brazilian tests. *Journal of Geophysical Research: Solid Earth*, 2017a.
- M Koslowski and M Ortiz. A multi-phase field model of planar dislocation networks. *Modelling and Simulation in Materials Science and Engineering*, 12(6):1087, 2004.
- T Takaki, Y Hisakuni, T Hirouchi, A Yamanaka, and Y Tomita. Multi-phase-field simulations for dynamic recrystallization. *Computational Materials Science*, 45(4):881–888, 2009.

- Ingo Steinbach. Phase-field models in materials science. *Modelling and simulation in materials science and engineering*, 17(7):073001, 2009.
- Bin Li, Christian Peco, Daniel Millán, Irene Arias, and Marino Arroyo. Phase-field modeling and simulation of fracture in brittle materials with strongly anisotropic surface energy. *International Journal for Numerical Methods in Engineering*, 102(3-4): 711–727, 2015.
- Magnus Ekh, Robert Lillbacka, and Kenneth Runesson. A model framework for anisotropic damage coupled to crystal (visco) plasticity. *International Journal of Plasticity*, 20(12):2143–2159, 2004.
- René de Borst and Clemens V Verhoosel. Gradient damage vs phase-field approaches for fracture: Similarities and differences. *Computer Methods in Applied Mechanics and Engineering*, 312:78–94, 2016a.
- Usik Lee, George A Lesieutre, and Lei Fang. Anisotropic damage mechanics based on strain energy equivalence and equivalent elliptical microcracks. *International Journal of Solids and Structures*, 34(33-34):4377–4397, 1997.
- JC Simo and JW Ju. On continuum damage-elastoplasticity at finite strains. *Computational Mechanics*, 5(5):375–400, 1989.
- Bernard Budiansky and Tai Te Wu. Theoretical prediction of plastic strains of polycrystals. Technical report, Havard University Cambridge Mass, 1961.
- C Barry Carter and M Grant Norton. *Ceramic materials: science and engineering*. Springer Science & Business Media, 2007.
- Lallit Anand and Morton E Gurtin. Thermal effects in the superelasticity of crystalline shape-memory materials. *Journal of the Mechanics and Physics of Solids*, 51(6):1015–1058, 2003.
- FT Meissonnier, EP Busso, and NP O’Dowd. Finite element implementation of a generalised non-local rate-dependent crystallographic formulation for finite strains. *International Journal of Plasticity*, 17(4):601–640, 2001.
- WaiChing Sun. A stabilized finite element formulation for monolithic thermo-hydro-mechanical simulations at finite strain. *International Journal for Numerical Methods in Engineering*, 103(11):798–839, 2015a.
- Blaise Bourdin, Gilles A Francfort, and Jean-Jacques Marigo. The variational approach to fracture. *Journal of elasticity*, 91(1):5–148, 2008a.
- Bernd Schmidt, Fernando Fraternali, and Michael Ortiz. Eigenfracture: an eigendeformation approach to variational fracture. *Multiscale Modeling & Simulation*, 7(3): 1237–1266, 2009.
- Kun Wang and WaiChing Sun. A unified variational eigen-erosion framework for interacting brittle fractures and compaction bands in fluid-infiltrating porous media. *Computer Methods in Applied Mechanics and Engineering*, 318:1–32, 2017a.
- L De Lorenzis, A McBride, and BD Reddy. Phase-field modelling of fracture in single crystal plasticity. *GAMM-Mitteilungen*, 39(1):7–34, 2016.

- John D Bredehoeft, AW England, DB Stewart, NJ Trask, and IJ Winograd. *Geologic disposal of high-level radioactive wastes: Earth-science perspectives*. US Department of the Interior, US Geological Survey, 1978.
- Morton E Gurtin. Generalized ginzburg-landau and cahn-hilliard equations based on a microforce balance. *Physica D: Nonlinear Phenomena*, 92(3-4):178–192, 1996a.
- Michael Johns Borden. *Isogeometric analysis of phase-field models for dynamic brittle and ductile fracture*. PhD thesis, 2012.
- Marreddy Ambati, Tymofiy Gerasimov, and Laura De Lorenzis. Phase-field modeling of ductile fracture. *Computational Mechanics*, 55(5):1017–1040, 2015.
- J.C. Simo and C. Miehe. Associative coupled thermoplasticity at finite strains: formulation, numerical analysis and implementation. *Computer Methods in Applied Mechanics and Engineering*, 98(1):41–104, 1992.
- Rohan Abeyaratne, Kim Sang-Joo, and James K Knowles. A one-dimensional continuum model for shape-memory alloys. *International journal of solids and structures*, 31(16): 2229–2249, 1994.
- VA Lubarda. On thermodynamic potentials in linear thermoelasticity. *International Journal of Solids and Structures*, 41(26):7377–7398, 2004.
- Qiang Yang, Laurent Stainier, and Michael Ortiz. A variational formulation of the coupled thermo-mechanical boundary-value problem for general dissipative solids. *Journal of the Mechanics and Physics of Solids*, 54(2):401–424, 2006.
- Michaël Peigney and Jean-Philippe Seguin. An incremental variational approach to coupled thermo-mechanical problems in anelastic solids. application to shape-memory alloys. *International Journal of Solids and Structures*, 50(24):4043–4054, 2013.
- Christian Miehe, M Hofacker, L-M Schaezel, and F Aldakheel. Phase field modeling of fracture in multi-physics problems. part ii. coupled brittle-to-ductile failure criteria and crack propagation in thermo-elastic-plastic solids. *Computer Methods in Applied Mechanics and Engineering*, 294:486–522, 2015a.
- Timo Heister, Mary F Wheeler, and Thomas Wick. A primal-dual active set method and predictor-corrector mesh adaptivity for computing fracture propagation using a phase-field approach. *Computer Methods in Applied Mechanics and Engineering*, 290: 466–495, 2015.
- Michael J Borden, Thomas JR Hughes, Chad M Landis, Amin Anvari, and Isaac J Lee. A phase-field formulation for fracture in ductile materials: Finite deformation balance law derivation, plastic degradation, and stress triaxiality effects. *Computer Methods in Applied Mechanics and Engineering*, 312:130–166, 2016a.
- Alexander Bartels, Thorsten Bartel, M Canadija, and Jörn Mosler. On the thermomechanical coupling in dissipative materials: a variational approach for generalized standard materials. *Journal of the Mechanics and Physics of Solids*, 82:218–234, 2015.
- M Canadija and Jörn Mosler. On the thermomechanical coupling in finite strain plasticity theory with non-linear kinematic hardening by means of incremental energy minimization. *International Journal of Solids and Structures*, 48(7):1120–1129, 2011.

- Jean Lemaitre. How to use damage mechanics. *Nuclear engineering and design*, 80(2): 233–245, 1984.
- JC Simo and JW Ju. Strain-and stress-based continuum damage models—ii. computational aspects. *International journal of solids and structures*, 23(7):841–869, 1987.
- JW Ju. On energy-based coupled elastoplastic damage theories: constitutive modeling and computational aspects. *International Journal of Solids and structures*, 25(7):803–833, 1989.
- René de Borst, Jerzy Pamin, and Marc GD Geers. On coupled gradient-dependent plasticity and damage theories with a view to localization analysis. *European Journal of Mechanics-A/Solids*, 18(6):939–962, 1999.
- Olivier Coussy. *Poromechanics*. John Wiley & Sons, 2004.
- W. Sun. A unified method to predict diffuse and localized instabilities in sands. *Geomechanics and Geoengineering*, 8(2):65–75, 2013a.
- W. Sun, J.T. Ostien, and A.G. Salinger. A stabilized assumed deformation gradient finite element formulation for strongly coupled poromechanical simulations at finite strain. *International Journal for Numerical and Analytical Methods in Geomechanics*, 37(16): 2755–2788, 2013a.
- K. Wang and W. Sun. Anisotropy of a tensorial bishop’s coefficient for wetted granular materials. *Journal of Engineering Mechanics*, page B4015004, 2015.
- S. Na and W. Sun. Wave propagation and strain localization in a fully saturated softening porous medium under the non-isothermal conditions. *International Journal for Numerical and Analytical Methods in Geomechanics*, 40(10):1485–1510, 2016. doi: 10.1002/nag.2505.
- SeonHong Na and WaiChing Sun. Computational thermo-hydro-mechanics for multi-phase freezing and thawing porous media in the finite deformation range. *Computer Methods in Applied Mechanics and Engineering*, 318:667–700, 2017a.
- John P Hirth and Jens Lothe. *Theory of dislocations*. John Wiley & Sons, 1982.
- John B Wachtman, W Roger Cannon, and M John Matthewson. *Mechanical properties of ceramics*. John Wiley & Sons, 2009.
- Milo A Durand. The temperature variation of the elastic moduli of nacl, kcl and mgo. *Physical Review*, 50(5):449, 1936.
- Richard A Bartels and DE Schuele. Pressure derivatives of the elastic constants of nacl and kcl at 295 k and 195 k. *Journal of Physics and Chemistry of Solids*, 26(3):537–549, 1965.
- Francis Birch. Finite strain isotherm and velocities for single-crystal and polycrystalline nacl at high pressures and 300 k. *Journal of Geophysical Research: Solid Earth*, 83(B3): 1257–1268, 1978.
- Warren Carl Oliver and George Mathews Pharr. An improved technique for determining hardness and elastic modulus using load and displacement sensing indentation experiments. *Journal of materials research*, 7(6):1564–1583, 1992.

- Hanen Amor, Jean-Jacques Marigo, and Corrado Maurini. Regularized formulation of the variational brittle fracture with unilateral contact: numerical experiments. *Journal of the Mechanics and Physics of Solids*, 57(8):1209–1229, 2009a.
- Weidong Qi and Albrecht Bertram. Anisotropic continuum damage modeling for single crystals at high temperatures. *International Journal of Plasticity*, 15(11):1197–1215, 1999.
- Wolfgang R Wawersik and David Henry Zeuch. *Creep and Creep Modeling of Three Domal Salts: A Comprehensive Update*, volume 84. Sandia National Laboratories, 1984.
- Juan C Simo and Thomas JR Hughes. *Computational inelasticity*, volume 7. Springer Science & Business Media, 2006.
- Jihoon Kim, Hamdi A Tchelepi, Ruben Juanes, et al. Stability, accuracy and efficiency of sequential methods for coupled flow and geomechanics. In *SPE reservoir simulation symposium*. Society of Petroleum Engineers, 2009.
- J.A. White, N. Castelletto, and H.A. Tchelepi. Block-partitioned solvers for coupled poromechanics: A unified framework. *Computer Methods in Applied Mechanics and Engineering*, 303:55–74, 2016a.
- K. Wang and W. Sun. A semi-implicit discrete-continuum coupling method for porous media based on the effective stress principle at finite strain. *Computer Methods in Applied Mechanics and Engineering*, 304:546–583, 2016b.
- K. Wang and W. Sun. A semi-implicit micropolar discrete-to-continuum method for granular materials. In *Proceedings of European Congress on Computational Methods in Applied Science and Engineering*, Papadarakakis M, Papadopoulos V, Stefanou G, Plevris V (eds.), June, Crete Island, pages 5–10, 2016c.
- D Tromans and JA Meech. Fracture toughness and surface energies of minerals: theoretical estimates for oxides, sulphides, silicates and halides. *Minerals Engineering*, 15(12):1027–1041, 2002.
- HR Wenk, G Canova, A Molinari, and H Mecking. Texture development in halite: comparison of taylor model and self-consistent theory. *Acta metallurgica*, 37(7):2017–2029, 1989.
- W. Bangerth, R. Hartmann, and G. Kanschat. deal. ii—a general-purpose object-oriented finite element library. *ACM Transactions on Mathematical Software (TOMS)*, 33(4):24, 2007a.
- W. Bangerth, T. Heister, L. Heltai, G. Kanschat, M. Kronbichler, M. Maier, B. Turcksin, and T.D. Young. The deal. ii library. *Version*, 8:1–5, 2013.
- C. Burstedde, L.C. Wilcox, and O. Ghattas. p4est: Scalable algorithms for parallel adaptive mesh refinement on forests of octrees. *SIAM Journal on Scientific Computing*, 33(3):1103–1133, 2011a.
- M.A. Heroux and J.M. Willenbring. A new overview of the trinos project. *Scientific Programming*, 20(2):83–88, 2012a.
- J. Choo and R.I. Borja. Stabilized mixed finite elements for deformable porous media with double porosity. *Computer Methods in Applied Mechanics and Engineering*, 293:131–154, 2015a.

- J.A. White and R.I. Borja. Block-preconditioned newton–krylov solvers for fully coupled flow and geomechanics. *Computational Geosciences*, 15(4):647–659, 2011a.
- J.A. White and R.I. Borja. Stabilized low-order finite elements for coupled solid-deformation/fluid-diffusion and their application to fault zone transients. *Computer Methods in Applied Mechanics and Engineering*, 197(49):4353–4366, 2008a.
- Ronaldo I Borja and Jinhyun Choo. Cam-clay plasticity, part viii: A constitutive framework for porous materials with evolving internal structure. *Computer Methods in Applied Mechanics and Engineering*, 309:653–679, 2016a.
- Mervyn S Paterson and Teng-fong Wong. *Experimental rock deformation-the brittle field*. Springer Science & Business Media, 2005.
- YJ Wei and Lallit Anand. Grain-boundary sliding and separation in polycrystalline metals: application to nanocrystalline fcc metals. *Journal of the Mechanics and Physics of Solids*, 52(11):2587–2616, 2004.
- Koo Hyun Nam, Il H Park, and Seung Hwan Ko. Patterning by controlled cracking. *Nature*, 485(7397):221, 2012.
- Brian M Adams, WJ Bohnhoff, KR Dalbey, JP Eddy, MS Eldred, DM Gay, K Haskell, Patricia D Hough, and LP Swiler. Dakota, a multilevel parallel object-oriented framework for design optimization, parameter estimation, uncertainty quantification, and sensitivity analysis: version 5.0 user’s manual. *Sandia National Laboratories, Tech. Rep. SAND2010-2183*, 2009.
- Guillaume Desbois, Janos L. Urai, and Johannes H.P. de Bresser. Fluid distribution in grain boundaries of natural fine-grained rock salt deformed at low differential stress (Qom Kuh salt fountain, central Iran): Implications for rheology and transport properties. *Journal of Structural Geology*, 43:128–143, 2012c.
- K. Collins and A. McGown. The form and function of microfabric features in a variety of natural soils. *Géotechnique*, 24(2):223–254, 1974.
- Waloddi Weibull. A statistical distribution function of wide applicability. *Journal of Applied Mechanics*, 18(3):293–297, 1951.
- P Habib, J Bernaix, et al. The fissuration of rocks. In *1st International Society for Rock Mechanics Congress*. International Society for Rock Mechanics, 1966.
- S. Forest. Nonlinear regularization operators as derived from the micromorphic approach to gradient elasticity, viscoplasticity and damage. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Science*, 472(2188):20150755, 2016.
- C Miehe, S Teichtmeister, and F Aldakheel. Phase-field modelling of ductile fracture: A variational gradient-extended plasticity-damage theory and its micromorphic regularization. *Phil. Trans. R. Soc. A*, 374(2066):20150170, 2016.
- Samuel Forest, JR Mayeur, and DL McDowell. Micromorphic crystal plasticity. *Handbook of Nonlocal Continuum Mechanics for Materials and Structures*, pages 1–44, 2018.
- KC Bennett, RA Regueiro, and DJ Luscher. Anisotropic finite hyper-elastoplasticity of geomaterials with Drucker-Prager/Cap type constitutive model formulation. *International Journal of Plasticity*, accepted.

- Anna Stankiewicz and Jerzy Pamin. Gradient-enhanced Cam-clay model in simulation of strain localization in soil. *Foundations of Civil and Environmental Engineering*, 7:293–318, 2006.
- O.C. Zienkiewicz, A.H.C. Chan, M. Pastor, B.A. Schrefler, and T. Shiomi. *Computational geomechanics*. Wiley Chichester, 1999.
- WaiChing Sun, Qiushi Chen, and Jakob T Ostien. Modeling the hydro-mechanical responses of strip and circular punch loadings on water-saturated collapsible geomaterials. *Acta Geotechnica*, 9(5):903–934, 2014.
- Guglielmo Scovazzi, Brian Carnes, Xianyi Zeng, and Simone Rossi. A simple, stable, and accurate linear tetrahedral finite element for transient, nearly, and fully incompressible solid dynamics: a dynamic variational multiscale approach. *International Journal for Numerical Methods in Engineering*, 106(10):799–839, 2016.
- Kun Wang and WaiChing Sun. A multiscale multi-permeability poroplasticity model linked by recursive homogenizations and deep learning. *Computer Methods in Applied Mechanics and Engineering*, 334:337–380, 2018.
- WaiChing Sun and José E Andrade. Diffuse bifurcations of porous media under partially drained conditions. In *Multiscale and Multiphysics Processes in Geomechanics*, pages 61–64. Springer, 2011.
- WaiChing Sun. A unified method to predict diffuse and localized instabilities in sands. *Geomechanics and Geoengineering*, 8(2):65–75, 2013b.
- Andreas Krischok and Christian Linder. On the enhancement of low-order mixed finite element methods for the large deformation analysis of diffusion in solids. *International Journal for Numerical Methods in Engineering*, 106(4):278–297, 2016a.
- A.J.L. Crook, J.-g. Yu, and S.M. Willson. Development of an orthotropic 3D elastoplastic material model for shale. In *SPE/ISRM Rock Mechanics Conference*. Society of Petroleum Engineers, 2002.
- Samuel Forest. Micromorphic approach for gradient elasticity, viscoplasticity, and damage. *Journal of Engineering Mechanics*, 135(3):117–131, 2009.
- Fadi Aldakheel and Christian Miehe. Coupled thermomechanical response of gradient plasticity. *International Journal of Plasticity*, 91:1–24, 2017.
- Andrew Schofield and Peter Wroth. *Critical state soil mechanics*, volume 310. McGraw-Hill London, 1968.
- David Muir Wood. *Soil behaviour and critical state soil mechanics*. Cambridge university press, 1990.
- Nabil Abboud and Guglielmo Scovazzi. Elastoplasticity with linear tetrahedral elements: A variational multiscale method. *International Journal for Numerical Methods in Engineering*, 115(8):913–955, 2018.
- S Reese, HR Bayat, and S Wulfinghoff. On an equivalence between a discontinuous galerkin method and reduced integration with hourglass stabilization for finite elasticity. *Computer Methods in Applied Mechanics and Engineering*, 325:175–197, 2017.

- Jiahao Cheng, Ahmad Shahba, and Somnath Ghosh. Stabilized tetrahedral elements for crystal plasticity finite element analysis overcoming volumetric locking. *Computational Mechanics*, 57(5):733–753, 2016.
- M. Itskov. On the theory of fourth-order tensors and their applications in computational mechanics. *Computer Methods in Applied Mechanics and Engineering*, 189(2):419–438, 2000.
- R.I. Borja and C. Tamagnini. Cam-clay plasticity, part III: Extension of the infinitesimal model to include finite strains. *Computer Methods in Applied Mechanics and Engineering*, 155(1-2):73–95, 1998.
- L. J. Walpole. Fourth-rank tensors of the thirty-two crystal classes: multiplication tables. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 391(1800):149–179, 1984.
- K.H. Roscoe and A. Schofield. Mechanical behaviour of an idealized ‘wet’ clay. In *Proc. 3rd Eur. Conf. Soil Mech. Wiesbaden, 1963*, volume 1, pages 47–54, 1963.
- K Hashiguchi and M Ueno. From hvorslev’s failure criterion to failure condition. In *Proc. Intern. Conf. Soil Mech. and Found. Eng., 9th., Tokyo*, volume 50, pages 123–126, 1977.
- R. Butterfield. A natural compression law for soils (an advance on e -log p'). *Géotechnique*, 29(4), 1979.
- David M Potts, Lidija Zdravkovic, and Lidija Zdravković. *Finite element analysis in geotechnical engineering: application*, volume 2. Thomas Telford, 2001.
- F. Hashagen and R. de Borst. Enhancement of the Hoffman yield criterion with an anisotropic hardening model. *Computers & Structures*, 79(6):637–651, 2001.
- Daniele Versino and Kane C Bennett. Generalized radial-return mapping algorithm for anisotropic von mises plasticity framed in material eigenspace. *International Journal for Numerical Methods in Engineering*, 116(3):202–222, 2018.
- J.C. Simo. Numerical analysis and simulation of plasticity. *Handbook of Numerical Analysis*, pages 183–499, 1998.
- K. Weinberg, A. Mota, and M. Ortiz. A variational constitutive model for porous metal plasticity. *Computational Mechanics*, 37(2):142–152, 2006.
- J.C. Simo and T.J.R. Hughes. *Computational Inelasticity*. Springer-Verlag New York, 1998.
- Christian Miehe, Fadi Aldakheel, and Steffen Mauthe. Mixed variational principles and robust finite element implementations of gradient plasticity at small strains. *International Journal for numerical methods in engineering*, 94(11):1037–1074, 2013.
- F. Aldakheel. Micromorphic approach for gradient-extended thermo-elastic–plastic solids in the logarithmic strain space. *Continuum Mechanics and Thermodynamics*, 26(6):1207–1217, 2017.
- Kun Wang, WaiChing Sun, Simon Salager, SeonHong Na, and Ghonwa Khaddour. Identifying material parameters for a micro-polar plasticity model via x-ray micro-computed tomographic (ct) images: lessons learned from the curve-fitting exercises. *International Journal for Multiscale Computational Engineering*, 14(4), 2016.

- J.D. Clayton and J. Knap. Phase field modeling of directional fracture in anisotropic polycrystals. *Computational Materials Science*, 98:158–169, 2015b.
- S. Teichtmeister, D. Kienle, F. Aldakheel, and M.A. Keip. Phase field modeling of fracture in anisotropic brittle solids. *International Journal of Non-Linear Mechanics*, 97:1–21, 2017a.
- Eric C Bryant and WaiChing Sun. A mixed-mode phase field fracture model in anisotropic rocks with consistent kinematics. *Computer Methods in Applied Mechanics and Engineering*, 342:561–584, 2018.
- M. Ortiz and L. Stainier. The variational formulation of viscoplastic constitutive updates. *Computer Methods in Applied Mechanics and Engineering*, 171(3-4):419–444, 1999.
- F. Armero and J.C. Simo. A new unconditionally stable fractional step method for non-linear coupled thermomechanical problems. *International Journal for Numerical Methods in Engineering*, 35(4):737–766, 1992.
- MF Wheeler, T Wick, and W Wollner. An augmented-lagrangian method for the phase-field approach for pressurized fractures. *Computer Methods in Applied Mechanics and Engineering*, 271:69–85, 2014.
- Christian Miehe, M Hofacker, L-M Schänzel, and Fadi Aldakheel. Phase field modeling of fracture in multi-physics problems. part ii. coupled brittle-to-ductile failure criteria and crack propagation in thermo-elastic–plastic solids. *Computer Methods in Applied Mechanics and Engineering*, 294:486–522, 2015b.
- J. Choo and W. Sun. Coupled phase-field and plasticity modeling of geological materials: From brittle fracture to ductile flow. *Computer Methods in Applied Mechanics and Engineering*, 2017.
- E.A. de Souza Neto, D. Peric, and D.R.J. Owen. *Computational Methods for Plasticity*. John Wiley & Sons, Ltd, 2008.
- Wolfgang Bangerth, R Hartmann, and Guido Kanschat. deal . II—A General-Purpose Object-Oriented Finite Element Library. 33(4):1–27, 2007b.
- Jinhyun Choo, Joshua A White, and Ronaldo I Borja. Hydromechanical modeling of unsaturated flow in double porosity media. *International Journal of Geomechanics*, 16(6): D4016002, 2016a.
- R.I. Borja. Conditions for instabilities in collapsible solids including volume implosion and compaction banding. *Acta Geotechnica*, 1(2):107–122, 2006a.
- A. Mota, Q. Chen, J.W. Foulk, J.T. Ostien, and Z. Lai. A Cartesian parametrization for the numerical analysis of material instability. *International Journal for Numerical Methods in Engineering*, 108(2):156–180, 2016.
- Conservation of historic stone buildings and monuments*. The National Academies Press, Washington, D.C., 1982.
- Nicholas Tsui, Robert J. Flatt, and George W. Scherer. Crystallization damage by sodium sulfate. *Journal of Cultural Heritage*, 4(2):109–115, 2003.

- Matthieu Angeli, Jean Philippe Bigas, David Benavente, Beatriz Menéndez, Ronan Hébert, and Christian David. Salt crystallization in pores: Quantification and estimation of damage. *Environmental Geology*, 52(2):187–195, 2007.
- Rosa Maria Espinosa-Marzal and George W. Scherer. Mechanisms of damage by salt. *Geological Society, London, Special Publications*, 331(1):61–77, 2010.
- Michael Steiger, A. Elena Charola, and Katja Sterflinger. Weathering and deterioration. In *Stone in Architecture*, pages 225–316. Springer, 2014.
- Robert J Flatt, Francesco Caruso, Asel Maria Aguilar Sanchez, and George W. Scherer. Chemo-mechanics of salt damage in stone. *Nature communications*, 5:4823, 2014.
- Ingo Sass and Ulrich Burbaum. Damage to the historic town of Staufen (Germany) caused by geothermal drillings through anhydrite-bearing formations. *Acta Carsologica*, 39(2): 233–245, 2010.
- K. Serafeimidis and G. Anagnostou. On the time-development of sulphate hydration in anhydritic swelling rocks. *Rock Mechanics and Rock Engineering*, 46(3):619–634, 2013.
- E E Alonso and A Ramon. Massive sulfate attack to cement-treated railway embankments. *Geotechnique*, 63(10):857–870, 2013.
- Peter B. Kelemen and Jürg Matter. In situ carbonation of peridotite for CO₂ storage. *Proceedings of the National Academy of Sciences of the United States of America*, 105(45): 17295–17300, 2008.
- George W. Scherer. Crystallization in pores. *Cement and Concrete Research*, 29(8):1347–1358, 1999.
- Carl W. Correns and W. Steinborn. Experimente zur Messung und Erklärung der sogenannten Kristallisationskraft. *Zeitschrift für Kristallographie - Crystalline Materials*, 101: 117–133, 1939.
- Carl W. Correns. Growth and dissolution of crystals under linear pressure. *Discussion of the Faraday Society*, 5:267–271, 1949.
- Michael Steiger. Crystal growth in porous materials—II: Influence of crystal size on the crystallization pressure. *Journal of Crystal Growth*, 282(3-4):470–481, 2005b.
- Robert J. Flatt and George W. Scherer. Thermodynamics of crystallization stresses in DEF. *Cement and Concrete Research*, 38(3):325–336, 2008.
- Peter B. Kelemen and Greg Hirth. Reaction-driven cracking during retrograde metamorphism: Olivine hydration and carbonation. *Earth and Planetary Science Letters*, 345-348: 81–89, 2012.
- J. Desarnaud, D. Bonn, and N. Shahidzadeh. The pressure induced by salt crystallization in confinement. *Scientific Reports*, 6(1):30856, 2016.
- Olivier Coussy. Deformation and stress from in-pore drying-induced crystallization of salt. *Journal of the Mechanics and Physics of Solids*, 54(8):1517–1547, 2006.

- Marcin Koniorczyk and Dariusz Gawin. Modelling of salt crystallization in building materials with microstructure - Poromechanical approach. *Construction and Building Materials*, 36:860–873, 2012.
- Ronaldo I. Borja. Cam-Clay plasticity. Part V: A mathematical framework for three-phase deformation and strain localization analyses of partially saturated porous media. *Computer Methods in Applied Mechanics and Engineering*, 193(48-51):5301–5338, 2004.
- Ronaldo I. Borja. On the mechanical energy and effective stress in saturated and unsaturated porous continua. *International Journal of Solids and Structures*, 43(6):1764–1786, 2006b.
- Ronaldo I. Borja and Azad Koliji. On the effective stress in unsaturated porous continua with double porosity. *Journal of the Mechanics and Physics of Solids*, 57(8):1182–1193, 2009.
- Xiaoyu Song and Ronaldo I. Borja. Mathematical framework for unsaturated flow in the finite deformation range. *International Journal for Numerical Methods in Engineering*, 97: 658–682, 2014.
- Jinhyun Choo, Joshua A. White, and Ronaldo I. Borja. Hydromechanical modeling of unsaturated flow in double porosity media. *International Journal of Geomechanics*, 16(6): D4016002, 2016b.
- Ronaldo I. Borja and Jinhyun Choo. Cam-Clay plasticity, Part VIII: A constitutive framework for porous materials with evolving internal structure. *Computer Methods in Applied Mechanics and Engineering*, 309:653–679, 2016b.
- Ronaldo I. Borja and Joshua A. White. Continuum deformation and stability analyses of a steep hillside slope under rainfall infiltration. *Acta Geotechnica*, 5(1):1–14, 2010a.
- Ronaldo I. Borja, Jinhyun Choo, and Joshua A. White. Rock Moisture Dynamics, preferential flow, and the stability of hillside slopes. In Paolo Gardoni and James M. LaFave, editors, *Multi-hazard Approaches to Civil Infrastructure Engineering*, chapter 20, pages 443–464. Springer, 2016.
- Blaise Bourdin, Gilles A. Francfort, and Jean Jacques Marigo. The variational approach to fracture. *Journal of Elasticity*, 91:5–148, 2008b.
- Hanen Amor, Jean Jacques Marigo, and Corrado Maurini. Regularized formulation of the variational brittle fracture with unilateral contact: Numerical experiments. *Journal of the Mechanics and Physics of Solids*, 57(8):1209–1229, 2009b.
- Christian Miehe, Fabian Welschinger, and Martina Hofacker. Thermodynamically consistent phase-field models of fracture: Variational principles and multi-field FE implementations. *International Journal for Numerical Methods in Engineering*, 82:1273–1311, 2010c.
- Christian Miehe, Martina Hofacker, and Fabian Welschinger. A phase field model for rate-independent crack propagation: Robust algorithmic implementation based on operator splits. *Computer Methods in Applied Mechanics and Engineering*, 199(45-48): 2765–2778, 2010d.

- Michael J. Borden, Clemens V. Verhoosel, Michael A. Scott, Thomas J R Hughes, and Chad M. Landis. A phase-field description of dynamic brittle fracture. *Computer Methods in Applied Mechanics and Engineering*, 217-220:77–95, 2012.
- Zachary A. Wilson, Michael J. Borden, and Chad M. Landis. A phase-field model for fracture in piezoelectric ceramics. *International Journal of Fracture*, 183(2):135–153, 2013.
- Michael J. Borden, Thomas J R Hughes, Chad M. Landis, and Clemens V. Verhoosel. A higher-order phase-field model for brittle fracture: Formulation and analysis within the isogeometric analysis framework. *Computer Methods in Applied Mechanics and Engineering*, 273:100–118, 2014.
- A. Mikelić, M. Wheeler, and T. Wick. A phase-field method for propagating fluid-filled fractures coupled to a surrounding porous medium. *Multiscale Modeling & Simulation*, 13(1):367–398, 2015.
- Christian Miehe and Steffen Mauthe. Phase field modeling of fracture in multi-physics problems. Part III. Crack driving forces in hydro-poro-elasticity and hydraulic fracturing of fluid-saturated porous media. *Computer Methods in Applied Mechanics and Engineering*, 304:619–655, 2015.
- Michael J. Borden, Thomas J R Hughes, Chad M. Landis, Amin Anvari, and Isaac J. Lee. A phase-field formulation for fracture in ductile materials: Finite deformation balance law derivation, plastic degradation, and stress triaxiality effects. *Computer Methods in Applied Mechanics and Engineering*, 312:130–166, 2016b.
- Zachary A. Wilson and Chad M. Landis. Phase-field modeling of hydraulic fracture. *Journal of the Mechanics and Physics of Solids*, 96:264–290, 2016.
- Sanghyun Lee, Andro Mikelić, Mary F. Wheeler, and Thomas Wick. Phase-field modeling of proppant-filled fractures in a poroelastic medium. *Computer Methods in Applied Mechanics and Engineering*, 312:509–541, 2016a.
- Sanghyun Lee, Jacqueline E. Reber, Nicholas W. Hayman, and Mary F. Wheeler. Investigation of wing crack formation with a combined phase-field and experimental approach. *Geophysical Research Letters*, 43(15):7946–7952, 2016b.
- Sanghyun Lee, Mary F. Wheeler, and Thomas Wick. Pressure and fluid-driven fracture propagation in porous media using an adaptive finite element phase field model. *Computer Methods in Applied Mechanics and Engineering*, 305:111–132, 2016c.
- Laura De Lorenzis, Andrew McBride, and B.D. Reddy. Phase-field modelling of fracture in single crystal plasticity. *GAMM-Mitteilungen*, 39(1):7–34, 2016.
- Sanghyun Lee, Mary F. Wheeler, and Thomas Wick. Iterative coupling of flow, geomechanics and adaptive phase-field fracture including level-set crack width approaches. *Journal of Computational and Applied Mathematics*, 314:40–60, 2017.
- Steffen Mauthe and Christian Miehe. Hydraulic fracture in poro-hydro-elastic media. *Mechanics Research Communications*, 80:69–83, 2017.
- Wolfgang Ehlers and Chenyi Luo. A phase-field approach embedded in the Theory of Porous Media for the description of dynamic hydraulic fracturing. *Computer Methods in Applied Mechanics and Engineering*, 315:348–368, 2017.

- David Santillán, Ruben Juanes, and Luis Cueto-Felgueroso. Phase-field model of fluid-driven fracture in elastic media: immersed-fracture formulation and validation with analytical solutions. *Journal of Geophysical Research: Solid Earth*, pages 1–25, 2017.
- S. Na, W. C. Sun, H. Yoon, and M. Ingraham. Effects of elastic heterogeneity on the fracture pattern and macroscopic effective toughness of Mancos Shale in Brazilian Tests. *Journal of Geophysical Research: Solid Earth*, 122(8):6202–6230, 2017b.
- T. Cajuhi, L. Sanavia, and L. De Lorenzis. Phase-field modeling of fracture in variably saturated porous media. *Computational Mechanics*, 2017. doi: 10.1007/s00466-017-1459-3.
- Jinhyun Choo and WaiChing Sun. Coupled phase-field and plasticity modeling of geological materials: From brittle fracture to ductile flow. *Computer Methods in Applied Mechanics and Engineering*, 330:1–32, 2018a.
- René de Borst and Clemens V. Verhoosel. Gradient damage vs phase-field approaches for fracture: Similarities and differences. *Computer Methods in Applied Mechanics and Engineering*, 312:78–94, 2016b.
- G. Castellazzi, C. Colla, S. De Miranda, G. Formica, E. Gabrielli, L. Molari, and F. Ubertini. A coupled multiphase model for hygrothermal analysis of masonry structures and prediction of stress induced by salt crystallization. *Construction and Building Materials*, 41:717–731, 2013.
- SeonHong Na and WaiChing Sun. Computational thermo-hydro-mechanics for multiphase freezing and thawing porous media in the finite deformation range. *Computer Methods in Applied Mechanics and Engineering*, 318:667–700, 2017b.
- Karl Terzaghi. *Theoretical Soil Mechanics*. John Wiley & Sons, 1943.
- Jinhyun Choo, Young-Hoon Jung, and Choong-Ki Chung. Effect of directional stress history on anisotropy of initial stiffness of cohesive soils measured by bender element tests. *Soils and foundations*, 51(4):737–747, 2011.
- Young-Hoon Jung, Jinhyun Choo, Wanjei Cho, and Choong-Ki Chung. Patterns of nonlinear shear stiffness degradation of reconstituted clay with different stress histories. *Marine Georesources & Geotechnology*, 31(4):309–331, 2013.
- Jinhyun Choo, Young-Hoon Jung, Wanjei Cho, and Choong-Ki Chung. Effect of pre-shear stress path on nonlinear shear stiffness degradation of cohesive soils. *Geotechnical Testing Journal*, 36(2):198–205, 2013.
- Robert J. Flatt, Michael Steiger, and George W. Scherer. A commented translation of the paper by C.W. Correns and W. Steinborn on crystallization pressure. *Environmental Geology*, 52:187–203, 2007.
- M. Th van Genuchten. A closed-form equation for predicting the hydraulic conductivity of unsaturated soils. *Soil Science Society of America Journal*, 44(5):892–898, 1980.
- Rosa Maria Espinosa-Marzal and George W. Scherer. Impact of in-pore salt crystallization on transport properties. *Environmental Earth Sciences*, 69(8):2657–2669, 2013.
- Jinhyun Choo. *Hydromechanical modeling framework for multiscale porous materials*. PhD thesis, Stanford University, 2016.

- Rosa Maria Espinosa-Marzal, Lutz Franke, and Gernod Deckelmann. Phase changes of salts in porous materials: Crystallization, hydration and deliquescence. *Construction and Building Materials*, 22:1758–1773, 2007.
- Morton E. Gurtin. Generalized Cahn-Hilliard equations based on a microforce balance. *Physica D*, 92:178–192, 1996b.
- Morton E. Gurtin. On the plasticity of single crystals: free energy, microforces, plastic strain gradients. *Journal of the Mechanics and Physics of Solids*, 48:989–1036, 2000.
- David L. Henann and Ken Kamrin. Continuum thermomechanics of the nonlocal granular rheology. *International Journal of Plasticity*, 60:145–162, 2014.
- Xiaoxuan Zhang, Andreas Krischok, and Christian Linder. A variational framework to model diffusion induced large plastic deformation and phase field fracture during initial two-phase lithiation of silicon electrodes. *Computer Methods in Applied Mechanics and Engineering*, 312:51–77, 2016.
- A.A. Griffith. The phenomena of rupture and flow in solids. *Philosophical Transactions. Series A*, 221:163–198, 1921.
- Kun Wang and Waiching Sun. A unified variational eigen-erosion framework for interacting brittle fractures and compaction bands in fluid-infiltrating porous media. *Computer Methods in Applied Mechanics and Engineering*, 318:1–32, 2017b.
- Xue Zhang, Chet Vignes, Scott W. Sloan, and Daichao Sheng. Numerical evaluation of the phase-field model for brittle fracture with emphasis on the length scale. *Computational Mechanics*, 59(5):737–752, 2017.
- Jack A Harison, Bobby O Hardin, and Kamyar Mahboub. Fracture toughness of compacted cohesive soils using ring test. *Journal of Geotechnical Engineering*, 120(5):872–891, 1994.
- Z X Zhang. An empirical relation between mode-I fracture toughness and the tensile strength of rock. *International Journal of Rock Mechanics and Mining Sciences*, 39:401–406, 2002.
- J Wang, J Zhu, C Chiu, and H Zhang. Experimental study on fracture toughness and tensile strength of a clay. *Engineering Geology*, 94(1-2):65–75, 2007.
- M.R. Lakshmikantha, P.C. Prat, and A. Ledesma. Discussion on “Experimental study on fracture toughness and tensile strength of a clay [Engineering Geology 94 (2007) 64–75]. *Engineering Geology*, 101:295–296, 2008.
- Ronaldo I. Borja and Joshua A. White. Conservation laws for coupled hydromechanical processes in unsaturated porous media: Theory and implementation. In Lyesse Laloui, editor, *Mechanics of Unsaturated Geomaterials*, chapter 8, pages 185–208. John Wiley & Sons, 2010b.
- Khalid Aziz, Louis Durlofsky, and Hamdi A. Tchelepi. *Notes on Reservoir Simulation*. Stanford University Lecture Notes, 2014.
- Birendra Jha and Ruben Juanes. Coupled multiphase flow and poromechanics: A computational model of pore pressure effects on fault slip and earthquake triggering. *Water Resources Research*, 50, 2014.

- Jihoon Kim, Eric Sonnenthal, and Jonny Rutqvist. A sequential implicit algorithm of chemo-thermo-poro-mechanics for fractured geothermal reservoirs. *Computers and Geosciences*, 76:59–71, 2015.
- Joshua A. White, Nicola Castelletto, and Hamdi A. Tchelepi. Block-partitioned solvers for coupled poromechanics: A unified framework. *Computer Methods in Applied Mechanics and Engineering*, 303:55–74, 2016b.
- Joshua A. White and Ronaldo I. Borja. Block-preconditioned Newton-Krylov solvers for fully coupled flow and geomechanics. *Computational Geosciences*, 15(4):647–659, 2011b.
- Nicola Castelletto, Joshua A. White, and Massimiliano Ferronato. Scalable algorithms for three-field mixed finite element coupled poromechanics. *Journal of Computational Physics*, 327:894–918, 2016.
- J. Kim, H. A. Tchelepi, and R. Juanes. Stability and convergence of sequential methods for coupled flow and geomechanics: Fixed stress and fixed-strain splits. *Computer Methods in Applied Mechanics and Engineering*, 200:1951–1606, 2011.
- Andro Mikelić and Mary F. Wheeler. Convergence of iterative coupling for coupled flow and geomechanics. *Computational Geosciences*, 17:455–461, 2013.
- Catherine Noiriel, François Renard, Mai Linh Doan, and Jean Pierre Gratier. Intense fracturing and fracture sealing induced by mineral growth in porous rocks. *Chemical Geology*, 269(3-4):197–209, 2010.
- Sheng Dai, Hosung Shin, and J. Carlos Santamarina. Formation and development of salt crusts on soil surfaces. *Acta Geotechnica*, 11(5):1103–1109, 2016.
- Jinhyun Choo and Ronaldo I. Borja. Stabilized mixed finite elements for deformable porous media with double porosity. *Computer Methods in Applied Mechanics and Engineering*, 293:131–154, 2015b.
- Carsten Burstedde, Lucas C. Wilcox, and Omar Ghattas. p4est: Scalable algorithms for parallel adaptive mesh refinement on forests of octrees. *SIAM Journal on Scientific Computing*, 33(3):1103–1133, 2011b.
- Michael A. Heroux and James M. Willenbring. A new overview of the Trilinos project. *Scientific Programming*, 20(2):83–88, 2012b.
- Bernard A. Schrefler and Zhan Xiaoyong. A fully coupled model for water flow and airflow in deformable porous media. *Water Resources Research*, 29(1):155–167, 1993.
- G Klubertanz, L Laloui, and L Vulliet. Numerical modeling of unsaturated porous media as a two and three phase medium: A comparison. *Proc. 9th Int. Conf. On Comp. Meth. and Advances in Geomech., IACMAG*, 2:1159–1164, 1997.
- Bernhard A. Schrefler and Roberto Scotta. A fully coupled dynamic model for two-phase fluid flow in deformable porous media. *Computer Methods in Applied Mechanics and Engineering*, 190(24-25):3223–3246, 2001.
- W. Ehlers, T. Graf, and M. Ammann. Deformation and localization analysis of partially saturated soil. *Computer Methods in Applied Mechanics and Engineering*, 193(27-29): 2885–2910, 2004.

- Enlong Liu, Hai Sui Yu, Gang Deng, Jianhai Zhang, and Siming He. Numerical analysis of seepage–deformation in unsaturated soils. *Acta Geotechnica*, 9(6):1045–1058, 2014.
- T. C. Rasmussen, D. D. Evans, P. J. Sheets, and J. H. Blandford. Permeability of apace leap tuff: Borehole and core measurements using water and air. *Water Resources Research*, 29(7):1997–2006, 1993.
- Joshua A. White and Ronaldo I. Borja. Stabilized low-order finite elements for coupled solid-deformation/fluid-diffusion and their application to fault zone transients. *Computer Methods in Applied Mechanics and Engineering*, 197(49-50):4353–4366, 2008b.
- Waiching Sun, Jakob T. Ostien, and Andrew G. Salinger. A stabilized assumed deformation gradient finite element formulation for strongly coupled poromechanical simulations at finite strain. *International Journal for Numerical and Analytical Methods in Geomechanics*, 37(16):2755–2788, 2013b.
- WaiChing Sun. A stabilized finite element formulation for monolithic thermo-hydro-mechanical simulations at finite strain. *International Journal for Numerical Methods in Engineering*, 103(11):798–839, 2015b.
- Andreas Krischok and Christian Linder. On the enhancement of low-order mixed finite element methods for the large deformation analysis of diffusion in solids. *International Journal for Numerical Methods in Engineering*, 106(4):278–297, 2016b.
- Brice Lecampion. Stress-induced crystal preferred orientation in the poromechanics of in-pore crystallization. *Journal of the Mechanics and Physics of Solids*, 58(10):1701–1715, 2010.
- George W. Scherer. Theory of Drying. *Journal of the American Ceramic Society*, 73(1):3–14, 1990.
- S J Kowalski and A Rybicki. Drying stress formation induced by inhomogeneous moisture and temperature distribution. *Transport in Porous Media*, 24:239–248, 1996.
- Stefan J Kowalski. *Thermomechanics of drying processes*. Springer, 2003.
- Kristopher L Kuhlman. Historic testing relevant to disposal of heat-generating waste in salt. Technical report, Sandia National Lab.(SNL-NM), Albuquerque, NM (United States), 2013.
- Laura Blanco Martin, Jonny Rutqvist, and Jens T Birkholzer. Long-term modeling of the thermal–hydraulic–mechanical response of a generic salt repository for heat-generating nuclear waste. *Engineering geology*, 193:198–211, 2015.
- Soheil Ghanbarzadeh, Marc A Hesse, Maša Prodanović, and James E Gardner. Deformation-assisted fluid percolation in rock salt. *Science*, 350(6264):1069–1072, 2015.
- Cheng Zhu, Ahmad Pouya, and Chloé Arson. Micro-macro analysis and phenomenological modelling of salt viscous damage and application to salt caverns. *Rock Mechanics and Rock Engineering*, 48(6):2567–2580, 2015.
- ME Houben, A ten Hove, CJ Peach, and CJ Spiers. Crack healing in rocksalt via diffusion in adsorbed aqueous films: Microphysical modelling versus experiments. *Physics and Chemistry of the Earth, parts A/B/C*, 64:95–104, 2013.

- Paula J Koelemeijer, Colin J Peach, and Christopher J Spiers. Surface diffusivity of cleaved nacl crystals as a function of humidity: Impedance spectroscopy measurements and implications for crack healing in rock salt. *Journal of Geophysical Research: Solid Earth*, 117(B1), 2012.
- RCMW Franssen and CJ Spiers. Deformation of polycrystalline salt in compression and in shear at 250–350 ° c. *Geological Society, London, Special Publications*, 54(1):201–213, 1990.
- Hugh C Heard. Steady-state flow in polycrystalline halite at pressure of 2 kilobars. *Flow and Fracture of Rocks*, 16:191–209, 1972.
- Ahmad Pouya, Cheng Zhu, and Chloé Arson. Micro–macro approach of salt viscous fatigue under cyclic loading. *Mechanics of Materials*, 93:13–31, 2016.
- Shi-Yuan Li and Janos L Urai. Rheology of rock salt for salt tectonics modeling. *Petroleum Science*, 13(4):712–724, 2016.
- VLADISLAV Kruzhanov and B Stöckhert. On the kinetics of elementary processes of pressure solution. *pure and applied geophysics*, 152(4):667–683, 1998.
- Stephen H Hickman and Brian Evans. Kinetics of pressure solution at halite-silica interfaces and intergranular clay films. *Journal of Geophysical Research: Solid Earth*, 100(B7): 13113–13132, 1995.
- Jacques Schott, Oleg S Pokrovsky, and Eric H Oelkers. The link between mineral dissolution/precipitation kinetics and solution chemistry. *Reviews in mineralogy and geochemistry*, 70(1):207–258, 2009.
- Reinier van Noort, Hendrica JM Visser, and Christopher J Spiers. Influence of grain boundary structure on dissolution controlled pressure solution and retarding effects of grain boundary healing. *Journal of Geophysical Research: Solid Earth*, 113(B3), 2008.
- R Van Noort, CJ Spiers, and CJ Peach. Effects of orientation on the diffusive properties of fluid-filled grain boundaries during pressure solution. *Physics and Chemistry of minerals*, 34(2):95–112, 2007.
- Marwan Alkattan, Eric H Oelkers, Jean-Louis Dandurand, and Jacques Schott. Experimental studies of halite dissolution kinetics, 1 the effect of saturation state and the presence of trace metals. *Chemical Geology*, 137(3-4):201–219, 1997.
- Ricardo A Lebensohn, Paul R Dawson, Hartmut M Kern, and Hans-Rudolf Wenk. Heterogeneous deformation and texture development in halite polycrystals: comparison of different modeling approaches and experimental data. *Tectonophysics*, 370(1-4):287–311, 2003.
- S Olivella, J Carrera, A Gens, and EE Alonso. Nonisothermal multiphase flow of brine and gas through saline media. *Transport in porous media*, 15(3):271–293, 1994b.
- Allan F Bower and Emeric Wininger. A two-dimensional finite element method for simulating the constitutive response and microstructure of polycrystals during high temperature plastic deformation. *Journal of the Mechanics and Physics of Solids*, 52(6): 1289–1317, 2004.

- Kwai S Chan, SR Bodner, DE Munson, and AF Fossum. A constitutive model for representing coupled creep, fracture, and healing in rock salt. Technical report, Sandia National Labs., Albuquerque, NM (United States), 1996.
- KS Chan, SR Bodner, and DE Munson. Application of isochronous healing curves in predicting damage evolution in a salt structure. *International Journal of Damage Mechanics*, 9(2):130–153, 2000.
- Zhengmeng Hou. Mechanical and hydraulic behavior of rock salt in the excavation disturbed zone around underground facilities. *International Journal of Rock Mechanics and Mining Sciences*, 40(5):725–738, 2003.
- Cheng Zhu and Chloé Arson. A model of damage and healing coupling halite thermo-mechanical behavior to microstructure evolution. *Geotechnical and Geological Engineering*, 33(2):389–410, 2015b.
- Xianda Shen and Chloé Arson. An isotropic self-consistent homogenization scheme for chemo-mechanical healing driven by pressure solution in halite. *International Journal of Solids and Structures*, 161:96–110, 2019.
- Yang Liu, WaiChing Sun, and Jacob Fish. Determining material parameters for critical state plasticity models based on multilevel extended digital database. *Journal of Applied Mechanics*, 83(1):011003, 2016b.
- Kun Wang, WaiChing Sun, and Qiang Du. A cooperative game for automated learning of elasto-plasticity knowledge graphs and models with ai-guided experimentation. *Computational Mechanics*, pages 1–33, 2019.
- Yujie Wei and Lallit Anand. On micro-cracking, inelastic dilatancy, and the brittle-ductile transition in compact rocks: A micro-mechanical study. *International Journal of Solids and Structures*, 45(10):2785–2798, 2008.
- Peter K Kang, Marco Dentz, Tanguy Le Borgne, and Ruben Juanes. Anomalous transport on regular fracture networks: Impact of conductivity heterogeneity and mixing at fracture intersections. *Physical Review E*, 92(2):022148, 2015.
- Martin Tjioe and Ronaldo I Borja. Pore-scale modeling of deformation and shear band bifurcation in porous crystalline rocks. *International Journal for Numerical Methods in Engineering*, 108(3):183–212, 2016.
- Guang Liu, WaiChing Sun, Steven M Lowinger, ZhenHua Zhang, Ming Huang, and Jun Peng. Coupled flow network and discrete element modeling of injection-induced crack propagation and coalescence in brittle rock. *Acta Geotechnica*, 14(3):843–868, 2019.
- Krishna Garikipati, Lori Bassman, and Michael Deal. A lattice-based micromechanical continuum formulation for stress-driven mass transport in polycrystalline solids. *Journal of the Mechanics and Physics of Solids*, 49(6):1209–1237, 2001.
- Aurélien Villani, Esteban P Busso, Kais Ammar, Samuel Forest, and MGD Geers. A fully coupled diffusional-mechanical formulation: numerical implementation, analytical validation, and effects of plasticity on equilibrium. *Archive of applied mechanics*, 84(9-11):1647–1664, 2014.

- Aurélien Villani, Esteban P Busso, and Samuel Forest. Field theory and diffusion creep predictions in polycrystalline aggregates. *Modelling and Simulation in Materials Science and Engineering*, 23(5):055006, 2015.
- François Renard, Dominique Bernard, Xavier Thibault, and Elodie Boller. Synchrotron 3d microtomography of halite aggregates during experimental pressure solution creep and evolution of the permeability. *Geophysical Research Letters*, 31(7), 2004.
- Chuanqi Liu and WaiChing Sun. Shift boundary material point method: an image-to-simulation workflow for solids of complex geometries undergoing large deformation. *Computational Particle Mechanics*, pages 1–18, 2019.
- Luv Sharma, Ron HJ Peerlings, Pratheek Shanthraj, Franz Roters, and Marc GD Geers. Fft-based interface decohesion modelling by a nonlocal interphase. *Advanced Modeling and Simulation in Engineering Sciences*, 5(1):7, 2018.
- Jinhyun Choo and WaiChing Sun. Cracking and damage from crystallization in pores: Coupled chemo-hydro-mechanics and phase-field modeling. *Computer Methods in Applied Mechanics and Engineering*, 335:347–379, 2018b.
- Morton E Gurtin. Generalized ginzburg-landau and cahn-hilliard equations based on a microforce balance. *Physica D: Nonlinear Phenomena*, 92(3-4):178–192, 1996c.
- Bin Li and Corrado Maurini. Crack kinking in a variational phase-field model of brittle fracture with strongly anisotropic surface energy. *Journal of the Mechanics and Physics of Solids*, 125:502–522, 2019.
- Clifford Truesdell and Walter Noll. The non-linear field theories of mechanics. In *The non-linear field theories of mechanics*, pages 1–579. Springer, 2004.
- Mark Messner, Armand Beaudoin, and Robert Dodds. Consistent crystal plasticity kinematics and linearization for the implicit finite element method. *Engineering Computations*, 32(6):1526–1548, 2015. ISSN 0264-4401.
- Ran Ma and Timothy J Truster. Fft-based homogenization of hypoelastic plasticity at finite strains. *Computer Methods in Applied Mechanics and Engineering*, 2019.
- Nathan R Barton, Athanasios Arsenlis, and Jaime Marian. A polycrystal plasticity model of strain localization in irradiated iron. *Journal of the Mechanics and Physics of Solids*, 61(2):341–351, 2013.
- Thomas Driesner and Christoph A Heinrich. The system h₂o–nacl. part i: Correlation formulae for phase relations in temperature–pressure–composition space from 0 to 1000 c, 0 to 5000 bar, and 0 to 1 xnacl. *Geochimica et Cosmochimica Acta*, 71(20):4880–4901, 2007.
- S Teichtmeister, D Kienle, F Aldakheel, and M-A Keip. Phase field modeling of fracture in anisotropic brittle solids. *International Journal of Non-Linear Mechanics*, 97:1–21, 2017b.
- Jan Zeman, Jaroslav Vondřejc, Jan Novák, and Ivo Marek. Accelerating a FFT-based solver for numerical homogenization of periodic media by conjugate gradients. *Journal of Computational Physics*, 229(21):8065–8071, 2010. ISSN 0021-9991.

- T. W. J. de Geus, J. Vondřejc, J. Zeman, R. H. J. Peerlings, and M. G. D. Geers. Finite strain FFT-based non-linear solvers made simple. *Computer Methods in Applied Mechanics and Engineering*, 318(Supplement C):412–430, 2017. ISSN 0045-7825.
- Julian Kochmann, Stephan Wulfinghoff, Lisa Ehle, Joachim Mayer, Bob Svendsen, and Stefanie Reese. Efficient and accurate two-scale fe-fft-based prediction of the effective material behavior of elasto-viscoplastic polycrystals. *Computational Mechanics*, 61(6): 751–764, 2018.
- Julian Kochmann, Kiran Manjunatha, Christian Gierden, Stephan Wulfinghoff, Bob Svendsen, and Stefanie Reese. A simple and flexible model order reduction method for fft-based homogenization problems using a sparse sampling technique. *Computer Methods in Applied Mechanics and Engineering*, 347:622–638, 2019.
- Nathan R Barton, Joel V Bernier, Ricardo A Lebensohn, and Donald E Boyce. The use of discrete harmonics in direct multi-scale embedding of polycrystal plasticity. *Computer Methods in Applied Mechanics and Engineering*, 283:224–242, 2015.
- Jingzhi Zhu, Long-Qing Chen, Jie Shen, and Veena Tikare. Coarsening kinetics from a variable-mobility cahn-hilliard equation: Application of a semi-implicit fourier spectral method. *Physical Review E*, 60(4):3564, 1999.
- R. Quey, P. R. Dawson, and F. Barbe. Large-scale 3D random polycrystals for the finite element method: Generation, meshing and remeshing. *Computer Methods in Applied Mechanics and Engineering*, 200(17–20):1729–1745, 2011. ISSN 0045-7825.