

Application of Rate-type Variational Formulations and Scalable Linear Solvers to the Thermo-Chemo-Mechanical Modeling of Hydrogels

Akshay B Jeeja, Bjoern Kiefer, Stefan Prüger, Oliver Rheinbach, Friederike Röver

Abstract. In this work, we compare two variational formulations of a chemo-mechanical problem: one as a minimization problem, the other in saddle point form. Our test problem is a mechanically-driven diffusion problem, using a Neo-Hooke energy. Our numerical results demonstrate clear advantages of the minimization approach. We observe experimentally that it yields symmetric positive definite system matrices, allowing the use of the conjugate gradient (CG) method with efficient preconditioning. This leads to noticeably lower computation times compared to GMRES, which is required for the indefinite saddle point system. Additionally, the minimization formulation results in sparser matrices, further reducing computational effort - especially in regimes where sparse direct solvers are still viable. These benefits are expected to extend to parallel domain decomposition methods that rely on sparse direct solvers, locally.

Key words: Variational Methods, Thermo-Chemo-Mechanics, Hydrogels, FROSch

1 INTRODUCTION

Hydrogels are water-swollen three-dimensional polymer networks, that possess a number of favorable properties such as high water absorption capability, tunable mechanical properties, easy functionalization, i.e., gels are sensitive to external stimuli, which makes them very attractive for medical or biomedical applications [1, pp. 453 ff.], [2, 3]. In order to support the development of hydrogel based medical devices like contact lenses, to predict cross-linking kinetics in hydrogels or to simulate the printing of 3D hydrogel scaffolds, a robust and efficient modeling framework is required. In the current contribution, such a modeling framework is based on two variational formulations of a simplified chemo-mechanical boundary value problem, in which the main phenomena of hydrogels are incorporated, namely the mutually coupled

Akshay B Jeeja & Bjoern Kiefer & Stefan Prüger
Technische Universität Bergakademie Freiberg, Institute of Mechanics and Fluid Dynamics, Lampadiusstrasse 4, 09599 Freiberg, Germany, e-mail: stefan.prueger@imfd.tu-freiberg.de

Oliver Rheinbach
Technische Universität Bergakademie Freiberg, Institute of Numerical Mathematics and Optimization, Akademiestrasse 6, 09599 Freiberg, Germany

Oliver Rheinbach & Friederike Röver
Technische Universität Bergakademie Freiberg, University Computing Center (URZ), Bernhard-v.-Cotta Strasse 1, 09599 Freiberg, Germany

solvent diffusion and the swelling-induced deformation of the gel. As the boundary value problem is solved by means of the Finite Element technique, the impact of different iterative solvers and the corresponding preconditioners is examined for the different variational formulations in the sense of a co-design of the model formulation and numerical solvers.

2 VARIATIONAL FORMULATIONS

This contribution employs the variational framework of chemo-mechanics, which couples the mechanical balance relations to mass diffusion, introduced in [4, 5]. In particular, the two dual formulations of mass diffusion, that lead either to a variational minimization or to a variational saddle point structure, are considered and briefly summarized in the subsequent subsections.

2.1 Minimization formulation of the chemo-mechanical problem

The rate-type potential associated with the minimization formulation is introduced as

$$\Pi(\dot{\boldsymbol{\varphi}}, \dot{v}, \mathbf{J}_v) = \int_B \frac{d}{dt} \hat{\psi}(\nabla \boldsymbol{\varphi}, v) dV + \int_B \hat{\phi}(\mathbf{J}_v; \nabla \boldsymbol{\varphi}, v) dV - P_{\text{ext}}^{\varphi}(\dot{\boldsymbol{\varphi}}) - P_{\text{ext}}^{\mu}(\mathbf{J}_v). \quad (1)$$

Herein, the specific free-energy function and the dissipation potential are denoted as $\hat{\psi}$ and $\hat{\phi}$, respectively. While the energy function depends on the material gradient of the deformation $\nabla \boldsymbol{\varphi}$ and the swelling volume fraction v , the state-dependent dissipation potential is a function of the volume flux $\mathbf{J}_v$ and is additionally parameterized by the former two fields. The power of the external forces corresponding to the deformation and the volume flux are given as

$$P_{\text{ext}}^{\varphi}(\dot{\boldsymbol{\varphi}}) = \int_B \mathbf{R}_{\varphi} \cdot \dot{\boldsymbol{\varphi}} dV + \int_{\partial B^T} \bar{\mathbf{T}} \cdot \dot{\boldsymbol{\varphi}} dA \quad P_{\text{ext}}^{\mu}(\mathbf{J}_v) = - \int_{\partial B^{\mu}} \bar{\mu} \underbrace{\mathbf{J}_v \cdot \mathbf{N}}_{H_v} dA. \quad (2)$$

in which the body force vector and the prescribed nominal traction vector are denominated as $\mathbf{R}_{\varphi}$ and $\bar{\mathbf{T}}$, respectively. Furthermore, the prescribed chemical potential and the volume flux projected into the outward normal direction $\mathbf{N}$ are called $\bar{\mu}$ and H_v .

Noting that the choice of the volume flux as the primary field in the diffusion problem renders the balance of solute volume

$$\dot{v} = -\text{Div}[\mathbf{J}_v] \quad (3)$$

an ordinary differential equation, allowing for the elimination of $\dot{v}$ from Eq. (1) and thus yielding a two-field problem. The deformation and the volume flux field that make the reduced rate-type potential stationary, are determined from

$$\{\dot{\boldsymbol{\varphi}}^*, \mathbf{J}_v^*\} = \text{Arg} \left\{ \inf_{\dot{\boldsymbol{\varphi}} \in \mathcal{W}_{\dot{\boldsymbol{\varphi}}}} \inf_{\mathbf{J}_v \in \mathcal{W}_{\mathbf{J}_v}} \Pi(\dot{\boldsymbol{\varphi}}, \mathbf{J}_v) \right\}. \quad (4)$$

The suitable function spaces for the two fields are detailed as

$$\mathcal{W}_{\dot{\boldsymbol{\varphi}}} = \{ \dot{\boldsymbol{\varphi}} \in H^1(\mathcal{B}) \mid \dot{\boldsymbol{\varphi}} = \dot{\boldsymbol{\varphi}} \text{ on } \partial \mathcal{B}^{\varphi} \}, \quad \mathcal{W}_{\mathbf{J}_v} = \{ \mathbf{J}_v \in H(\text{Div}, \mathcal{B}) \mid \mathbf{J}_v \cdot \mathbf{N} = H_v \text{ on } \partial \mathcal{B}^{H_v} \} \quad (5)$$

where both fields have to satisfy the respective Dirichlet boundary conditions along $\partial\mathcal{B}^\varphi$ and $\partial\mathcal{B}^{H_v}$. While the deformation field $\dot{\varphi}$ is taken from the standard Sobolev space [6, p. 47f], i.e. the norm of the field and its first partial derivatives are bounded in an integral sense [7, p.136ff.], the choice for the flux field is less restrictive, as it only requires that the norms of the field and its divergence are bounded [6, p.49f]. The conforming choice of finite element ansatz functions is given in Section 3.1.

2.2 Saddle point formulation of the chemo-mechanical problem

As an alternative to the two-field formulation introduced above, the coupled chemo-mechanical problem can also be cast into the three-field variational formulation

$$\Pi^*(\dot{\varphi}, \dot{v}, \mu) = \int_{\mathcal{B}} \left[\frac{d}{dt} \hat{\psi}(\nabla \varphi, v) - \mu \dot{v} - \hat{\phi}^*(-\nabla \mu; \nabla \varphi, v) \right] dV - P_{\text{ext}}^\varphi(\dot{\varphi}) - P_{\text{ext}}^{H_v}(\mu), \quad (6)$$

in which the dissipation potential $\hat{\phi}$ is replaced by its Legendre transform and the power of the external forces associated with the diffusion problem is redefined as

$$P_{\text{ext}}^{H_v}(\mu) = \int_{\partial\mathcal{B}^{H_v}} \mu \bar{H}_v dA. \quad (7)$$

The state-dependent dissipation potential $\hat{\phi}^*$ emerges as a function of the negative material gradient of the scalar chemical potential with an additional parameterization based on the gradient of the deformation and the swelling volume fraction. Furthermore, the role of the chemical potential and the normal component of the volume flux H_v has changed in the sense that $\bar{H}_v$ is now prescribed as a Neumann boundary condition on the $\partial\mathcal{B}^{H_v}$, while the Neumann boundary condition for the minimization formulation is specified in terms of $\bar{\mu}$ on $\partial\mathcal{B}^\mu$. Once again, primary fields satisfying the underlying balance equations are determined by demanding that the rate-type potential is stationary.

$$\{\dot{\varphi}^*, \dot{v}^*, \mu^*\} = \text{Arg} \left\{ \inf_{\dot{\varphi} \in \mathcal{W}_{\dot{\varphi}}} \inf_{\dot{v} \in L^2} \sup_{\mu \in \mathcal{W}_\mu} \Pi^*(\dot{\varphi}, \dot{v}, \mu) \right\} \quad (8)$$

Here, the deformation field as well as the chemical potential are taken from standard Sobolev space as

$$\mathcal{W}_{\dot{\varphi}} = \{\dot{\varphi} \in H^1(\mathcal{B}) \mid \dot{\varphi} = \dot{\bar{\varphi}} \text{ on } \partial\mathcal{B}^\varphi\}, \quad \mathcal{W}_\mu = \{\mu \in H^1(\mathcal{B}) \mid \mu = \bar{\mu} \text{ on } \partial\mathcal{B}^\mu\}, \quad (9)$$

where the associated Dirichlet boundary conditions have to be fulfilled on $\partial\mathcal{B}^\varphi$ and $\partial\mathcal{B}^\mu$. From the inspection of Eq. (8), it turns out that the minimum of Π^* with respect to $\dot{v}$ can be obtained point-wise for given fields $\dot{\varphi}$ and μ . Thus, a reduced form of Eq. (6) can be constructed that only depends on the deformation and the chemical potential, see [4] for further details. The choice of the conforming finite element ansatz functions and the suitable numerical handling of the local condensation of $\dot{v}$ is given in Section 3.1.

2.3 Constitutive functions

The two variational formulations outlined in Section 2.1 and 2.2 introduce only the dependencies of the specific free-energy function and the dissipation potentials and are therefore valid for a large class of materials. However, this contribution focuses on an isotropic, hyperelastic material model for hydrogels that accounts for a strong two-sided coupling between the deformation and the flux field. Regarding the finite strain kinematics, the multiplicative split of the deformation gradient $\mathbf{F}_d = \mathbf{F} \cdot J_0^{1/3} \mathbf{I}$ is assumed, where $\mathbf{F}_d$ maps from the initial configuration associated with the dry-state of the hydrogel to the current configuration, while J_0 corresponds to a material parameter that characterizes the volume change stemming from the pre-swelling of the initial configuration. The deformation gradient $\mathbf{F}$ and its determinant J specify the mechanical deformation of the pre-swollen hydrogel. A coupled free-energy function of Neo-Hookean type, suitable for compressible material behavior, is specified relative to the pre-swollen configuration as

$$\hat{\psi}(\nabla \boldsymbol{\varphi}, v) = \frac{\gamma}{2J_0} \left[J_0^{2/3} \mathbf{F} : \mathbf{F} - 3 - 2 \ln(JJ_0) \right] + \frac{\lambda}{2J_0} [JJ_0 - 1 - v]^2 + \frac{\alpha}{J_0} \left[v \ln \left(\frac{v}{1+v} \right) + \frac{\chi v}{1+v} \right], \quad (10)$$

in which γ and λ correspond to the shear modulus and the first Lamé constant, while the mixing modulus and the mixing control parameter in the Flory-Rehner type contribution of the energy are denoted as α and χ , respectively. The latter accounts for the mixing of the solvent and the polymer network, where $\chi = 0$ represents an ideal mixture. The scaling factor $\frac{1}{J_0}$ is introduced to enforce the equality of the energy in the initial and the pre-swollen configuration.

The first Piola-Kirchhoff stress and the chemical potential are computed as

$$\mathbf{P} = \frac{\partial \hat{\psi}}{\partial \mathbf{F}} \stackrel{(10)}{=} \frac{\gamma}{J_0^{1/3}} \mathbf{F} + \left[\lambda J [JJ_0 - 1 - v] - \frac{\gamma}{J_0} \right] \mathbf{F}^{-T} \quad (11)$$

and

$$\mu = \frac{\partial \hat{\psi}}{\partial v} \stackrel{(10)}{=} -\frac{\lambda}{J_0} [JJ_0 - 1 - v] + \frac{\alpha}{J_0} \left[\ln \left(\frac{v}{1+v} \right) + \frac{1}{1+v} + \frac{\chi}{(1+v)^2} \right]. \quad (12)$$

As the undeformed state, i.e. $\mathbf{F} = \mathbf{I}$, is assumed to be stress-free, the swelling volume fraction has to be initialized for a given value of the pre-swollen Jacobian J_0 as

$$v_0 = \frac{\gamma}{\lambda} \left[J_0^{-1/3} - \frac{1}{J_0} \right] + J_0 - 1. \quad (13)$$

The state-dependent dissipation potentials corresponding to the minimization and the saddle point formulation are taken as the quadratic forms

$$\hat{\phi}(\mathbf{J}_v; \mathbf{C}, v) = \frac{1}{2Mv} \mathbf{C} : (\mathbf{J}_v \otimes \mathbf{J}_v) \quad \text{and} \quad \hat{\phi}^*(-\nabla \mu; \mathbf{C}, v) = \frac{Mv}{2} \mathbf{C}^{-1} : (-\nabla \mu \otimes -\nabla \mu), \quad (14)$$

which yield the following linear relations between the volume flux and the gradient of the chemical potential, employing the volumetric diffusivity parameter M .

$$-\nabla\mu = \frac{\partial\hat{\phi}}{\partial\mathbf{J}_v} \stackrel{(14)_1}{=} \frac{1}{Mv} \mathbf{C} \cdot \mathbf{J}_v \quad \text{and} \quad \mathbf{J}_v = \frac{\partial\hat{\phi}^*}{\partial(-\nabla\mu)} \stackrel{(14)_2}{=} Mv \mathbf{C}^{-1} \cdot (-\nabla\mu) \quad (15)$$

The additional dependency on the right Cauchy-Green tensor $\mathbf{C}$ or its inverse, is due to the assumption of an isotropic behavior in the current configuration, which is then pulled back to the pre-swollen configuration by means of Nanson's formula. The material parameters are chosen identical to [8].

3 Implementation

3.1 Implementation of the chemo-mechanical problem in deal.II

The variational formulations described in Section 2 are the same as the formulations in [8]. We use our implementation employing the `deal.II` software library [9] Version 9.5.0. For a more detailed description, we refer to [8, Section 3]. In the following, we denote the minimization formulation as *Min* formulation and the saddle point formulation as *SP* formulation to improve the readability. We examine different combinations of conforming approximations. For the *Min* formulation we consider tri-linear (Q_1) Lagrange ansatz functions for the deformation field and lowest order Raviart-Thomas (RT_0) ansatz functions for the flux field $\mathbf{J}_v$. The latter ensures the continuity of the normal trace across the element boundaries. In order to compute the swelling volume fraction, Eq. (3) is integrated numerically by a backward Euler scheme.

We employ Taylor-Hood elements (Q_2Q_1) for the approximation of the primary variables φ and μ in the saddle point formulation. In addition, the computation of the swelling volume fraction v is carried out by means of the interior affine trust-region method [10], which strictly ensures its non-negativity. The nonlinear algebraic systems emerging from the finite element discretization are solved by a standard Newton-Raphson scheme in each load increment. The solution of the linear system of equations arising in the Newton iterations is obtained by the iterative minimal residual method (*MINRES*) and the generalized minimal residual method (*GMRES*) method with the splitting preconditioners Jacobi and SSOR [11] using the implementation provided by `deal.II`. We use the default settings for *GMRES*, therefore the *GMRES* method employs Gram Schmidt with delayed reorthogonalization [12]. In [8], the limitations of sparse direct linear solvers were already shown for the two variational formulations outlined in Section 2.1 and 2.2. Here, we investigate the performance of iterative methods with simple preconditioners. It is well known that these simple preconditioners used here are not scalable. More advanced, scalable preconditioning methods based on domain decomposition methods [13] will be considered in a next step. In [14], we have already successfully applied the parallel overlapping Schwarz preconditioner *FROSch* [15] of the Trilinos software library [16] to the *Min* formulation.

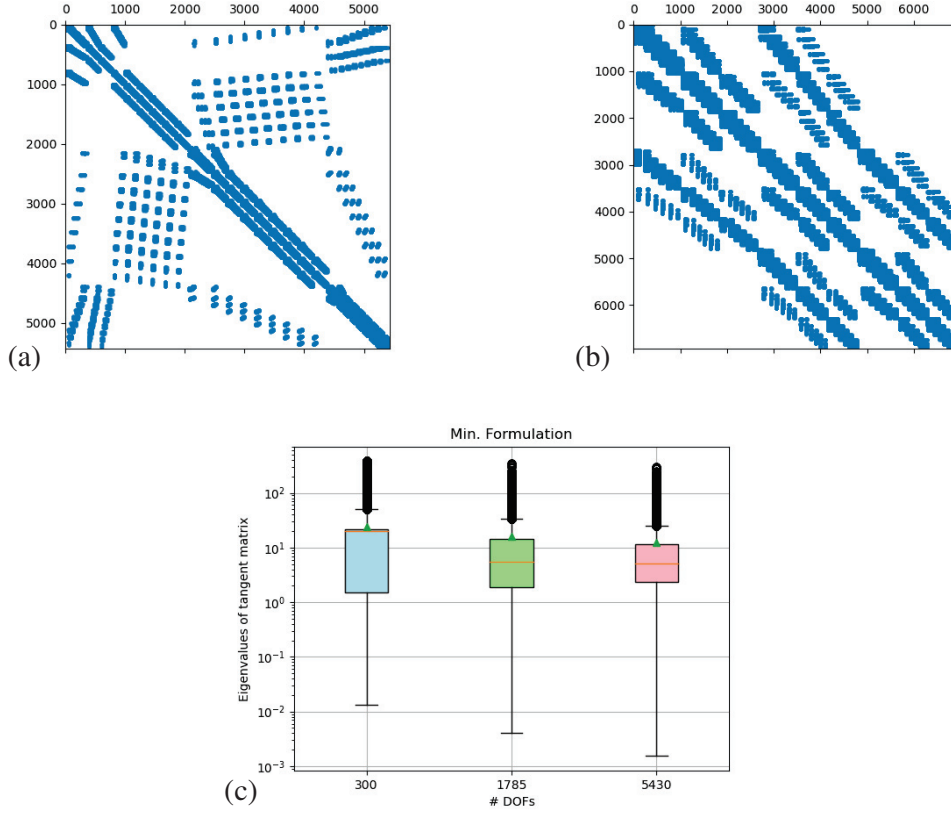


Figure 1: Sparsity pattern of the tangent matrix for the *Min* formulation (a) with 5 430 DOFs and 385 164 non-zero entries and the *SP* formulation (b) with 6 934 DOFs and 1 244 446 non-zero entries. Box plot of the eigenvalues of the tangent stiffness matrices (c) evaluated in the simulation of the mechanically-driven diffusion problem, employing the *Min* formulation, for three different mesh refinements. The smallest eigenvalues are: 0.0131 (300 DOFs), 0.0041 (1785 DOFs), 0.0015 (5430 DOFs).

4 RESULTS

4.1 Comparison of iterative methods in the chemo-mechanical problem

To evaluate the performance of iterative methods for the different variational formulations, we apply our finite element implementation to a mechanically-driven diffusion problem in a cubic domain. The problem considers the indentation of a hydrogel cube with a flat, rigid indenter, where the mechanical stress field leads to a redistribution of the solvent within the sealed domain. It is equivalent to the one described in [8, Section 4]. The same relative stopping criterion of $\|r_k\|/\|r_0\| \leq 10^{-6}$ is used for all iterative solvers, to obtain the solution of the linear system of equations. We set the maximum number of iteration in dependence of the size of the system matrix, such that the maximum number of iterations is four times the number of degrees of freedom. We have observed experimentally that the *Min* formulation yields symmetric posi-

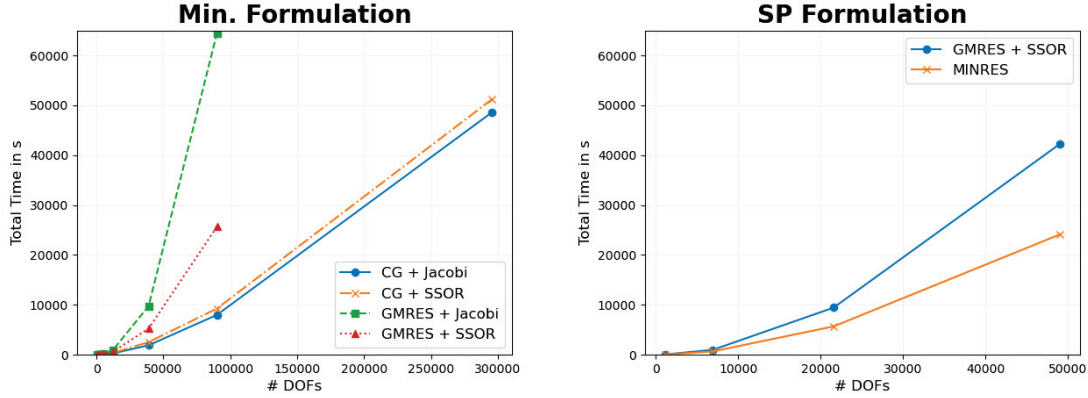


Figure 2: Total Time of the calculations performed with $\Delta t = 0.1$ s. Considered are the *Min* formulation (left) with different iterative methods and preconditioner as well as the *SP* formulation (right) employing GMRES with SSOR and MINRES without any preconditioner.

tive definite tangent matrices for our test problem, see Fig. 1(a,c). The smallest eigenvalues are bounded away from zero, which is expected as the boundary value problem does not contain any intrinsic buckling or snap-back/snap-through instabilities. Therefore, we also consider the conjugate gradient (CG) method for this formulation. Note that we have additionally verified the definiteness of all tangents for problem sizes up to 5430 DOFs, using a Cholesky decomposition. In the iterative solvers, the Jacobi and the SSOR preconditioner are employed. In order to obtain a reference for the performance of sparse direct solvers, we also consider the direct solver UMFPACK[17, 18] for both formulations.

The suitability of the iterative solvers in connection with the different preconditioners is first investigated for *Min* formulation. Herein, the relaxation parameter, required for the SSOR preconditioner, is set to $\omega = 0.65$. Considering the average number of linear iterations for the *Min* formulation, it is observed for all methods that the number of iterations increases with increasing number of DOFs, as expected, see Table 1 and Figure 3. However, the CG method, combined with the SSOR preconditioner (CG+SSOR), consistently required the lowest number of iterations. In the subsequent discussion of the computing times, we focus on the *Solver Time*, i.e. the time to solve the linear system in the Newton-Raphson iterations, as it dominates the total time to solution (*Total Time*), except for very small problems. The CG method with the Jacobi preconditioner (CG+Jacobi) is the fastest except for the smallest test case with 300 DOFs, where UMFPACK is faster by 0.01 s. For the SSOR preconditioner, the *Solver Time* is slightly higher compared to CG+Jacobi. However, the slopes by which the *Total Time* is increasing with increasing number of DOFs are similar, compare Figure 2. Overall, the CG method performs better than GMRES with regards to the avg. number of iterations and the *Solver Time*. The higher *Solver Time* for GMRES is due to, both, the higher number of iterations, and of the quadratic complexity in the Arnoldi process. Based on the comparison of the two iterative solvers and the two preconditioners employed for the *Min* formulation, it can be

Minimization								
$Q_1 RT_0$								
60 time steps, $\Delta t = 0.1$ s								
DOFs		300	1785	5430	12 207	39 045	90 075	295 815
Avg # lin. iter per Newton step	UMFPACK	-	-	-	-	-	-	-
	CG + Jacobi	157	442	726	1053	1777	2577	4250
	CG + SSOR	112	242	362	506	818	1085	1672
	CG + SSOR [†]	167	397	601	862	1363	1897	2823
	GMRES + Jacobi	410	1197	2088	3637	8568	18 342	*
	GMRES + SSOR	177	391	542	948	1641	2594	*
	GMRES + SSOR [†]	309	678	1216	1877	3857	6839	*
Assemble Time	UMFPACK	1.20	9.30	30.70	78.80	250	595	#
	CG + Jacobi	1.18	9.20	30.30	73.60	249	596	2062
	CG + SSOR	1.19	9.20	30.50	73.40	250	599	2068
	CG + SSOR [†]	1.18	9.20	30.20	73.50	249	596	2064
	GMRES + Jacobi	1.19	9.20	30.60	74.50	259	621	*
	GMRES + SSOR	1.20	9.20	30.40	74	259	616	*
	GMRES + SSOR [†]	1.22	9.30	31.20	76.40	260	646	*
Solver Time	UMFPACK	0.72	12.80	125.60	632	8208	49 210	#
	CG + Jacobi	0.73	11.20	54.60	176.80	1587	7207	46 040
	CG + SSOR	1.13	17.30	79.40	240.60	2198	8492	48 650
	CG + SSOR [†]	1.67	28	128.70	411.70	3486	14 980	81 470
	GMRES + Jacobi	2.50	41.30	204	779	9444	63 650	*
	GMRES + SSOR	2.15	33.10	136	495.50	4914	24 870	*
	GMRES + SSOR [†]	3.85	57.30	307.30	996	11 550	68 640	*
Total Time	UMFPACK	4.70	26.40	167.80	733	8538	49 970	#
	CG + Jacobi	3.80	34.90	98.40	273	1909	7994	48 630
	CG + SSOR	5.00	33.50	124.70	347	2552	9265	51 240
	CG + SSOR [†]	4.50	42.60	170.60	509.90	3804	15 740	84 070
	GMRES + Jacobi	5.70	59.20	253	881	9779	64 430	*
	GMRES + SSOR	5.50	48.10	185	597	5259	25 650	*
	GMRES + SSOR [†]	7.30	71.40	351	1118	11 890	69 460	*
total # Newton steps	UMFPACK	300	300	300	300	301	304	#
	CG + Jacobi	300	300	300	300	301	304	312
	CG + SSOR	300	300	300	300	301	304	312
	CG + SSOR [†]	300	300	300	300	301	304	312
	GMRES + Jacobi	300	300	301	304	311	317	*
	GMRES + SSOR	300	300	301	301	311	315	*
	GMRES + SSOR [†]	307	302	310	310	314	330	*

Table 1: Computation times for the *Min* formulation. Comparison of different (iterative) linear solvers and different levels of grid refinement. The combination of the CG and the GMRES solver with the SSOR preconditioner, marked with [†], have a common relaxation parameter of $\omega = 0.18$. For all the entries in the table, indicated by a *, the maximum number of GMRES linear iterations were exceeded, whereas the ones, marked with #, the maximum walltime limit of 96 hours was not sufficient to obtain a solution. We mark the lowest number of linear iterations as well as the fastest *Solver* and *Total time* in bold.

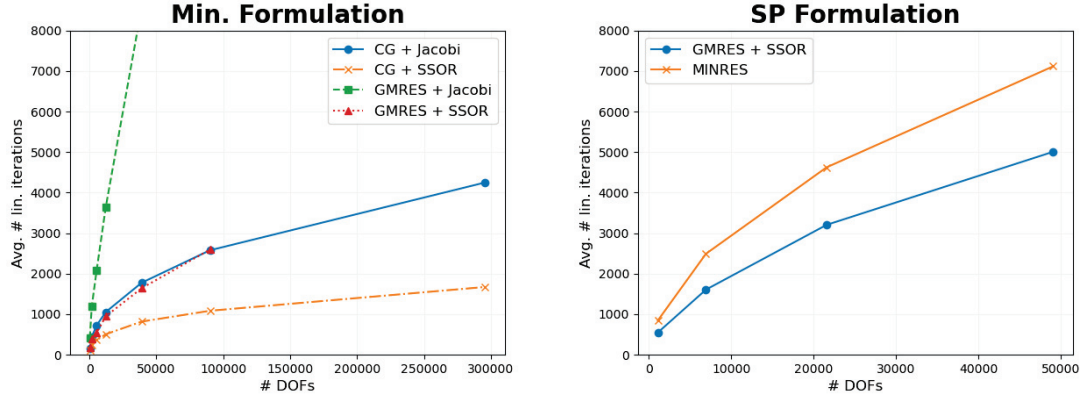


Figure 3: Average number of linear iteration over all Newton steps for the calculations performed with $\Delta t = 0.1$ s. In the *Min* formulation (left) with different iterative methods and preconditioner are considered, while the *SP* formulation (right) employs GMRES with SSOR and MINRES without any preconditioner.

concluded that CG+Jacobi is the most efficient method for this particular problem and the SSOR preconditioner does not reduce the *Solver Time* although the number of linear iterations can be significantly reduced. In contrast to CG, GMRES does not exploit the positive definiteness of the system matrix. Because GMRES builds an orthonormal basis for the Krylov subspace via the Arnoldi process, its computational cost grows with the number of iterations. As the update of the orthogonal basis in one GMRES iteration is computationally more expensive than a single CG iteration, even the application of the SSOR preconditioner, which reduces the number of linear iterations to that of CG, cannot fully compensate the computational overhead. This emphasizes the importance of exploiting known matrix properties when selecting the iterative solution method and the preconditioner.

For the *SP* formulation we consider the direct solver UMFPACK and iterative solvers MINRES and GMRES as the system is symmetric, but indefinite. Employing the simple Jacobi preconditioner in combination with GMRES did not lead to convergence of the iterative solver within the maximum number of linear iterations allowed. In the case of GMRES combined with the SSOR preconditioner, an adjustment of the relaxation parameter to $\omega = 0.18$ was necessary to ensure convergence. In fact, the value has been obtained by systematically varying the relaxation parameter in the range of $[0.1, 0.2]$, where the value of $\omega = 0.18$ led to lowest number of linear iterations. Similarly, as in the *Min* formulation, the number of linear iterations increases with increasing problem size. Consequently, the *Solver* and *Total Time* also increase. For the *SP* formulation, for the range of problem sizes considered, UMFPACK is always faster than the iterative methods, but the advantage reduces with the size of the problem. Note that we have considered only problems of modest size for the *SP* formulation. For larger problems, direct solvers will not be applicable anymore. We note that the system matrix in the *SP* formulation possesses a significantly larger number of non-zero entries as the one corresponding to the *Min* formulation, see Fig. 1. We observe that the number of GMRES+SSOR iterations is quite high

Saddle Point Formulation					
60 time steps, $\Delta t = 0.1$ s					
DOFs		1093	6934	21 577	49 072
Avg #	UMFPACK	-	-	-	-
lin. iter	MINRES	845	2 490	4 623	7 115
per Newton step	GMRES + SSOR	547	1 607	3 204	5 007
Assemble Time	UMFPACK	35.20	283	957	2 297
	MINRES	35.30	286	983	2 319
	GMRES + SSOR	35.30	283	961	2 505
Solver Time	UMFPACK	4.30	188	1 807	14 450
	MINRES	17.60	386	4 683	21 770
	GMRES + SSOR	38.10	706	8463	39 740
Total Time	UMFPACK	41.30	481	2 791	16 790
	MINRES	54.60	681	5 688	24 140
	GMRES + SSOR	75	997	9 448	42 300
total # Newton steps	UMFPACK	200	201	202	204
	MINRES	201	201	202	206
	GMRES + SSOR	201	201	202	222

Table 2: Computation times for the *SP* formulation. Comparison of direct and iterative linear solvers and different levels of grid refinement. The fastest *Solver* and *Total time* are marked in bold. An additional grid refinement step, yielding 158818 DOFs, has been excluded from the comparison, as the wall time was exceeded for GMRES+SSOR and the condition $\det(\mathbf{F}) > 0$ was violated in a not yet converged Newton iteration for UMFPACK, see [8] for further details on the latter issue.

for the *SP* formulation when compared with the *Min* formulation. As an alternative to GMRES, the MINRES method without a preconditioner is employed to solve the linear system in the *SP* formulation. Due to absence of a preconditioner, the number of linear iterations exceeds even the linear iterations required by GMRES+SSOR. However, for MINRES, the total solution times are still significantly lower, because its computational complexity is comparable to CG and thus yields a much more efficient method than GMRES+SSOR.

In summary, the comparison of the *Min* and *SP* formulation, both yielding a symmetric system matrix, as illustrated in Fig. 1, highlights that the property of being symmetric and positive definite is a significant advantage of the *Min* formulation since the CG method can be applied, leading to faster computation times, see also Fig. 2. When comparing GMRES+SSOR, the *Min* formulation is faster overall; see Tab. 1 and 2. In particular, the *SP* formulation required a *Total Time* of 42 300 s for a medium sized problem with 49 072 DOFs, while the simulations, using the *Min* formulation with 39 045 DOFs, were conducted in a *Total Time* of 5259 s ($\omega = 0.65$) and 11 890 s ($\omega = 0.18$), respectively.

5 CONCLUSIONS

Within the chemo-mechanical modeling framework discussed here, comparing the same problem, but cast either in a minimization formulation or in saddle point form, we observe significant advantages for the minimization problem: As the minimization formulation leads to symmetric positive definite problems, preconditioned CG can be applied, leading to significant gains in computation time compared to GMRES. Moreover, the system matrices are more sparse. As a result, for the range of problem sizes, where sparse direct solvers are still applicable, we see better computation times for the minimization formulation. This will carry over

to parallel domain decomposition methods, where sparse direct solvers are the local computational kernel. We can conclude that the choice of the variational formulation, combined with the suitable discretization, significantly influences the performance.

REFERENCES

- [1] M. Abu Jafar Mazumder, H. Sheardown, and A. Al-Ahmed, *Functional Biopolymers*, 1st ed., ser. Polymers and Polymeric Composites: A Reference Series. Springer Cham, 2019.
- [2] H. Cao, L. Duan, Y. Zhang, J. Cao, and K. Zhang, “Current hydrogel advances in physicochemical and biological response-driven biomedical application diversity,” *Signal Transduct Target Ther*, vol. 6, no. 1, pp. 1–31, Dec. 2021.
- [3] T.-C. Ho, C.-C. Chang, H.-P. Chan, T.-W. Chung, C.-W. Shu, K.-P. Chuang, T.-H. Duh, M.-H. Yang, and Y.-C. Tyan, “Hydrogels: Properties and Applications in Biomedicine,” *Molecules*, vol. 27, no. 9, p. 2902, May 2022.
- [4] L. Böger, A. Nateghi, and C. Miehe, “A minimization principle for deformation-diffusion processes in polymeric hydrogels: Constitutive modeling and FE implementation,” *Int J Solids Struct*, vol. 121, pp. 257–274, 2017.
- [5] L. Böger, *Saddle-Point and Minimization Principles for Diffusion in Solids : Phase Separation, Swelling and Fracture*. Stuttgart : Institute of Applied Mechanics, 2020.
- [6] D. Boffi, F. Brezzi, and M. Fortin, *Mixed Finite Element Methods and Applications*, 1st ed., ser. Springer Series in Computational Mathematics. Berlin, Heidelberg: Springer, 2013, no. 44.
- [7] W. Han and B. Reddy, *Plasticity*, 2nd ed. Springer New York, NY, 2013.
- [8] A. B. Jeeja, B. Kiefer, S. Prüger, O. Rheinbach, and F. Röver, “Comparison of a minimization and a saddle point formulation of a coupled mechanics-diffusion problem implemented in deal.II,” *PAMM*, vol. 25, no. 1, p. e202400188, 2025. [Online]. Available: <https://onlinelibrary.wiley.com/doi/abs/10.1002/pamm.202400188>
- [9] D. Arndt, W. Bangerth, M. Bergbauer, M. Feder, M. Fehling, J. Heinz, T. Heister, L. Heltai, M. Kronbichler, M. Maier, P. Munch, J.-P. Pelteret, B. Turcksin, D. Wells, and S. Zampini, “The deal.II library, version 9.5,” *J Numer Math*, vol. 31, no. 3, pp. 231–246, Aug. 2023. [Online]. Available: <https://www.degruyter.com/document/doi/10.1515/jnma-2023-0089/html>
- [10] S. Bellavia and B. Morini, “An interior global method for nonlinear systems with simple bounds,” *Optim Method Softw*, vol. 20, no. 4-5, pp. 453–474, Aug. 2005. [Online]. Available: <https://doi.org/10.1080/10556780500140516>

- [11] A. Meister, *Präkonditionierer*. Wiesbaden: Springer Fachmedien Wiesbaden, 2015, pp. 230–255. [Online]. Available: https://doi.org/10.1007/978-3-658-07200-1_5
- [12] D. Bielich, J. Langou, S. Thomas, K. Świrydowicz, I. Yamazaki, and E. G. Boman, “Low-synch Gram–Schmidt with delayed reorthogonalization for Krylov solvers,” *Parallel Comput*, vol. 112, p. 102940, 2022. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0167819122000394>
- [13] A. Toselli and O. Widlund, *Domain decomposition methods—algorithms and theory*, ser. Springer Series in Computational Mathematics. Springer-Verlag, Berlin, 2005, vol. 34. [Online]. Available: <https://doi.org/10.1007/b137868>
- [14] B. Kiefer, S. Prüger, O. Rheinbach, and F. Röver, “Monolithic parallel overlapping Schwarz methods in fully-coupled nonlinear chemo-mechanics problems,” *Comput Mech*, vol. 71, no. 4, pp. 765–788, 2023.
- [15] A. Heinlein, A. Klawonn, S. Rajamanickam, and O. Rheinbach, “FROSch: A fast and robust overlapping Schwarz domain decomposition preconditioner based on Xpetra in Trilinos,” in *Domain Decomposition Methods in Science and Engineering XXV*, R. Haynes, S. MacLachlan, X.-C. Cai, L. Halpern, H. H. Kim, A. Klawonn, and O. Widlund, Eds. Cham: Springer, 2020, pp. 176–184.
- [16] T. Trilinos Project Team, *The Trilinos Project Website*.
- [17] T. A. Davis and I. S. Duff, “An unsymmetric-pattern multifrontal method for sparse lu factorization,” *SIAM J Matrix Anal A*, vol. 18, no. 1, pp. 140–158, 1997. [Online]. Available: <https://doi.org/10.1137/S0895479894246905>
- [18] R. F. Boisvert, T. A. Davis, and I. S. Duff, “A combined unifrontal/multifrontal method for unsymmetric sparse matrices,” *ACM Trans Math Softw*, vol. 25, no. 1, p. 1–20, 1999. [Online]. Available: <https://doi.org/10.1145/305658.287640>