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Dynamic optimization and event location in atmospheric chemistry

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A model coupling differential equations and an optimization problem with equality and inequality constraints is described. The first order optimality conditions are coupled with the differential equations to form a differential-algebraic system. Inequality constraints for the optimization variables induce discontinuities in the first derivatives of the solution in time. Tracking techniques based on sensitivity analysis and second order multistep methods are proposed to locate the discontinuities. Application to the dynamics of organic atmospheric aerosol particles is given to illustrate the detection of the activation of constraints.

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1 Optimization-Constrained Differential Equations

The modeling of the dynamics of organic-containing aerosol particles is a crucial step in the simulation of atmospheric processes. A model problem for the description of the dynamics and mass transfer between an aerosol particle and the ambient gas phase is presented following the model described in [1]. The thermodynamic equilibrium inside the particle is modeled by a constrained global (non-convex) minimization problem, while differential equations describe the fluxes between the particle and the surrounding gas arising in the mass transfer. The coupling appears in the fluxes of the differential equations and the equality constraints of the optimization problem.

Let $\mathbf{f} : \mathbb{R} \times \mathbb{R}^p \times \mathbb{R}^m \to \mathbb{R}^p$ Lipschitz continuous, $g \in C^\infty(\mathbb{R}^m)$ and $A \in \mathbb{R}^{p\times m}$. The problem reads: find $\mathbf{b} : (0,T) \to \mathbb{R}^p$ and $\mathbf{z} : (0,T) \to \mathbb{R}^m$ satisfying

$$\begin{aligned} &\frac{d}{dt}\mathbf{b}(t) = \mathbf{f}(t, \mathbf{b}(t), \mathbf{z}(t)), \quad \text{a.e. } t \in (0,T), \quad \mathbf{b}(0) = \mathbf{b}_0, \\ &\mathbf{z}(t) = \arg\min_{\tilde{\mathbf{z}}} g(\tilde{\mathbf{z}}) \quad \text{s. t.} \quad A\tilde{\mathbf{z}} = \mathbf{b}(t), \quad \tilde{\mathbf{z}} \geq 0. \end{aligned} \tag{1}$$

The variable $\mathbf{z}(t)$ loses regularity when one of the inequality constraints $\mathbf{z}_i(t) \geq 0$, $i = 1, \ldots, m$ is activated or deactivated. More precisely the first derivative of $\mathbf{z}_i(t)$ is discontinuous at the transition points when one component of $\mathbf{z}(t) \geq 0$ tends to zero (activation) or when one component of $\mathbf{z}(t) = 0$ becomes strictly positive (deactivation).

2 Numerical Model

We propose a numerical method that relies on a primal-dual interior-point method for the minimization problem coupled with a first order implicit scheme for the time discretization of the resulting system of differential-algebraic equations [1]. Let $h > 0$ be a fixed time step, $t^n = nh$ discrete times, and $\mathbf{z}^n$ and $\mathbf{b}^n$ denote respectively approximations of $\mathbf{z}(t^n)$ and $\mathbf{b}(t^n)$, with $\mathbf{b}^0 = \mathbf{b}_0$. The problem discretized in time corresponds to finding $\mathbf{z}^n \in \mathbb{R}^m$ and $\mathbf{b}^n \in \mathbb{R}^p$ at each time step satisfying:

$$\begin{aligned} &\frac{1}{h}(\mathbf{b}^n - \mathbf{b}^{n-1}) = \mathbf{f}(t^n, \mathbf{b}^n, \mathbf{z}^n), \quad n = 1, 2, \ldots, \\ &\mathbf{z}^n = \arg\min_{\tilde{\mathbf{z}}} g(\tilde{\mathbf{z}}) \quad \text{s. t.} \quad A\tilde{\mathbf{z}} = \mathbf{b}^n, \quad \tilde{\mathbf{z}} \geq 0. \end{aligned} \tag{2}$$

The sequence of discrete variables is constrained by an optimization problem at each time step. The minimization problem is replaced by its first order optimality conditions. Both are equivalent in the case of a convex optimization problem, but the second order optimality conditions have to be satisfied to obtain the global minimum in the non-convex case [2]. The first order optimality conditions provide the following system of nonlinear equations: find $\mathbf{z}^n \in \mathbb{R}^m$ and $\mathbf{b}^n \in \mathbb{R}^p$ satisfying:

$$\begin{aligned} &\frac{1}{h}(\mathbf{b}^n - \mathbf{b}^{n-1}) = \mathbf{f}(t^n, \mathbf{b}^n, \mathbf{z}^n), \\ &\nabla g(\mathbf{z}^n) + A^T \boldsymbol{\lambda}^n - \boldsymbol{\theta}^n = 0, \quad A\mathbf{z}^n = \mathbf{b}^n, \\ &\mathbf{z}^n \geq 0, \quad \boldsymbol{\theta}^n \geq 0, \quad \mathbf{z}_i^n \boldsymbol{\theta}_i^n = 0, \quad \forall i = 1, \ldots, m, \end{aligned} \tag{3}$$

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where $\boldsymbol{\lambda}^n$ and $\boldsymbol{\theta}^n$ are the dual multipliers.

An interior-point method [4] relaxes the complementarity conditions $\mathbf{z}_i^n \boldsymbol{\theta}_i^n = 0$ by introducing a *penalty parameter* $\nu > 0$ that tends to zero at each time step. Thus these conditions are replaced by $\mathbf{z}_i^n \boldsymbol{\theta}_i^n = \nu$, $i = 1, \ldots, m$, $\mathbf{z}^n > 0$, $\boldsymbol{\theta}^n > 0$, and then the variables $\boldsymbol{\theta}_i^n$ are eliminated from the system by writing $\boldsymbol{\theta}_i^n = \nu / \mathbf{z}_i^n$ for $i = 1, \ldots, m$. Starting with an interior-point parameter ν^0, the relaxed system of nonlinear equations is solved by applying one Newton iteration, then decreasing the parameter $\nu^k = \xi \nu^{k-1}$, $\xi \in (0, 1)$ and repeating the process until convergence is reached [1, 2].

3 Tracking of Discontinuities

When an inequality constraint $\mathbf{z}_i \geq 0$ is activated or deactivated, the variable $\mathbf{z}(t)$ is not continuously differentiable anymore. In order to ensure the global convergence order of a numerical method for the time-discretization of the differential-algebraic system (for instance any Runge-Kutta methods), the discontinuity points have to be located with a higher order method. We discuss here only the activation case (when one component $\mathbf{z}_i > 0$ tends to zero).

Let us assume that the interval $[t^{n-1}, t^n)$ contains a discontinuity point for the ith component of $\mathbf{z}$. The fractional time step $h^\star \in (0, h)$ needed to locate the discontinuity point is obtained with a Taylor expansion and a first order approximation:

$$0 = \mathbf{z}_i(t^{n-1} + h^\star) = \mathbf{z}_i(t^{n-1}) + h^\star \frac{d\mathbf{z}_i}{dt}(t^{n-1}) + \mathcal{O}((h^\star)^2), \quad i.e. \quad h^\star \simeq -\mathbf{z}_i(t^{n-1}) / \frac{d\mathbf{z}_i}{dt}(t^{n-1}). \tag{4}$$

The approximation of the first derivative $\frac{d\mathbf{z}_i}{dt}(t^{n-1})$ is based on sensitivity analysis techniques [4]. The differentiation of the algebraic part of (3) with $\nu = 0$ allows to compute the variations of the variable $d\mathbf{z}/dt(t^{n-1})$ and $d\boldsymbol{\lambda}/dt(t^{n-1})$:

$$\begin{bmatrix} \nabla^2 g(\mathbf{z}) & A^T \\ A & 0 \end{bmatrix} \begin{bmatrix} d\mathbf{z}/dt(t^{n-1}) \\ d\boldsymbol{\lambda}/dt(t^{n-1}) \end{bmatrix} = \begin{bmatrix} 0 \\ d\mathbf{b}/dt(t^{n-1}) \end{bmatrix} = \begin{bmatrix} 0 \\ \mathbf{f}(t^{n-1}, \mathbf{b}^{n-1}, \mathbf{z}^{n-1}) \end{bmatrix}$$

Once the fractional time step $h^\star$ is obtained with (4), a predictor-corrector method based on two-steps Adams-Bashforth and Adams-Moulton schemes [3] allows to determine the approximation $\mathbf{b}^n$ of $\mathbf{b}(t)$ at time $t^{n-1} + h^\star$ as follows:

$$\tilde{\mathbf{b}}^n = \mathbf{b}^{n-1} + h^\star \left[\left(1 + \frac{h^\star}{2h}\right) \mathbf{f}(t^{n-1}, \mathbf{b}^{n-1}, \mathbf{z}^{n-1}) - \frac{h^\star}{2h} \mathbf{f}(t^{n-2}, \mathbf{b}^{n-2}, \mathbf{z}^{n-2}) \right] \quad \text{(predictor)},$$

$$\mathbf{b}^n = \mathbf{b}^{n-1} + \frac{1}{2} h^\star \left(\mathbf{f}(t^{n-1} + h^\star, \tilde{\mathbf{b}}^n, \tilde{\mathbf{z}}^n) + \mathbf{f}(t^{n-1}, \mathbf{b}^{n-1}, \mathbf{z}^{n-1}) \right) \quad \text{(corrector)}.$$

The variable $\tilde{\mathbf{z}}^n$ is obtained by solving the pure optimization problem for given $\tilde{\mathbf{b}}^n$ [2]. Numerical results for the detection of discontinuities are presented for the phase equilibrium problem described in [1, 2], applied to the hexacosanol-pinic acid-water system. In the special case when all components of $\mathbf{z}(t)$ are strictly positive, the exact solution $\mathbf{b}(t)$ and the exact time of first activation $t^\star$ are known. Figure 1 illustrates the numerical approximation of $\mathbf{b}(t)$ and $\mathbf{z}_1(t)$ and a convergence result when activating the constraint $\mathbf{z}_1(t) > 0$ and going from three to two strictly positive components of $\mathbf{z}(t)$.

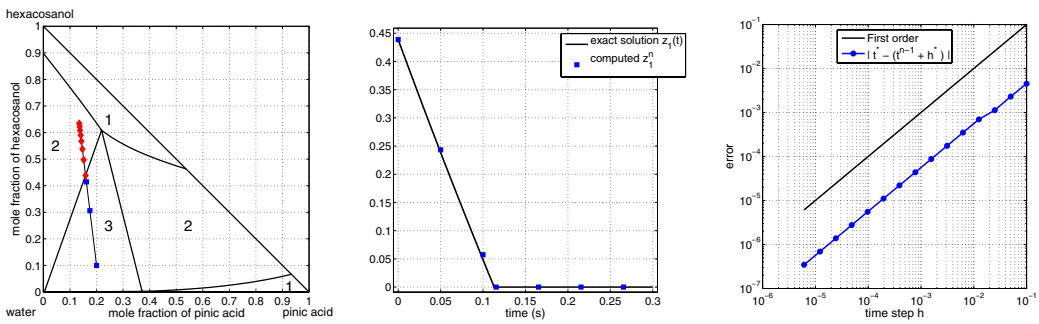

Fig. 1 Numerical approximation of the trajectory $\mathbf{b}(t)$ (left) and first component of $\mathbf{z}(t)$ (middle), with computation of fractional time step and detection of the discontinuity point, and log-log convergence plot of the error $\left|t^\star - (t^{n-1} + h^\star)\right|$ (right).

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