Lagrange–Galerkin Approximation For
Advection–Dominated Contaminant Transport With
Nonlinear Equilibrium Or Non–equilibrium
Adsorption *

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Abstract

An extension of the Lagrange–Galerkin approach is developed for advection-dominated problems with nonlinear adsorption, either being in equilibrium or in non–equilibrium, possibly with isotherms of the Freundlich type. The scheme should be feasible also for the hyperbolic limit case.

The basic problems to deal with are the possibility of non–unique characteristics due to the nonlinear isotherms and the incorporation of the time–dependent non–equilibrium adsorption kinetics, avoiding a strong restriction on the CFL–number. The one–dimensional scheme derived is able to handle shock solutions.

In the non–equilibrium case a more severe restriction on the timestep has to be regarded.

Key words. method of characteristics, degenerate parabolic equation, finite elements

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Introduction

The numerical simulation of the transport of solutes by dispersion and advection, which in addition can be adsorbed to the soil matrix by equilibrium/non-equilibrium kinetics, is one of the main problems in soil science and subsurface hydrology.

The aim of this paper is to develop an appropriate Lagrange-Galerkin discretization for a one-site transport model with either equilibrium or non-equilibrium adsorption sites. The considered model, in the case of non-equilibrium adsorption reactions, takes the following form. The unknowns $u, v$, the dissolved and the adsorbed concentration, have to fulfill:

\[
\begin{align*}
\partial_t u + \partial_x v + q \nabla u - \nabla \cdot (D \nabla u) &= 0 & \text{in} & & (0, T] \times \Omega \subset (0, T] \times \mathbb{R}^3 \\
\partial_t v &= k(\varphi(u) - v) & \text{in} & & (0, T] \times \Omega \\
u(0, x) &= u_0(x) & x & \in \Omega \\
v(0, x) &= v_0(x) & x & \in \Omega,
\end{align*}
\]

and appropriate boundary conditions.

The water content and the bulk density are assumed to be constant and scaled to unity, and

\[
\begin{align*}
\varphi &\in C^r([0, \infty)) \cap C_{\text{loc}}^1((0, \infty)) & r &\in (0, 1) \\
\varphi(\cdot) &\geq 0 \\
\infty &> \varphi'(s) \geq 0 & \text{for} & s > 0 \\
\infty &\geq \lim_{s \to +0} \varphi'(s) \geq 0,
\end{align*}
\]

The diffusion/dispersion coefficient $D$ and the flow field $q$ are assumed to be smooth functions, such that:
\[ q : (0, T] \times \Omega \rightarrow \mathbb{R}^N, \quad ||q|| \neq 0, \quad \nabla \cdot q = 0, \quad (4) \]
\[ D : (0, T] \times \Omega \rightarrow \mathbb{R}^{N^2}, \quad \nu ||\xi||^2 \leq \xi^T D(t, x) \xi \leq \mu ||\xi||^2 \text{ for } \nu, \mu > 0. \quad (5) \]

The exact conditions under which (1) has weak or strong solutions are analysed in Knabner [3].

The equilibrium model can be obtained formally from (1) by the limiting process \( k \) to \( \infty \), just leaving the dissolved concentration \( u \) as unknown.

We will consider Lagrange–Galerkin approaches for discretising the two models. For a linear instationary convection–diffusion equation, this type of method is characterised by writing the hyperbolic part as derivative along the characteristics, discretising this derivative by standard differences in time and dealing with the diffusive part implicitly in time. Finally the finite element discretization is applied to the weak formulation of the semi–discretised equation. The main advantage of this scheme are small truncation errors in time and thus large CFL–numbers.

A Lagrange–Galerkin approximation of the model considered should have this property as far as possible, too. Developing such a discretization has to handle three basic problems:

- the incorporation of the hyperbolic limit case \( D = 0 \), leading to discontinuous solutions.

- the incorporation of degenerate nonlinearities of the Freundlich type \( (\varphi'(0+) = \infty) \) leading to solutions with a sharp front, which have much in common with the hyperbolic limit case (in particular the speed of propagation), cf. Knabner [3].

- the incorporation of kinetic adsorption reactions.

Throughout this paper we will restrict the derivation of the discretization to the one–dimensional case, i.e. \( N = 1 \), but it can be extended to two dimensions, as well. Furthermore we use linear finite elements with vertex quadrature to avoid instabilities, caused by quadrature formulas of higher order (see Morton et al. [4]).

Let \( \Omega = [0, L], L \in \mathbb{R}^+ \) and for the sake of simplicity \( q \) a constant, say \( q > 0 \). \( \phi \) denotes a differentiable approximation of \( \varphi \). Just for the sake of simplicity uniform discretizations
in time and in space are chosen.

\[ 0 = t^0 < \cdots < t^{n-1} < t^n < \cdots < t^{N_t} = T, \quad \Delta t = t^i - t^{i-1}, \quad 0 < i \leq N_t, \quad (6) \]

\[ 0 = x_0 < \cdots < x_{j-1} < x_j < \cdots < x_{N_x} = L, \quad h = x_i - x_{i-1}, \quad 0 < i \leq N_x. \quad (7) \]

From now on we will define characteristics not with respect to a given equation, but to a given flux. A Lagrange–Galerkin approach can result in small time truncation errors only, if the characteristics are computed sufficiently accurate. As consequence, a given flux \( b_j \) \((0 \leq j \leq N_x)\), represented by its nodal values, will be approximated piecewise constant on each element and the characteristics are then calculated exactly with respect to this approximative flux. \( \bar{x}_j \) denotes the point in space where the characteristic, starting at \( t^n \) at the node \( x_j \), arrives at \( t^n - \Delta t \) and is called ‘backtrace’.

In formulas:

\[ b_{j-1/2} := b(x_j), \quad (8) \]

\[ \bar{x}_j = x_{j-m} + b_{j-m-1/2} \sum_{k=1}^{m} \left( \frac{h}{b_{j+1/2-k}} \right) - \Delta t, \quad (9) \]

and \( m \geq 0 \) is the first integer for which

\[ \sum_{k=1}^{m+1} \left( \frac{h}{b_{j+1/2-k}} \right) > \Delta t. \quad (10) \]

Furtheron the following notations will be used:

\[ Q_j(x, u^{n-1}) := \begin{cases} \phi(U^{n-1}(x)) - \phi(U^{n-1}_j), & \text{if } U^{n-1}_j \neq U^{n-1}(x) \\ \phi'(U^{n-1}_j), & \text{otherwise.} \end{cases} \quad (11) \]

\[ \delta(D\delta U)^n_j = \rightleftharpoons \left\{- \left( \frac{D_j - D_{j-1}}{2} \right) u^n_{j-1} + \left( \frac{D_{j+1} + 2D_j + D_{j+1}}{2} \right) u^n_j - \left( \frac{D_j + D_{j+1}}{2} \right) u^n_{j+1} \right\}. \quad (12) \]
Equilibrium Adsorption Reactions

A first Lagrange–Galerkin discretization was already presented in Knabner, Kappmeier, and Barrett [2]. Therefore we only summarize the derivation, restricted to the hyperbolic case $D = 0$, as the discretization of the diffusive term causes no problems and will be neglected therefore. There will be some small but important differences to the previous version.

The starting point is to introduce an ansatz–function $\alpha(t, x, u)$ for a given timestep and to define the characteristics with respect to the flux $b = \frac{q}{1 + \alpha(t, x, u)}$. That is:

$$
\begin{align*}
(\partial_t \phi(u) - \alpha(t, x, u) \partial_t u) + \\
(1 + \alpha(t, x, u)) \left[ \partial_t u + \frac{q}{1 + \alpha(t, x, u)} \partial_x u \right] = 0 & \quad \text{in } (t^{n-1}, t^n) \\
u(t, 0) = g(t) & \quad \text{in } (t^{n-1}, t^n) \\
u(t^{n-1}, x) = U^{n-1}(x).
\end{align*}
\tag{13}
$$

$U^{n-1}$ denotes the computed solution of the last timestep. Here and in the following we use a Dirichlet boundary condition on the inflow boundary $x = 0$.

Known choices for $\alpha$ are $\alpha(t, x, u) = 0$ (‘Pore–Velocity–Scheme’) and $\alpha(t, x, u) = \alpha'(u)$ (‘Retarded–Velocity–Scheme’ (RVS)). Choosing large CFL–numbers, the Pore–Velocity–Scheme tends to compute ‘zig–zag–solutions’, smears a lot in the case of steep profiles and the RVS fails, if shocks are present.

As a new approach we defined $\alpha$ on the discrete level in such a way, that a pure (discrete) derivative along the characteristic resulted. By this idea (see Knabner, Kappmeier, Barrett [2] for details) the following scheme is derived from (13). We denote this new scheme by FIS or by FISD in the case with diffusion:

$$U^n_j + \phi(U^n_j) = (1 + \alpha_j)U^{n-1}(\bar{x}_j) - \alpha_j U^{n-1}_j + \phi(U^{n-1}_j), \tag{14}$$

where $\alpha_j$ is the infimum of all solutions $\beta \geq 0$ of

$$\beta = Q_j(\bar{x}_j, U^{n-1}), \tag{15}$$

for $\bar{x}_j$ defined by $\tilde{b}$

$$\tilde{b}_i = \frac{q}{1 + \alpha_i} \quad i \neq j, \tag{16}$$
\[
\hat{b}_j = \frac{q}{1 + \beta}.
\]

(17)

In this case

\[
U_j^n = U^{n-1}(\bar{x}_j).
\]

(18)

is equivalent to (14).

Note that in the present simple situation of one space dimension and a constant \( q > 0 \) the resolution of (14)–(17), which in general defines a nonlinear coupling between all values \( U_i^n \) and \( \alpha_i \), can proceed according to the numbering of the nodes. Then \( U_i^n, \alpha_i, \)
\( i = 1, \cdots, j - 1 \) are already known and (14)–(17) define a nonlinear equation for \( \alpha_j \) and \( U_j^n \). In general for ‘most’ fluxes a ‘downwind numbering’ is possible, which preserves the property, that only known values \( \alpha_i \) additionally to \( \alpha_i \) are involved in the computation of the backtrace \( \bar{x}_j \).

An open item in Knabner, Kappmeier, Barrett [2] was the existence of solutions \( \beta \) for the equations (15)–(17).

**Theorem 1** The computed solution \( U^{n-1} \) of the old time level is extended to \( \mathbf{R} \). Furthermore \( U^{n-1} \geq 0, \phi \in C^1([0, \infty)), q = \text{const} > 0 \) are assumed. \( \alpha_i \in [0, \infty) \) are given for \( 0 \leq i < j \). The size of the timestep \( \Delta t \) from \( U^{n-1} \) to \( U^n \) is chosen.

Then there exists a solution \( \beta \geq 0 \) for (15)–(17).

The solutions \( \beta \) for (15)–(17) can be computed by using the bisection method.

A speed up of convergence can be obtained by using a method of higher order (secant method, Brent’s method). The main difference to the first version in Knabner, Kappmeier, Barrett [2] is the different piecewise constant approximation of the flux \( b \), now defined by the downwind node (see (8)) and not by the midvalue of the flux at both nodes as in the previous paper. Now we have for \( \Delta t \to 0 \) that \( b_{j-1/2} \to \frac{x}{1 + \phi(U_j^{n-1})} \) independent of the spatial discretization, i.e. the situation of the retarded-velocity-scheme.
Nonequilibrium Adsorption Reactions

The system (1) with the unknowns $u, v$ can be transformed to an equivalent partial differential equation in the unknown $u$ alone:

$$\begin{align*}
\partial_t u + \partial_t \varphi(u) + q \nabla u - \nabla \cdot (D \nabla u) + k^{-1} \partial_t \left[ \partial_t u + q \nabla u - \nabla \cdot (D \nabla u) \right] &= 0 \quad \text{in } (0, T) \times \Omega \subset (0, T] \times \mathbb{R}^N \\
u(0, x) &= u_0(x) \quad x \in \Omega \\
k^{-1} \left| \partial_t u + q \nabla u - \nabla \cdot (D \nabla u) \right| (0, x) &= v_0(x) - \varphi(u_0(x)) \quad x \in \Omega.
\end{align*}$$

(19)

By (19) the non-equilibrium case can be interpreted as the equilibrium model amended by a perturbation term of the order $\frac{1}{k}$ in $\partial_t u$. Only in special cases this term can be rewritten as a purely spatial operator. In the one-dimensional case for constant coefficients we have for travelling wave solutions $\partial_t = -a \partial_x$ with a wave speed $a < q$ (cf. v.Duijn, Knabner [1]) and thus:

$$\begin{align*}
k^{-1} \partial_t [\partial_t u + q \partial_x u - D \partial_{xx} u] &= \\
-k^{-1} \left[ a(q - a) \partial_{xx} u - Da \partial_{xxx} u \right].
\end{align*}$$

(20)

In general an extension of the FIS/FISD to the non-equilibrium case will have an additional time truncation error due to the discretization of $\partial_t u$.

As it makes no additional difficulties to allow for diffusion, we will neglect diffusion during the analysis and derivation of the different schemes. But we formulate them for the case with diffusion.

Introducing a nonnegative ansatz function $\alpha(t, x, u, v)$, equation (1), restricted to $(t^{n-1}, t^n)$ and with $U^{n-1}, V^{n-1}$ as initial condition, becomes:

$$\begin{align*}
\partial_t v - \alpha(t, x, u, v) \partial_t u + \left( 1 + \alpha(t, x, u, v) \right) \left[ \partial_t u + \frac{q}{1 + \alpha(t, x, u, v)} \partial_x u \right] &= 0 \quad \text{in } (t^{n-1}, t^n) \\
\partial_t v &= k(\varphi(u) - v) \quad \text{in } (t^{n-1}, t^n) \\
u(t, 0) &= g(t) \quad \text{in } (t^{n-1}, t^n) \\
\end{align*}$$

(21)
\[ u(t^{n-1}, x) = U^{n-1}(x) \]
\[ v(t^{n-1}, x) = V^{n-1}(x). \]

In the equilibrium case the FIS chooses \( \alpha(t, x, u) \), so that \( \partial_t \phi(u) - \alpha(t, x, u) \partial_t u \) vanishes on the discrete level. The corresponding requirement for (21) is to define \( \alpha \) by \( \partial_t v - \alpha(t, x, u) \partial_t u = 0 \) on the discrete level. However this idea leads to a flux \( b \), which is possibly unbounded and changes sign.

The flux \( b \) should model the flux, or a part of it, of the dissolved concentration, \( \alpha \) should depend on \( u \). Furthermore \( b \) should incorporate the influence of the kinetics on the flux of \( u \), by using the corresponding equilibrium reaction and a \( k \)-dependent correction term.

Till the end of this Section \( u, v \) are assumed to be regular enough, to allow for the following transformations.

The ODE in (21) implies:

\[ (\partial_t + k)(v) = k(\phi(u)), \]  \quad (22)
\[ (\partial_t + k)(\partial_t v) = k(\partial_t \phi(u)). \]  \quad (23)

(23) can now be used in two ways:

1. By variation of the constant one gets:

\[
\partial_t v(t) = \exp(-k(t - t^{n-1})) \partial_t v(t^{n-1}) + 
k \int_{t^{n-1}}^{t} \exp(-k(t - s)) \partial_s \phi(u) ds \quad t \in [t^{n-1}, t^n), \tag{24}
\]

Replacing \( \partial_t v \) in (21) by (24) and then \( \partial_t v(t^{n-1}) \) by means of (22) leads to:

\[
k \int_{t^{n-1}}^{t} \exp(-k(t - s)) \partial_s \phi(u) ds - \alpha(t, x, u, v) \partial_t u + 
(1 + \alpha(t, x, u, v)) \left[ \partial_t u + \frac{q}{1 + \alpha(t, x, u, v)} \partial_x u \right] = 
-k \exp(-k(t - t^{n-1}))(\phi(u(t^{n-1})) - v(t^{n-1})).
\]  \quad (25)

To arrive at a Lagrange–formulation \( \alpha(t, x, u, v) \) should be defined in such a way, that \( k \int_{t^{n-1}}^{t} \exp(-k(t - s)) \partial_s \phi(u) ds - \alpha(t, x, u, v) \partial_t u = 0 \) (on the discrete level).
A possible scheme is determined by the chosen approximation of
\[ k \int_{t_{n-1}}^{t_n} \exp(-k(t-s)) \partial_s \phi(u) \, ds. \]

2. Replacing \( \partial_t v \) in (21) by means of (23) results in:

\[
(1 + \alpha(t, x, u, v)) \partial_t u + \frac{q}{1 + \alpha(t, x, u, v)} \partial_x u = 0.
\]

Now \( \alpha(t, x, u, v) \) will be defined according to the FIS, implying \( \partial_t \phi(u) - \alpha(t, x, u, v) \partial_t u \) = 0 on the discrete level. This should restrict the time truncation error due to \( \partial_t \phi(u) - \alpha(t, x, u, v) \partial_t u \).

The main difference to the equilibrium case is the perturbation term \( k^{-1} \partial_{tt} v \). One way to handle this part is to replace it by \( k^{-1} \partial_t \left[ \partial_t u + q \partial_x u \right] \) and to end up with equation (19). This second term can be written as a derivative in time of one along the characteristic of the underlying flowfield. Due to the derivative along the characteristic it may be expected, that this leads to a smaller time truncation error than the discretization of \( k^{-1} \partial_{tt} v \) itself.

The scheme presented next is a consequence of (25), by approximating
\[ k \int_{t_{n-1}}^{t_n} \exp(-k(t-s)) \partial_s \phi(u) \, ds \] by \( k \int_{t_{n-1}}^{t_n} \exp(-k(t-s)) \, ds \partial_t \phi(u) = 
(1 - \exp(-k(t - t^{n-1}))) \partial_t \phi(u) \) and \( \exp(-k(t - t^{n-1})) \) by \( \frac{1}{1 + k(t - t^{n-1})} \).

**Discretising (25)**: The resulting scheme for interior nodes is, with \( b_j = \frac{q_j}{1 + \alpha_j} \):

\[
U_j^n = \frac{\Delta t}{h^2} \delta(D \delta U)_j^n + \frac{k \Delta t}{1 + k \Delta t} \phi(U_j^n) = 
U_j^{n-1}(\bar{x}_j) + \frac{k \Delta t}{1 + k \Delta t} \phi(U_j^{n-1}(\bar{x}_j)) + \frac{k \Delta t}{1 + k \Delta t} \left[ V_j^{n-1} - \phi(U_j^{n-1}) \right],
\]

where \( \alpha_j \) is the infimum of all solutions \( \beta \geq 0 \) of

\[
\beta = \frac{k \Delta t}{1 + k \Delta t} Q_j(\bar{x}_j, U_j^{n-1}),
\]

(28)
for \( \bar{x}_j \) defined by \( \bar{b} \)

\[
\dot{b}_i = \frac{q}{1 + \alpha_i} \quad i \neq j, \quad (29)
\]

\[
\dot{b}_j = \frac{q}{1 + \beta}, \quad (30)
\]

\[
V^n_j = \frac{1}{1 + k\Delta t} \left( k\Delta t \phi(U^n_j) + V^{n-1}_j \right) \quad \forall n \geq 1. \quad (31)
\]

**Discretising (19) :** As consequence of (26) the resulting scheme for interior nodes is, with

\[
b_j = \frac{q_j}{1 + \alpha_j};
\]

\[
U^n_j - \frac{\Delta t}{\hbar^2} \delta(D\delta U)^n_j + \frac{k\Delta t}{1 + k\Delta t} \phi(U^n_j) =
\]

\[
\frac{k\Delta t}{1 + k\Delta t} \left[ U^{n-1}(\bar{x}_j) + \phi(U^{n-1}(\bar{x}_j)) \right] + \frac{1}{1 + k\Delta t} U^{n-1}(x_j - q\Delta t)
\]

\[
+ \begin{cases} 
\frac{1}{1 + k\Delta t} \left( U^{n-1} - \frac{\Delta t}{\hbar^2} \delta(D\delta U)^{n-1}_j - U^{n-2}(x_j - q\Delta t) \right) & \text{if } n \geq 2 \\
\frac{k\Delta t}{1 + k\Delta t} (v_0(x_j) - \phi(u_0(x_j))) & \text{if } n = 1, 
\end{cases} \quad (32)
\]

where \( \alpha_j \) is the infimum of all solutions \( \beta \geq 0 \) of

\[
\beta = Q_j(\bar{x}_j, U^{n-1}), \quad (33)
\]

for \( \bar{x}_j \) defined by \( \bar{b} \)

\[
\dot{b}_i = \frac{q}{1 + \alpha_i} \quad i \neq j, \quad (34)
\]

\[
\dot{b}_j = \frac{q}{1 + \beta}, \quad (35)
\]

\( V^n_j \) is defined as in (31).

It should be noticed, that both schemes ((32), (27)) reduce to the FIS/FISD for \( k \to \infty \) and the linear finite element discretization of the instationary diffusion equation with adsorption, if \( q = 0 \).
Numerical Results

In the equilibrium case the FIS/FISD turned out to be superior to the Retarded-Velocity-Scheme or the Pore-Velocity-Scheme (see Knabner, Kappmeier, Barrett [2]). Therefore we take the Pore-Velocity-Scheme in the non-equilibrium case as benchmark, as well. For interior nodes the Pore-Velocity-Scheme (PVS) is:

\[
U_j^n - \frac{\Delta t}{h^2} \delta(D\delta U)_j^n + \frac{k\Delta t}{1 + k\Delta t} \phi(U_j^n) = U_{j-1}^{n-1}(x_j - q\Delta t) + \frac{k\Delta t}{1 + k\Delta t} V_{j-1}^{n-1}(x_j)
\]

\[
V_j^n = \frac{1}{1 + k\Delta t} (k\Delta t \phi(U_j^n) + V_j^{n-1}) \quad \forall n \geq 1.
\]

The three schemes ((32), (27), (36)) were tested in three situations:
For travelling waves with nonlinear isotherm \( \varphi(u) = u^p, p \in (0, 1) \) (situation 1), pulse with linear isotherm \( \varphi(u) = K_d u, K_d > 0 \) (situation 2), pulse with nonlinear isotherm \( \varphi(u) = u^p, p \in (0, 1) \) (situation 3).

In the equilibrium case (27) and (32) coincide with the FIS/FISD. For linear and nonlinear isotherms the FIS/FISD allows for larger CFL-numbers and smears much less than the Pore-Velocity-Scheme (36), if steep fronts are present. However strong refinement in space can be necessary to conserve mass, in the case of degenerate nonlinear isotherms, sharp fronts and CFL-numbers larger than one. In summary the FIS/FISD seems to be the better choice.

In the non-equilibrium case the experiments show:
Scheme (27) requires smaller timesteps than the Pore-Velocity-Scheme (36), otherwise peaks and oscillations can be observed and therefore seems to be no good extension of the FIS/FISD.
In comparison to the equilibrium case a stronger restriction of the timestep size is necessary for Scheme (32), as well. Too large timesteps cause peaks and oscillations.
For \( \frac{q}{1+\alpha} \) nearby \( q \) or small rate parameter \( k \) the Pore-Velocity-Scheme (36) allows for larger timesteps. In the other cases scheme (32) is superior.
The Pore-Velocity-Scheme (36) tends to smear more. In the case of too large timesteps
the Pore–Velocity–Scheme (36) computes zig–zag–solutions, resulting in a lot of smearing afterwards.

In consequence, scheme (32) seems to be an acceptable extension of the FIS/FISD to the non–equilibrium case.

Figure 1: Scheme (32); $t = 20 \cdot \Delta t$  
Figure 2: PVS (36); $t = 20 \cdot \Delta t$

Figure 3: Scheme (32); $t = 35 \cdot \Delta t$  
Figure 4: PVS (36); $t = 35 \cdot \Delta t$
The thick line displays the computed and the thin one the exact solution.
The figures 1, 2 present an example of situation 1 for \( k = 1.0, p = 0.6, h = 0.05, \Delta t = 0.737777, D = 0, q = 1. \)
The figures 3, 4 show an example of situation 2 for \( k = 10.0, K_d = 1.0, h = 0.05, \Delta t = 8 \cdot 0.073777, D = 10^{-8}, q = 1. \)
The exact solutions of situation 2 were computed with the package of Toride et al. [5].

References


