R^3T : RADIOACTIVE-RETARDATION-REACTION-TRANSPORT-PROGRAM FOR THE SIMULATION OF RADIOACTIVE WASTE DISPOSALS. *

JÜRGEN GEISER †

Abstract.
We present a software-tool for the simulation of a transport and reaction model for a potential waste scenarios of a radioactive waste-disposals. We introduce the complex model for the transport-, reaction- and sorption-processes. The model is based on systems of linear convection-diffusion-dispersion-reaction equations with equilibrium sorption. The discretisation is done with finite volume discretisation and a reconstruction with higher order polynoms. The convection-reaction term is explicit discretised with embedded analytical solutions. The diffusion-dispersion-term is discretised with implicit methods and the solvers use the BiCGStab with multi-grid methods. We focus us to the simulation of a 2d and 3d test case which we present with different grid- and time-step-refinements. The potential waste scenario for 2d and 3d is discussed.
At the end we will discuss the further works.

1. Introduction and mathematical model. The motivation of studying the presented model in this paper come from the simulation of a waste scenario of a radioactive contaminant transport in flowing groundwater [4, 6]. The idea is to study the simulation for the transportation of radioactive contaminants in 10000 years to have a further tool of predicting the waste scenario coming from a potential damage case.

For this studying we introduce our model. The model is based on the following equations

$$\partial_t R_i(c_i) \; + \; \nabla \cdot (\mathbf{v} c_i - D \nabla c_i) + \lambda_i R_i(c_i) \; = \; \sum_{e=e(i)} \lambda_e R_e(c_e) \; . \tag{1.1}$$

The unknown concentrations are $c_i = c_i(x,t)$ and are considered in $\Omega \times T$ where $\Omega \subset I\!R^n$, $T \subset I\!R^+$ and n is the space dimension. The parameters $R_i(c_i) \equiv (\phi + (1 - \phi)\, \rho K_d^i)\, c_i$ are the retardation factors. They delay the transport of the contaminants. Whereby ϕ is the porosity, ρ is the density and K_d is the Henry-sorption. We decided us for the equilibrium sorption for this model, confer [6]. The parameter $\mathbf{v}$ is the divergence free groundwater velocity, D is the dispersion-diffusion matrix and λ_i are the constant reaction-parameters. The index $e(i)$ describe the predecessor indices. We consider only decay-chains with one successor, but with possible more predecessors.

The aim of this paper is to describe the methods which are used to solve the equations. They are based on the methods used in [9] and [10]. The second part is to describe potential damage cases in 2d and in 3d to simulate a realistic case in a waste disposal. The methods and the used software-tools are considered and the computing time for the calculations. The results of the simulations are discussed and the new insights for such waste disposals. At the end of this paper we derive the future works for new simulations and the use of new discretisation and solver methods. We consider the software-tools as a control tool for first and carefully usability of future waste-disposal based on a salt-dome disposal and an overlying rock with clay and sand layers.

*This work is funded by the Federal Ministry of Economics and Technology (BMWi) under the contract number 02 E 9148 2

† Juergen.Geiser@IWR.Uni-Heidelberg.DE, Institute of Computer Science, Im Neuenheimer Feld 368, 69120 Heidelberg, Germany

2. Mathematical Methods for Solving the Equations. In this section we describe our used mathematical methods to solve the system of convection-diffusion-dispersion-reaction-equations. The idea is to solve such complex equations with decoupling the multi-physical and multidimensional equations in simpler physical and one-dimensional equations. This methods are based on the Operator-Splitting-methods, [17]. The simpler equations are discretised with the best adapted discretisation methods for this types of equation.
We distinguish between two time-discretisation. The explicit discretisation in time (forward Euler-method) is used for the convection-reaction equation and we get direct the solution, see [9]. The implicit discretisation in time (backward Euler-method) is used for the diffusion-dispersion-equation and use an iterative method to get the solution, see [8].

2.1. Operator-Splitting-Method. The operator-splitting-methods we used are based on the first and second order methods. The space-discretised terms are given with A and B.

$$\partial_t c = Ac + Bc \ ,$$

whereby the initial-conditions are $c(t^n) = c^n$. The splitting-method of the first order is done with

$$\begin{aligned} \partial_t c^* &= Ac^* \quad \text{with} \quad c^*(t^n) = c^n \ , \\ \partial_t c^{**} &= Bc^{**} \quad \text{with} \quad c^{**}(t^n) = c^*(t^{n+1}) \ , \end{aligned}$$

whereby the result of the methods are $c(t^{n+1}) = c^{**}(t^{n+1})$. We have a splitting error of the first order $O(\tau)$ if A, B do not commute, otherwise we get an exact solution. Where $\tau = t^{n+1} - t^n$ is the time-step. We could improve our results with a second order splitting-method, so called Strang-Splitting-method, [16], the error is $O(\tau^2)$. The underlying discretisation methods for the space discretisation are described in the following section.

2.2. Discretisation-Methods. The discretisation methods are based on the vertex-centered finite volume methods. First we describe the explicit discretisation of the convection equation. The equation is given as

$$\partial_t R(c) \ + \ \nabla \cdot (\mathbf{v}c) \ = \ 0 \tag{2.1}$$

with details for the simplest case $R(c) \equiv \phi c$. To simplify further our presentation we consider only trivial inflow and outflow boundary conditions where $c = 0$ is considered at inflow. The initial-conditions are $c(x, t^0) = c^0(x)$.
The integration over space and time are

$$\int_{\Omega_j} \int_{t^n}^{t^{n+1}} \partial_t (R\, c)\, dt\, dx = - \int_{\Omega_j} \int_{t^n}^{t^{n+1}} \nabla \cdot (\mathbf{v}\, c)\, dt\, dx \ ,$$

whereby Ω_j is the j-th cell with $\Omega = \cup_{j=1}^{I} \overline{\Omega}_j$.

$$|\Omega_j|(R(c_j^{n+1}) - R(c_j^n)) = -\tau^n \sum_{k \in out(j)} v_{jk} \tilde{c}_{jk}^n + \tau^n \sum_{l \in in(j)} v_{lj} \tilde{c}_{lj}^n \ ,$$

The discretisation-scheme with the mass-notation is given as

$$m_j^{n+1} - m_j^n = - \sum_{k \in out(j)} m_{jk}^n + \sum_{l \in in(j)} m_{lj}^n \ ,$$

whereby the inflow of j is $in(j) := \{k \in \Theta_j, v_{jk} < 0\}$, the outflow of j is $out(j) := \{k \in \Theta_j, v_{jk} \geq 0\}$ and Θ_j are the neighbour nodes of j. The masses are given as

$$m_j^n = V_j \; R \; c_j(t^n) \; , \; m_{jk}^n = \tau \; \tilde{c}_{jk}^n v_{jk} \; , \text{ whereby } v_{jk} = \mathbf{n}_{jk} \cdot \int_{\Gamma_{jk}} \mathbf{v}(\gamma) \; d\gamma \; .$$

We use the limitation to avoid oscillations, coming from over-shootings on discontinuous solutions. The limitation fulfil the monotonicity (local min-max-property) and we use the combination of two limiters. The first limiter is based on the slope and given as

$$\begin{aligned} 2c_i^n - \max_{k \in in(i)} \{c_i^n, c_k^n\} \leq c_{jk}^n \leq 2c_i^n - \min_{k \in in(i)} \{c_i^n, c_k^n\} \; , \; j \in out(i) \\ \min_{k \in in(i)} \{c_i^n, c_k^n\} \leq c_{ki}^n \leq \max_{k \in in(i)} \{c_i^n, c_k^n\} \; , \; k \in in(i) \end{aligned}$$

We get the limited values $\hat{c}_{jk}^n$.
The second limiter is based on the flux to avoid an overshooting in mass.

$$\begin{aligned} \tilde{c}_{jk}^n &= \hat{c}_{jk}^n + \frac{\tau}{\tau_j}(c_j^n - \hat{c}_{jk}^n) \\ \tau_j &= \frac{V_j}{\nu_j} \; , (\text{max. time-step with Cour.-Numb. 1}) \; , \; \nu_j = \sum_{k \in out(j)} v_{jk} \; . \end{aligned}$$

The limiters are described in [9].

On this higher order discretisation for the convection-term, we enlarge the discretisation for the reaction-term. We embed the analytical solution of a one-dimensional convection-reaction equation in the mass-discretisation, given above.

The equation has the following terms

$$\partial_t R_i c_i = \underbrace{-\nabla \cdot \mathbf{v} \; c_i}_{transport} \; \underbrace{-R_i \lambda_i c_i}_{sink} \; \underbrace{+R_{i-1} \lambda_{i-1} c_{i-1}}_{source} \; ,$$

whereby the initial-conditions are $c_1(x, t^0) = c_1^0(x)$ otherwise 0.0 .

The notation for the mass-terms in the systems are given as

$$m_{i,j}^{n+1} - m_{i,j}^n = - \sum_{k \in out(j)} m_{i,jk}^n + \sum_{l \in in(j)} m_{i,lj}^n \; , \tag{2.2}$$

whereby $m_{i,jk}^n$ is the mass of the concentration i from cell j to cell k for the transport- and reaction-term. The embedding of the mass for the one-dimensional convection-reaction equation is given in the following procedure.

We calculate the total-flux over all outflow-boundaries

$$\nu_j = \sum_{k \in out(j)} v_{jk} \; .$$

We calculate the velocity $v_{i,j}$ for every cell j over the norm-interval $(0, 1)$

$$v_{i,j} = \frac{1}{\tau_{i,j}} \; , \text{ with } \tau_{i,j} = \frac{V_j R_i}{\nu_j} \; , \text{ max. time-step with Cour.-number 1 .}$$

The analytical solution of the mass, confer [10] is calculated as

$$m_{i,jk,out}^n = m_{i2}(a, b, \tau^n, v_{1,j}, \ldots, v_{i,j}, R_1, \ldots, R_i, \lambda_1, \ldots, \lambda_i) \; ,$$

whereby $\tau^n \leq \min_{\substack{i=1,\dots,M \\ j=1,\dots,I}} \tau_{i,j}$ is the limitation of the time-steps, M is the number of components and I is the number of cells .

Further $a = V_j R_i (c^n_{i,jk} - c^n_{i,jk'})$, $b = V_j R_i c^n_{i,jk'}$ are the parameter for the linear initial-impulse for the finite-volume cell. Whereby $c^n_{i,jk'}$ is the concentration at left side of the cell j and $c^n_{i,jk}$ is the concentration at the right side of the cell j . The partial masses $m^n_{i,jk}$, described in equation (2.2) , are computed with the percentages of the total-masses with the outflow-boundaries

$$m^n_{i,jk} = \frac{v_{jk}}{\nu_j} \, m^n_{i,jk,out}$$

For the diffusion-dispersion-term we use an implicit discretisation in time and the underlying finite volume methods with the central method. The method is a higher order method, described in [10]. We have used a standard method for the diffusion-dispersion-term in [5].

2.3. Solvers for the implicit methods. For the solvers we use iterative methods based on multi-grid methods. We use the BiCGStab-method as solver and a multiplicative multi-grid method as pre-conditioner, described in [12] and [18]. We also use standard methods for the solving of the implicit methods and will not more focus on this topic.

In the next section we introduce our software-tool r^3t.

3. Software-tool r^3t. The mathematical methods described in the last sections are programmed in our software-toolbox r^3t, confer [4]. The concept is a script-based user-environment for which one could control the different test cases. The equation-parameters and the parameters for the domain-layers are done in scripts which are read in the start-process of the program. The underlying velocity-field of the domain is also read as an ASCII-file in runtime and is calculated by another software-tool called d^3f, [2]. The underlying equation are coupled flow- and transport equations for a density driven groundwater flow, confer [13]. The software-toolbox r^3t is based on the flexible software-tool ug, confer [1], which is a numerical program for computing on unstructed grids. This toolbox has a large amount of modules which are concerted to the discretisations and solvers for partial differential equations. We use the flexibility and the object-based programming of this tool. Two concepts are done in r^3t, one is the sparse-matrix concept, confer [14], which use the structure of the local discretisation and the structure for the reaction-matrix for an efficient storage of the dates. The second concept is the coupling concept, which is based on the exchange of input and output data-files. Therefore we could visualise our solutions with extern visualisation-tools, for which we use GRAPE [11]. We also could initialise our equation-parameters with data-files created from extern preprocessors for radioactive transport and reaction equations.

The applications of the software-tool r^3t as a control tool for radioactive waste disposals is described in the next sections.

4. Test-cases. In the test-cases we describe 2 potential test-cases one in 2d the other in 3d, because of the 3d effects. The cases are based on realistic dates, given by the project-partner GRS [3].

We have the following test case :

We have a transport of radioactive contaminants through an overlying rock, which is spooled with groundwater flow. This process should be simulated over a model time

for 10000 $[a]$. The contaminants are flown in at the bottom of the overlying rock and are time-dependent over the whole simulation.

The transport, done with the groundwater flow, and the dispersion in the overlying rock should be simulated with realistic parameters. The velocity is calculated with a density driven flow code (d^3f) and for the retardation we underly realistic parameters done with experiments. After the simulation time of 10000 $[a]$ one should have a knowledge about the transport and dispersion for of the contaminations.

The computing time should be done in 1-3 days, so that massive parallel computers are necessary.

This conditions are fulfilled with the r^3t software-tool and test cases are presented in the next sections.

4.1. 2d Test-case of a damage event. The test-case for two dimensions have the following description. The model-domain has the size of $6000[m] \times 150[m]$, with 4 different layers, [3]. The domain is flown through groundwater. It flows from right to left and from the top down to the middle of the bottom and up to the left boundary of the domain. The groundwater is flown faster through the permeable layers, confer figure 4.1. At the middle of the bottom we have a time-dependent inflow-source.

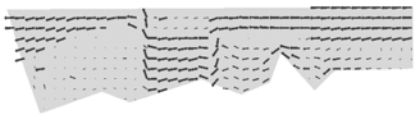

FIG. 4.1. *Velocity-field for the 2d test-case.*

With the stationary flow the contamination transport is calculated with r^3t and they are transported till the top of the overlying rock. The results of the calculations are introduced in the figure 4.2. In the left picture the concentration-flow of the Uran isotope $U-236$ after $100[a]$ is presented. The concentration-flow is transported with the the velocity-field and is spread out with the diffusion and dispersion. In the right figure the concentration flow after $10000[a]$ is presented. Therefore the concentration is flown through the permeable layer and is infiltrated through the impermeable layer because of the diffusion.

The two dimensional computations are done at Linux-PC-Cluster with Pentium II processors with a frequency of 0.4 GHz. The calculation is done with 26 radionuclids, which are coupled through the decay, confer equation (1.1) . The coarse-grid has 595 elements, we refined the domain till level 2 uniform and then the left part of the domain till level 4 uniform. Further computation is done adaptive till the level 6 with an error-estimator presented in [15]. The time-step is controlled by the Courant-number, confer equation (2.2) . The important times and refinements of the 2d computations are presented in table 4.1.

5. 3d test case of a damage event. The test-case for three dimensions have the following description. The model-domain has the size of $6000[m] \times 4000[m] \times$

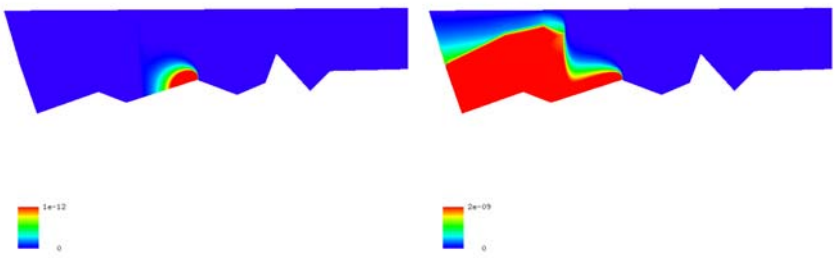

FIG. 4.2. *Concentration at* $100a$ *and end concentration at* $10000a$ *for the* $U-236$ *specie in 2d.*

Processors	Refinement	Number of elements	Number of time-steps	time for one time step	total time
30	uniform	75000	3800	5.0 sec.	5.5 h
64	adaptive	350000	3800	14.0 sec.	14.5 h

TABLE 4.1
Two dimensional calculation-times of a realistic potential damage event.

$1500[m]$, with 4 different layers, confer [3]. The domain is flown through groundwater from right to left and flows in the middle of the domain down to the bottom. The curls are situated in the middle and spooled the water to the top of the left side into 2 sinks situated at the top of the domain. The velocity-field is presented in figure 4.2 as a 2d cut-plan through the sinks. The arrows indicate the directions and strengths of the flow. The velocity-field is calculated with d^3f-tool, confer [13]. The 3d case is interested because of the curls done with the salted water. Therefore the 3d effects are carefully to study. These influences are important for the further understanding of the transported contaminants.

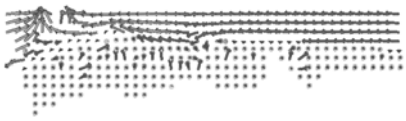

FIG. 5.1. *Velocity-field for the 3d test-case.*

The groundwater-flow transport the radioactive contaminants and this is calculated by the r^3t-tool, confer [10].

In the figure 5.2 we present the concentration of the important concentration $U-236$. The figure show the concentration in vertical and horizontal cut plans of the 3d domain. The concentration is situated in the middle of the bottom in the domain.

The red color is a high concentration about $10^{-9} - 10^{-7}$ and the blue color is no concentration. We have a time-dependent point-source in the whole simulation-time. The contaminants are flown over the impermeable layer and are spread out with the diffusion and dispersion, presented in figure 5.3.

The contaminants are transported to the sinks at the top in the left side of the domain. The end situation after 10000 $[a]$ is shown in figure 5.3.

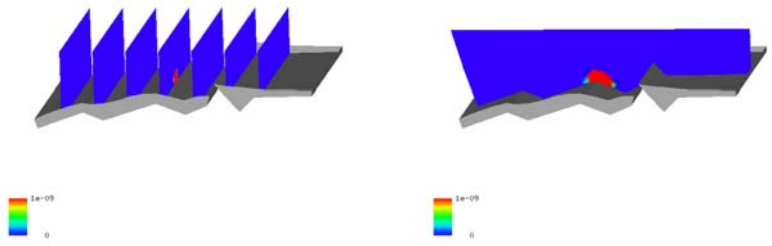

FIG. 5.2. *Concentration at* $100a$ *for the* $U - 236$ *specie in 3d.*

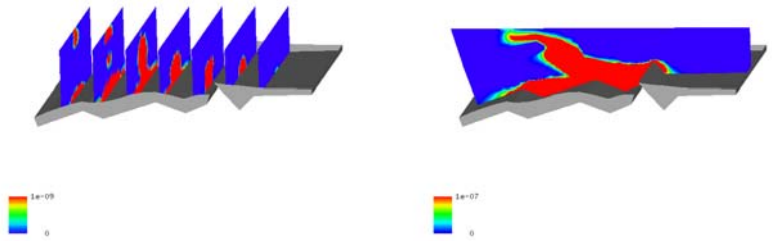

FIG. 5.3. *Concentration at* $10000a$ *for the* $U - 236$ *specie in 3d.*

The computations are done uniform and adaptive with massive parallel computers. We use a Linux-PC-Cluster with Athlon computers with a frequency of 1.6 GHz.

We have a coarse-grid of 8301 elements. In the uniform case we calculate the 26 components till the grid-level 2 uniform. The results are confirmed with the adaptive calculation, which are also done with 26 components. We refined till the level 2 uniform and till the level 5 adaptive. The time-steps are controlled with the Courant-number, confer equation (2.2) .

Processors	Refinement	Number of elements	Number of time-steps	time for one time step	total time
16	uniform	531264	3600	13.0 sec.	13.0 h
72	adaptive	580000	3600	18.5 sec.	18.5 h

TABLE 5.1
Three dimensional calculation-times of a realistic potential damage event.

6. Conclusions. We presented a new software-tool for the simulation of radioactive contaminant transport through porous media with groundwater flow. The tool is for predicting different scenarios for potential waste disposal. One could use it flexible as control tool together with other prediction methods. We have further presented the framework to get such a tool. We begin with the model-problem, the mathematical methods and end with the test-cases. Further works will be new mathematical methods, e.g. higher order discretisation methods and it should pay more attention to the physical model, e.g. kinetic sorption and nonlinear processes. We develop further the methods for massive parallel computers so that the complex simulations are computable.

REFERENCES

[1] P. Bastian, K. Birken, K. Eckstein, K. Johannsen, S. Lang, N. Neuss, and H. Rentz-Reichert. *UG - a flexible software toolbox for solving partial differential equations.* Computing and Visualisation in Science, 1(1):27–40, 1997.

[2] E. Fein, A. Schneider. *d^3f- Ein Programmpaket zur Modellierung von Dichteströmungen.* Abschlussbericht, Braunschweig, 1999.

[3] E. Fein. *Dates for examples for a potential damage case.* Private communications, Braunschweig, 2000.

[4] E. Fein, T. Kühle, and U. Noseck. *Entwicklung eines Programms zur dreidimensionalen Modellierung des Schadstofftransportes.* Fachliches Feinkonzept, Braunschweig, 2001.

[5] P. Frolkovič and H. De Schepper. *Numerical modelling of convection dominated transport coupled with density driven flow in porous media.* Advances in Water Resources, 24:63–72, 2001.

[6] P. Frolkovič and J. Geiser. *Numerical Simulation of Radionuclides Transport in Double Porosity Media with Sorption.* Proceedings of Algorithmy 2000, Conference of Scientific Computing, pp.28-36.

[7] P. Frolkovič. *Flux-based method of characteristics for contaminant transport in flowing groundwater.* Computing and Visualization in Science, to appear.

[8] J. Geiser. *Numerical Simulation of a Model for Transport and Reaction of Radionuclides.* Proceedings of the Large Scale Scientific Computations of Engineering and Environmental Problems, Sozopol, Bulgaria, 2001.

[9] P. Frolkovič and J. Geiser. *Discretization methods with discrete minimum and maximum property for convection dominated transport in porous media.* Proceeding of NMA 2002, Bulgaria, 2002.

[10] J. Geiser. *Diskretisierungsverfahren für Systeme von Konvektions- Diffusions-Dispersions-Reaktions-Gleichungen und Anwendungen.* Doktor-Arbeit, Universität Heidelberg, 2004.

[11] GRAPE. *GRAphics Programming Environment for mathematical problems, Version 5.4.* Institut für Angewandte Mathematik, Universität Bonn und Institut für Angewandte Mathematik, Universität Freiburg, 2001.

[12] W. Hackbusch. *Multi-Gird Methods and Applications.* Springer-Verlag, Berlin, Heidelberg, 1985.

[13] K. Johannsen. *Robuste Mehrgitterverfahren für die Konvektions-Diffusions Gleichung mit wirbelbehafteter Konvektion.* Doktor-Arbeit, Universität Heidelberg, 1999.

[14] N. Neuss. *A new sparse matrix storage methods for adaptive solving of large systems of reaction-diffusion-transport equations.* In Keil et. al., editor, Scientific computing in chemical Engineering II, Springer-Verlag Berlin-Heidelberg-New York, 175–182, 1999.

[15] M. Ohlberg. *A Posteriori Error Estimates for Vertex Centered finite Volume Apprximations of Convection-Diffusion-Reaction equations.* Preprints 12/2000, Mathematische Fakultät, Freiburg, May 2000.

[16] G. Strang. *On the construction and comparision of difference schemes.* SIAM J. Numer. Anal., 5:506–517, 1968.

[17] J.,G. Verwer and B. Sportisse. *A note on operator splitting in a stiff linear case.* MAS-R9830, ISSN 1386-3703, 1998.

[18] G. Wittum. *Multi-grid Methods : A introduction.* Bericht 1995-5, Institut für Computeranwendungen der Universität Stuttgart, Juni 1995.