

INVERSE PROBLEM OF NON-ISOTHERMAL MULTIPHASE FLOW AND REACTIVE TRANSPORT IN DEFORMABLE POROUS MEDIA

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ABSTRACT. We present a methodology for solving the inverse problem of non-isothermal multiphase flow and multicomponent reactive solute transport which can take into account various types of data including heads or pressures, temperatures, stress/deformation, aqueous and total (dissolved plus sorbed) concentrations, water fluxes, water contents and exchanged cations as well as parameter prior information. The inverse problem is solved by minimizing a generalized least-squares criterion with a Gauss-Newton-Levenberg-Marquardt method. Approximate confidence intervals are computed from the parameter covariance matrix. All these features have been implemented in INVERS-FADES-CORE, a general code which solves both forward and inverse problems. The code can estimate a wide range of parameters including: intrinsic permeability, parameters of retention and relative permeability curves, parameters of the constitutive model of deformation, tortuosity of vapor, thermal conductivity, dispersivity, distribution coefficient, molecular diffusion coefficient, initial and boundary concentrations, pH and pE, selectivity coefficients, cation exchange capacity, specific surface of minerals and initial volume fraction of minerals. Synthetic data have been used to verify the formulation and study the convergence, uniqueness and stability of the algorithm. The methodology has been applied to the estimation of transport and chemical parameters from both laboratory and in situ tests performed during the FEBEX project (Full-scale Engineered Barrier Experiment).

RESUMEN. Se presenta una metodología numérica para resolver el problema inverso del flujo multifásico no isoterma y transporte de solutos reactivos a través de medios porosos deformables que tiene en cuenta una amplia gama de diferentes datos: niveles o presiones, temperaturas, tensiones/deformaciones, concentraciones acuosas y totales (disuelta más adsorbida), caudales, contenidos humedad así como información previa de los parámetros. El problema inverso se resuelve minimizando una función objetivo mediante el algoritmo de Gauss-Newton-Levenberg-Marquardt. La incertidumbre de los parámetros estimados se calcula mediante intervalos de confianza aproximados de los parámetros estimados. La formulación se ha implementado en INVERS-FADES-CORE, un código que

resuelve tanto el problema directo como el inverso. El código permite la estimación de distintos tipos de parámetros incluyendo: permeabilidad intrínseca, parámetros de la curva de permeabilidad relativa, parámetros de la curva de retención, parámetros de ecuaciones constitutivas para el comportamiento tenso-deformacional del suelo, tortuosidad de los gases y del vapor, conductividades térmicas, dispersividad, coeficiente de distribución, coeficiente de difusión molecular, concentración inicial y de contorno, pH y pE, coeficiente de intercambio, capacidad de intercambio catiónico, superficie específica de mineral y fracciones iniciales de minerales. Se han utilizado datos sintéticos para verificar la formulación y estudiar la convergencia, unicidad, y estabilidad del algoritmo inverso. La metodología se ha aplicado también a la estimación de los parámetros de la bentonita compactada utilizada en los ensayos de laboratorio e in situ del proyecto FEBEX..

1. Introduction

Compacted bentonites are being used in several countries as backfill and sealing material for high-level radioactive waste (HLW) disposal. The study of thermo-hydro-mechanical-geochemical (THMC) processes in the bentonite barrier is a key point in the performance assessment. Coupled inverse THMC models greatly enhance our ability to study THMC processes and estimate key parameters. Existing THMC codes include TOUGH2-CHEM by White (1995), TOUGHREACT by Xu and Pruess (1998), RETRASO by Saaltink et al. (1997; 2004), MULTIFLO by Lichtner (1996) and CRUNCH by Steefel (2001) which simulate multiphase THMC processes by incorporating reactive transport in a preexisting multiphase flow code. Multiphase flow codes have been widely developed within the realm of soil mechanics. Some of them solve for non-isothermal multiphase flow coupled with soil deformation. Among them, we can mention CODE_BRIGHT (Olivella, 1996) and FADES (Navarro,

1997; Navarro & Alonso, 2000). Recently, CODE_BRIGHT has been updated to include geochemical calculations by Guimarães (2002).

FADES-CORE is a THMC code developed within the FEBEX project (ENRESA, 2000; Juncosa et al., 2002) by coupling FADES (Navarro, 1997) and CORE (Samper et al., 2000). CORE is a finite element CODE for modeling water flow saturated or unsaturated, heat transport and multi-component REactive solute transport under local chemical equilibrium which uses the sequential iterative approach. The code accounts for a wide range of chemical processes such as aqueous complexation, acid-base, redox, mineral dissolution and precipitation, gas dissolution/exsolution, cation exchange, and adsorption via surface complexation. This code has been widely used in several research projects dealing with: 1) Radioactive waste disposal at laboratory and field experiments (Samper et al. 2000; 2005a;b,c,d; Dai and Samper, 2004; Molinero and Samper, 2004; Samper et al., 2004), 2) Groundwater pollution studies, 3) Coupled geo-microbial models (Molinero et al., 2004), 4) Evaluation of impacts of underground civil works; and 5) Geothermal systems.

FADES is based on a complete analysis of the structure of soil and multiphase flow and allows the simulation of the coupled thermo-hydro-mechanical behavior of partially saturated soils (Navarro, 1997). FADES-CORE can model the multiphase thermo-hydrodynamic behavior of partially saturated soils taking into account solute transport and chemical reactions. Recently, FADES-CORE has been extended with many new features such as a constitutive law for permeability depending on the salinity of pore water, the use of Pitzer equations to calculate activity coefficients for high salinity solutions and double porosity structures (Samper et al., 2005d).

Inverse algorithms have been used to derive soil hydraulic parameters from transient infiltration events (Yeh et al., 1990; Russo et al., 1991; Eching et al., 1994; Simunek and van Genuchten, 1996; Inoue et al., 1998; Pan and Wu, 1998). Inverse modeling of flow and solute transport has also been extensively performed (Kool et al., 1987; Gailey et al., 1991; Wagner, 1992; Xiang et al., 1993; Medina and Carrera, 1996;). The inverse problem of reactive solute transport has been traditionally solved by trial and error (Appelo et al., 1998; Keating and Bahr, 1998a; Keating and Bahr, 1998b; Saaltink et al., 1998). Recently Dai and Samper (1999), Dai (2000), Dai and Samper (2004) and Dai et al. (2005) have developed an automatic inverse code, INVERSE-CORE. INVERS-FADES-CORE is based on the formulation of FADES-CORE and uses the inverse algorithm of INVERSE-CORE.

INVERS-FADES-CORE has been used to estimate transport and chemical parameters from a heating and hydration laboratory experiment and an in situ test. The governing equations of multiphase flow reactive solute transport are presented first. Then, the formulation of the inverse algorithm is described. Code verification and implementation are discussed. Finally, the application to laboratory and in situ experiments is presented.

2. Formulation of non-isothermal multiphase flow and reactive transport

The mathematical formulation of non-isothermal multiphase flow and deformation of INVERS-FADES-CORE is that of FADES-CORE (see Navarro, 1997; Juncosa et al, 2002).

The solute transport model accounts for advection, molecular diffusion and mechanical dispersion. Each of these processes produces a solute flux per unit surface and time. There are as many transport equations as chemical components (primary species) in the system. The mass balance equation for the j-th component is given by:

$$\rho_i X_i^w \theta_i \frac{\partial C_j}{\partial t} + \frac{\partial(\rho_i X_i^w \theta_i P_j)}{\partial t} + \frac{\partial(\rho_i X_i^w \theta_i W_j)}{\partial t} + \frac{\partial(\rho_i X_i^w \theta_i Y_j)}{\partial t} = L^*(C_j) + r_i(C_j^0 - C_j) \quad (1)$$

where

$$L^*(\cdot) = \nabla \cdot (\rho_i X_i^w \theta_i \mathbf{D}^j \nabla(\cdot)) - \rho_i X_i^w \mathbf{q}_i \nabla(\cdot) + (r_e - r_c)(\cdot) \quad (2)$$

C_j is total dissolved concentration of j-th species, r_e is evaporation rate, r_i is sink term, C_j^0 is dissolved concentration of j-th species in the sink term r_i , W_j is total exchanged concentration of j-th component, \mathbf{D}^j is dispersion coefficient, Y_j is total sorbed concentration of j-th component, N_c is number of solutes, P_j is precipitated concentration of j-th component, and θ is water content.

The chemical system is defined by the concentration of primary species from which it is possible to compute the concentration of secondary species by means of appropriate mass balance and mass action equations. Concentrations of mineral phases, exchanged and adsorbed species can be obtained using similar equations. Sink/source chemical terms in the transport equation take into account the amount of mass gained or lost due to mineral dissolution and precipitation, ion exchange and adsorption. Relevant equations linking primary to secondary species as well as those for adsorbed, exchanged and precipitated species can be found in Samper et al. (2000).

3. Formulation of the inverse problem

INVERS-FADES-CORE uses the same inverse algorithm adopted in INVERSE-CORE (Dai, 2000). The inverse problem is solved by minimizing a generalized least-squares criterion with a Gauss-Newton-Levenberg-Marquardt method. Let $\mathbf{P} = (p_1, p_2, p_3, \dots, p_M)$ be the unknown parameter vector defined by a parameterization method, then the least squares criterion $E(\mathbf{p})$ can be expressed as:

$$\begin{aligned}
 E(p) &= \sum_{i=1}^{NE} W_i E_i(p) \\
 &= \sum_{i=1}^{NE} W_i \sum_{l=1}^{L_i} w_l^2 [u_l(p) - F_l]^2 \\
 &= \sum_{i=1}^{NE} W_i \sum_{l=1}^{L_i} w_l^2 r_l^2(p)
 \end{aligned}
 \tag{3}$$

where NE is the number of different types of data such as liquid pressure, dissolved concentration, total concentration, water flux, water content and prior information of the parameters to be estimated; $E_i(p)$ is the least-squares for each type of data; W_i are weighting coefficients for each type of data and prior information; w_l are weighting coefficients for different observation points. Their values depend on the accuracy of the observations. $u_l(p)$ is the value of the variable computed with the numerical model; F_l are observed values and prior information of the parameters; L_i is the number of observation points in the space-time domain; and r_l are residuals or differences between model outcomes and actual field or laboratory measurements. Equation (3) is solved with a Gauss-Newton-Levenberg-Marquardt method (see Dai, 2000; Dai and Samper, 2004).

4. Implementation of the formulation

The main structure of INVERS-FADES-CORE is shown in Figure 1. The program starts by reading all relevant data for inverse modeling which includes control parameters, lower and upper bounds, initial values, and prior information of the parameters, as well as all kinds of measured data. Then, it continues by reading the data for the forward modeling of multiphase flow, heat and solute transport. At the beginning of a run of Gauss-Newton-Marquardt-Levenberg iteration, it is necessary to provide a set of initial parameter values. These are the values that the algorithm uses to start the first optimization iteration. At each iteration, the Jacobian matrix is calculated using finite differences (forward or central difference method). After solving the Gauss-Newton- Marquardt-Levenberg equation, a set of updated parameters is obtained. The forward model FADES-CORE is called again to calculate an improved objective function value. By comparing parameter changes and objective function improvement achieved through the current iteration with those achieved in previous iteration; the algorithm can tell whether it is worth undertaking another optimization iteration. If so, the whole process is repeated. Finally, the program stops when a convergent solution is attained or when the maximum number of iterations is exceeded.

INVERS-FADES-CORE enables to estimate a wide range of parameters by taking into account different types of observations including: 1) liquid and gas pressure, 2) concentrations of chemical species, 3) total concentration in solid and liquid phase, 4) water inflows, 5) water content, 6) exchanged cations, 7) temperature, 8) total stress 9) porosity. The following parameters can be estimated:

1. Intrinsic permeability and the parameters of the relative permeability and retention curves
 2. Vapor tortuosity and diffusion coefficient,
 3. Thermal conductivity of the medium,
 4. Dispersivity,
 5. Accessible porosity, which may be different from total porosity if ion exclusion is considered,
 6. Distribution coefficient,
 7. Initial concentrations and pH and pE,
 8. Boundary concentrations of chemical species,
 9. Selectivity coefficients of exchangeable cations,
 10. Cation Exchange Capacity (CEC),
 11. Specific surface of minerals,
 12. Initial volume fraction of minerals,
- Parameters of mechanical modules..

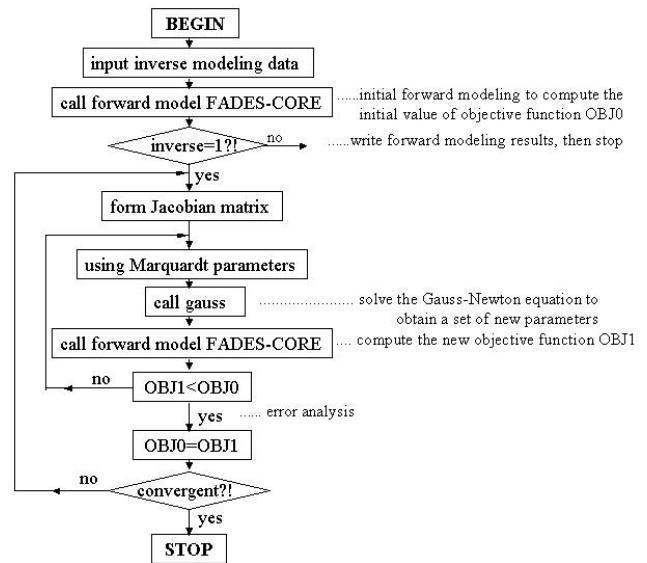


Fig. 1. Flow chart of INVERSE-FADES-CORE main program.

5. Code verification

Verification of the inverse routines of INVERS-FADES-CORE has been focused on the study of convergence, uniqueness and stability of the inverse algorithm and the estimation of different types of parameters with different observations. Synthetic data have been used to verify the estimation capabilities. Then the code has been applied to the estimation of transport and chemical parameters of heating and hydration laboratory and in situ experiments.

5.1. Estimation of soil deformation parameters

Navarro (1997) used a one dimensional flow and displacement model to verify FADES. Here his modeling results are used as synthetic data. Based on them, parameters of void ratio state surface were estimated. The void ratio state surface is given by:

$$e = 1.1849 + A \cdot \log(\sigma') + B \cdot \log(s) + C \cdot \log(\sigma') \log(s) \tag{4}$$

where e is void ratio, σ' is effective stress, s is suction, and A , B and C are empirical parameters for the state surface.

Liquid pressures and tensions are considered in the objective function. Parameters A , B and C in Equation (4) are estimated simultaneously. For noise-free synthetic data, estimated values coincide with the true values. For noise-corrupted data with a standard deviation of 0.05, estimated parameters are similar to the true values. Estimated results for noisy data are listed in Table 1

Table 1. Estimated values of parameters A , B and C in Equation (4).

Parameter	Initial guess	Estimated value	True value	Confidence Interval
A	0.153	0.2080	0.2062	(0.1994, 0.2166)
B	-0.06	-0.1261	-0.1385	(0.085, 0.1502)
C	0.015	0.07407	0.0741	($7.385 \cdot 10^{-2}$, $7.43 \cdot 10^{-2}$)

5.2. Estimation of Multiphase Flow and Transport parameters

Synthetic data were taken from the modeling results of a forward model. This forward model is a non-isothermal multiphase conservative solute transport model in 1-D column. It deals with the transport of chloride through a bentonite barrier. Water enters into bentonite with a prescribed pressure through the left boundary. Using liquid pressure and chloride concentration data, the following parameters were estimated: 1) intrinsic permeability of liquid, 2) parameters of relative permeability curve, 3) parameters of the retention curve, 4) tortuosity of vapor, 5) solute molecular diffusion coefficient, and 6) accessible porosity for chloride. These parameters were estimated first one-by-one and then all simultaneously. When only one parameter is estimated, INVERS-FADES-CORE provides excellent estimation results which are insensitive to the initial guess. However, when more than one parameter is estimated, the goodness of estimation depends on the correlation of these parameters. For example, when diffusion coefficient D_0 and accessible porosity ϕ_a are estimated simultaneously, good estimates are obtained (Table 2). However, when diffusion coefficient and intrinsic permeability are estimated simultaneously, poor estimation results were obtained due to their large correlation.

Table 2. Estimation results when diffusion coefficient and accessible porosity are estimated simultaneously. True values of these two parameters are $2.0 \cdot 10^{-8}$ m²/s and 0.75 respectively. Accessible porosity is expressed as a fraction of total porosity.

Parameter	Initial guess	Estimated value	Standard deviation	Confidence Interval
D_0 (m ² /s)	$2.5 \cdot 10^{-9}$	$2.0 \cdot 10^{-8}$	$2.61 \cdot 10^{-12}$	($1.996 \cdot 10^{-7}$, $1.998 \cdot 10^{-7}$)
ϕ_a	0.55	0.75	$2.65 \cdot 10^{-5}$	(0.75, 0.7502)
D_0 (m ² /s)	$1.5 \cdot 10^{-7}$	$2.0 \cdot 10^{-8}$	$2.61 \cdot 10^{-12}$	($1.996 \cdot 10^{-7}$, $1.998 \cdot 10^{-7}$)
ϕ_a	0.95	0.75	$2.65 \cdot 10^{-5}$	(0.75, 0.7502)

Note: True values of these two parameters are $2.0 \cdot 10^{-8}$ m²/s and 0.75 respectively, accessible porosity is expressed as a fraction of total porosity.

In the second step, we analyze the estimation of chemical parameters. The following chemical processes are

considered: aqueous complexation, acid-base, mineral dissolution/precipitation and cation exchange. 10 primary species are considered. Both noise-free synthetic data and noise-corrupted data with different standard deviations were considered in the objective function. Initial concentrations of Mg^{2+} , Na^+ , K^+ , Cl^- , SO_4^{2-} and HCO_3^- are estimated. Since Ca^{2+} is initially in equilibrium with calcite, its initial concentration is not estimated. With noise-free synthetic data, estimated values are equal to the true values. With noise-corrupted data with a standard deviation of 0.05, estimated parameters are similar to the true values (Table 3).

Table 3. Estimation results of initial pore water concentrations (mol/L).

Parameter	Initial guess	Estimated value	Standard deviation	Confidence Interval
Mg^{2+}	$1.44 \cdot 10^{-2}$	$1.48 \cdot 10^{-2}$	$3.48 \cdot 10^{-6}$	($1.47 \cdot 10^{-2}$, $1.48 \cdot 10^{-2}$)
Na^+	$1.88 \cdot 10^{-1}$	$1.92 \cdot 10^{-1}$	$1.01 \cdot 10^{-5}$	($1.91 \cdot 10^{-1}$, $1.92 \cdot 10^{-1}$)
K^+	$1.68 \cdot 10^{-3}$	$1.72 \cdot 10^{-3}$	$9.48 \cdot 10^{-7}$	($1.71 \cdot 10^{-3}$, $1.72 \cdot 10^{-3}$)
Cl^-	$1.87 \cdot 10^{-1}$	$1.91 \cdot 10^{-1}$	$1.19 \cdot 10^{-5}$	($1.91 \cdot 10^{-1}$, $1.92 \cdot 10^{-1}$)
SO_4^{2-}	$2.12 \cdot 10^{-2}$	$2.16 \cdot 10^{-2}$	$3.59 \cdot 10^{-6}$	($2.16 \cdot 10^{-2}$, $2.17 \cdot 10^{-2}$)
HCO_3^-	$5.8 \cdot 10^{-4}$	$5.74 \cdot 10^{-4}$	$1.31 \cdot 10^{-7}$	($5.73 \cdot 10^{-4}$, $5.74 \cdot 10^{-4}$)

6. Iodide migration in the FEBEX 'in situ' test

INVERS-FADES-CORE has been applied to a thermo-hydraulic cell experiment in which optimum values of diffusion coefficient and accessible porosity have been obtained (Zheng and Samper, 2004). Here it has been applied to the large scale in situ test.

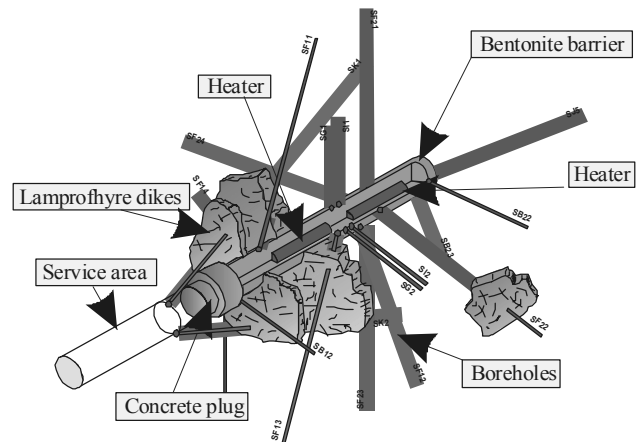


Fig. 2. Scheme of the "in situ" test

The in situ test is one of the two main experiments in the FEBEX project (ENRESA, 2000). The in situ test is being performed in a gallery excavated in the northern zone of the Grimsel underground laboratory, operated by NAGRA in Switzerland (Figure 2). The test began in February 27th, 1997. Heater 1 was switched-off in February 2002. Dismantling of section 1 of the in situ test was performed from May to September 2002. A comprehensive post-mortem bentonite sampling and analysis program was designed. Iodide was emplaced in filter paper located on the outer rings of the bentonite barrier at the central section of heater 1 during the installation phase. Concentrations of

iodide were analyzed in clay samples along a radius of the clay buffer during the dismantling in order to study the migration processes during the heating and hydration of the bentonite barrier.

A one dimensional axisymmetric THG model was set up to predict iodide migration. In the model it was assumed that the tracer was readily available to migration in bentonite at $t = 0$. This was achieved by assigning a high initial iodide concentration to the nodes located at the bentonite-granite interface.

An inverse model (inverse model 1) was used to derive optimum estimates of the diffusion coefficient and accessible porosity. Estimated results are listed in Table 4. It should be noticed that accessible porosity is expressed as its ratio to the total porosity, for example, 0.614 means accessible porosity is 61.4% of total porosity. Figure 3 shows the comparison of computed concentration to measurements for the optimum parameter values.

Table 4. Estimated diffusion coefficient and accessible porosity in inverse model 1.

Parameters	Estimated values.	Variance	Confidence interval (95%)
D_0	$8.3 \cdot 10^{-13}$	$1.81 \cdot 10^{-22}$	$(0.12 \cdot 10^{-13}, 0.33 \cdot 10^{-10})$
Accessible porosity	0.614	$1.42 \cdot 10^{-4}$	(0.58, 0.64)

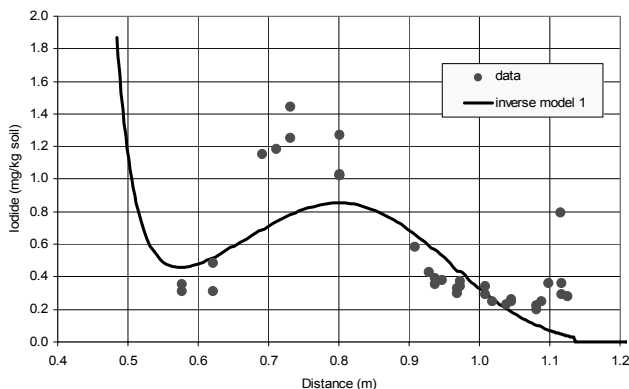


Fig. 3. Inverse modelling results of inverse model 1.

Figure 3 indicates that the model computes too much evaporation near the heater. Therefore, another inverse run was performed in which, in addition to diffusion coefficient and accessible porosity vapor tortuosity was also estimated (inverse model 2). Estimated parameters are listed in Table 5. Figure 4 shows the comparison of measured data and modeling results. The fit of computed results to measured data improves when vapor tortuosity is also estimated.

Table 5. Estimated parameters in inverse model 2.

Parameters	Estimated values.	Variance	Confidence interval (95%)
D_0	$1.3 \cdot 10^{-12} \text{ m}^2/\text{s}$	$1.03 \cdot 10^{-21}$	$(0.83 \cdot 10^{-12}, 0.91 \cdot 10^{-10})$
Accessible porosity	0.572	$4.60 \cdot 10^{-4}$	(0.51, 0.63)
Vapor tortuosity	0.099	$4.56 \cdot 10^{-8}$	(0.098, 0.099)

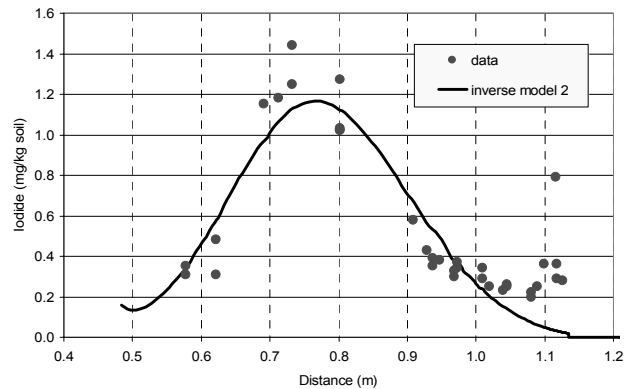


Fig. 4. Comparison of measured data and inverse modelling results.

7. Conclusions

A numerical formulation of the inverse problem of non-isothermal multiphase flow and reactive transport in deformable media has been presented. The formulation has been implemented in INVERS-FADES-CORE, a code that can estimate a wide range of thermal, hydrodynamic, mechanical, transport and geochemical parameters using head, tension, concentration and temperature data. The formulation has been verified with synthetic examples and has been used to estimate flow and transport parameters of compacted bentonite from heating and hydration laboratory and in situ tests. The inverse algorithm has proved to be an efficient tool to derive optimum estimates of THMC model parameters.

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