

Modelling of experiment BenchMark 1.3

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Abstract: Danger waste disposal systems are typically comprised a series of barriers that act to protect the environment and human health. The presence of several barriers enhances confidence that the waste will be adequately contained. This article is focused on one of barriers, bentonite buffer. Bentonite buffer intervene between the waste canister and the host rock. One of main tasks of the bentonite buffer is to reduce and delay water flow from the host rock in order to prevent the contact of water with the waste canister as long as possible. Model, described in this article, solve coupled task of unsteady heat and water vapor flow in bentonite. In the end, model results are compared with experimental results.

Key-Words: Modelling, bentonite, coupled processes, unsteady heat flow, unsteady water vapor flow

1 Introduction

The major problem of dangerous (radioactive or toxic) waste repositories is water which present in the host rock [3], [4]. The water can not only jeopardize the integrity of the waste canister, but also, form possible pathway to the biosphere for the hazardeous substances.

The problem of resaturation of the initially air-dry bentonite is very complex. This phenomenon is influenced by hydraulic, mechanic, thermal and chemical processes. These processes are coupled by a series of parameters and equations. Some of them are highly nonlinear and some of them are not yet completely understood.

Many scientists have worked with models, which solve this problem using Darcy law. But these type of

models do not reflect effect of water vapor. In this article is presented model, which solve this task in a different way. This model is based on water vapor diffusion. Model solve the unsteady coupled heat and mass problem by using primal formulation of finite element method.

2 Problem formulation

Heat and mass transportation parameters and the distribution of moisture and temperature within porous materials are based on the energy and moisture conservation equations during the transportation. The transfer is described by partial differential equations derived from the set of equations published by Henry [6]

$$\begin{aligned}
 c_v(T, C_a, C_b) \frac{\partial T}{\partial t} - \chi(T, C_a, C_b) \frac{\partial C_b}{\partial t} &= \nabla \cdot (\lambda(T, C_a, C_b) \nabla T) \quad , \\
 \epsilon \frac{\partial C_a}{\partial t} + (1 - \epsilon) \frac{\partial C_b}{\partial t} &= \nabla \cdot \left(\frac{\epsilon}{\tau} D_a(T, C_a, C_b) \nabla C_a \right) \quad , \\
 \frac{1}{\epsilon} \frac{\partial C_b}{\partial t} &= \left(\frac{C_a}{C_a^{100}(T)} - \varphi(T, C_a, C_b) \right) \gamma(T, C_a, C_b) \quad .
 \end{aligned} \tag{1}$$

Domain of task is denoted Ω with boundary Γ . The problem is discreted in the space variable $\mathbf{x} = \{x, y\}$.

Unknown variables are T temperature, C_a water vapor concentration in the air between bentonite grains and C_b water vapor concentration in the bentonite grains.

Furthermore t time, $c_v(T, C_a, C_b)$ volumetric heat capacity, $\chi(T, C_a, C_b)$ latent heat of sorption or adsorption of water vapor by solid grains (it gives information on interaction forces between the water vapor molecules and the sorbent surface-binding energy), $\lambda(T, C_a, C_b)$ thermal conductivity, ϵ porosity of bentonite, τ effective tortuosity (it is related to the hindrance imposed on diffusing particle by the bentonite grains), $D_a(T, C_a, C_b)$ diffusion coefficient of water vapor in the air, $C_a^{100}(T)$ water vapor concentration for 100% relative humidity (RH) in the air, $\varphi(T, C_a, C_b)$ inversion sorption curve, and $\gamma = \gamma(T, C_a, C_b)$ general function defining sorption/adsorption speed of bentonite.

Material parameter c_v , χ , λ , D_a , C_a^{100} , γ and φ are assumed as functions and must be positive and finiteness.

2.1 Boundary conditions

The set of boundary conditions is added. In the model are assume three kind of boundary conditions. Splitting of boundary Γ on three disjunction parts $\Gamma = \Gamma_1 \cup \Gamma_2 \cup \Gamma_3$ is assumed. \mathbf{n} is outter normal of boundary Γ .

Dirichlet boundary condition defined temperature $T_D(t)$, eventually water vapor concentration $C_D^a(t)$ on boundary Γ_1 ,

$$\begin{aligned} T(\mathbf{x}, t) &= T_D(t) \\ C_a(\mathbf{x}, t) &= C_D^a(t) \end{aligned} \quad \mathbf{x} \in \Gamma_1 \quad . \quad (2)$$

Neumann boundary condition defined heat flow $q_T(t)$, eventually water vapor flow $q_{C_a}(t)$ through boundary Γ_2 ,

$$\begin{aligned} \lambda \nabla T(\mathbf{x}, t) \cdot \mathbf{n} &= q_T(t) \\ D_a \nabla C_a(\mathbf{x}, t) \cdot \mathbf{n} &= q_{C_a}(t) \end{aligned} \quad \mathbf{x} \in \Gamma_2 \quad . \quad (3)$$

Newton boundary condition defined heat flow, eventually water vapor flow generated by heat drop $T - T_W(t)$, eventually by water vapor concentration drop $C_a - C_W^a(t)$ on boundary Γ_3 ,

$$\begin{aligned} \lambda \nabla T \cdot \mathbf{n} + \sigma_T(t)(T - T_W(t)) &= 0 \\ D_a \nabla C_a \cdot \mathbf{n} + \sigma_{C_a}(t)(C_a - C_W^a(t)) &= 0 \end{aligned} \quad \mathbf{x} \in \Gamma_3 \quad , \quad (4)$$

where $\sigma_T(t) > 0$ and $\sigma_{C_a}(t) > 0$.

2.2 Initial conditions

Initial conditions are defined by general functions on domain Ω . These function can be known, for example constant functions, or can be obtained by solutions of steady task.

$$\begin{aligned} T(\mathbf{x}, 0) &= T_o(\mathbf{x}) \\ C_a(\mathbf{x}, 0) &= C_o^a(\mathbf{x}) \\ C_b(\mathbf{x}, 0) &= C_o^b(\mathbf{x}) \end{aligned} \quad \mathbf{x} \in \Omega \quad . \quad (5)$$

2.3 Weak formulation

To use the finite element method, the weak formulation has to be derived. Let be $H_0(\Omega) = \{f \in W_2^1(\Omega), f|_{\Gamma} = 0\}$ the space of testing functions. Further, is denoted the scalar products as $(\varphi, \psi) = \int_{\Omega} \varphi \psi d\Omega$, $\langle \varphi, \psi \rangle = \int_{\Gamma} \varphi \psi d\Gamma$. Equations in set of equations (1) are multiplied by testing function $w \in H_0(\Omega)$ and integrated over Ω . Then a Green formula is used and substitution of boundary conditions gives integral identities

$$\begin{aligned} \left(c_v \frac{\partial T}{\partial t}, w \right) - \left(\chi \frac{\partial C_b}{\partial t}, w \right) &= \left\langle \lambda \nabla T \cdot \mathbf{n}, w \right\rangle - (\lambda \nabla T, \nabla w), \\ \left(\epsilon \frac{\partial C_a}{\partial t}, w \right) + \left((1 - \epsilon) \frac{\partial C_b}{\partial t}, w \right) &= \left\langle \frac{\epsilon}{\tau} D_a \nabla C_a \cdot \mathbf{n}, w \right\rangle - \left(\frac{\epsilon}{\tau} D_a \nabla C_a, \nabla w \right), \\ \left(\frac{1}{\epsilon} \frac{\partial C_b}{\partial t}, w \right) &= \left(\gamma \frac{C_a}{C_a^{100}}, w \right) - (\gamma \varphi, w). \end{aligned} \quad (6)$$

The problem is solved in time interval $I = \langle 0, t^* \rangle$. Then $T^*, C_a^*, C_b^* \in AC(I, W_2^1(\Omega))$ is denoted as the function fulfilling the Dirichlet boundary conditions (2). Let

$$\begin{aligned} T(x, t) &= T^*(x, t) + T_0(x, t), \\ C_a(x, t) &= C_a^*(x, t) + C_{a0}(x, t), \\ C_b(x, t) &= C_b^*(x, t) + C_{b0}(x, t), \end{aligned}$$

where $T_0, C_{a0}, C_{b0} \in AC(I, H_0(\Omega))$. Then functions T, C_a, C_b are the weak solution of set of equations (1) with boundary conditions (2)-(4) and initial conditions (5) in time interval I , if they fulfill the identities (6) for arbitrary $w \in H_0(\Omega)$.

Existence of integrals in (6) is allowed by finiteness of functions $c_v, \chi, \lambda, D_a, C_a^{100}, \gamma$ and φ .

2.4 Spatial discretization

For spatial discretization the tetrahedrons with linear base functions are used (see figure 1).

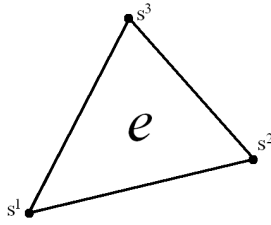


Figure 1: Element of discretization

Area Ω is then approximated by the set Ω^h ,

$$\Omega^h = \bigcup_{e \in E^h} e,$$

where E^h is the set of all discretization nodes. On every simplex e with nodes (s^1, s^2, s^3) , three base functions are established,

$$w_i = \alpha_0^i + \alpha_1^i x_1 + \alpha_2^i x_2 \quad ,$$

$i = 1, 2, 3$. They fulfill the condition $w_i(s^j) = \delta_{ij}$. The approximation of weak solution is looked for in the form (r is number of nodes)

$$\begin{aligned} T^h(\mathbf{x}, t) &= \sum_{i=1}^r T^i(t) w_i(\mathbf{x}), \\ C_a^h(\mathbf{x}, t) &= \sum_{i=1}^r C_a^i(t) w_i(\mathbf{x}), \\ C_b^h(\mathbf{x}, t) &= \sum_{i=1}^r C_b^i(t) w_i(\mathbf{x}). \end{aligned} \quad (7)$$

Coefficients $T^i(t), C_a^i(t), C_b^i(t)$ are values of unknowns at the nodes of discretization in time t . The approximations (7) are entered into identities(6). Then is searched for their fulfilment for all base functions $w_j, j \in \hat{r}$. The resulting system of ordinary differential equations has the block structure

$$\begin{pmatrix} \mathbb{B}_T & 0 & \mathbb{C}_T \\ 0 & \mathbb{B}_{C_a} & \mathbb{C}_{C_a} \\ 0 & 0 & \mathbb{B}_{C_b} \end{pmatrix} \frac{d}{dt} \begin{pmatrix} T \\ C_a \\ C_b \end{pmatrix} + \begin{pmatrix} \mathbb{A}_T & 0 & 0 \\ 0 & \mathbb{A}_{C_a} & 0 \\ 0 & \mathbb{C}_{C_b} & \mathbb{A}_{C_b} \end{pmatrix} \begin{pmatrix} T \\ C_a \\ C_b \end{pmatrix} = \begin{pmatrix} R_T \\ R_{C_a} \\ R_{C_b} \end{pmatrix}, \quad (8)$$

where

$$\begin{aligned} [\mathbb{A}_T]_{i,j} &= \lambda(\nabla w_i, \nabla w_j) \quad , & [\mathbb{A}_{C_a}]_{i,j} &= \frac{D_a \epsilon}{\tau} (\nabla w_i, \nabla w_j) \quad , \\ [\mathbb{A}_{C_b}]_{i,j} &= \gamma \varphi(w_i, w_j) \quad , & [\mathbb{B}_T]_{i,j} &= c_v(w_i, w_j) \quad , \\ [\mathbb{B}_{C_a}]_{i,j} &= \epsilon(w_i, w_j) \quad , & [\mathbb{B}_{C_b}]_{i,j} &= \frac{1}{c} (w_i, w_j) \quad , \\ [\mathbb{C}_T]_{i,j} &= \chi(w_i, w_j) \quad , & [\mathbb{C}_{C_a}]_{i,j} &= (1 - \epsilon)(w_i, w_j) \quad , \\ [\mathbb{C}_{C_b}]_{i,j} &= -\frac{\gamma}{C_a^{100}} (w_i, w_j) \quad , & & \end{aligned}$$

$$\begin{aligned} [R_T]_i &= \lambda \langle \alpha, w_i \rangle, \\ [R_{C_a}]_i &= \frac{D_a \epsilon}{\tau} \langle \beta, w_i \rangle, \\ [R_{C_b}]_i &= 0, \end{aligned}$$

$$\begin{aligned} [T]_i &= T^i(t), \\ [C_a]_i &= C_a^i(t), \\ [C_b]_i &= C_b^i(t). \end{aligned}$$

Values of functions $c_v, \chi, \lambda, D_a, C_a^{100}, \gamma$ and φ in given time are chosen to be piecewise constant on each element (in the manner described further). Values of functions ϵ and τ are also chosen to be piecewise constant on each element, but as material characteristic, independent on time.

3 Numerical model

System (8) with initial conditions (5) can be solved e.g. by the Euler method. Its advantage is, that it can be used for case, when the system has coefficients depending on unknown quantities ($c_v, \chi, \lambda, D_a, C_a^{100}, \gamma$ and φ).

3.1 Time discretization

The implicit scheme for approximation of time derivatives is used,

$$\left. \frac{\partial f}{\partial t} \right|_{t=n} \equiv \frac{f^{n+1} - f^n}{\Delta t}.$$

This scheme provides sufficient numerical stability. Then the system (8) can be rewritten more simply,

$$\mathbb{D}\dot{X} + \tilde{\mathbb{D}}X = R,$$

where

$$\mathbb{D} = \begin{pmatrix} \mathbb{B}_T & 0 & \mathbb{C}_T \\ 0 & \mathbb{B}_{C_a} & \mathbb{C}_{C_a} \\ 0 & 0 & \mathbb{B}_{C_b} \end{pmatrix},$$

$$\tilde{\mathbb{D}} = \begin{pmatrix} \mathbb{A}_T & 0 & 0 \\ 0 & \mathbb{A}_{C_a} & 0 \\ 0 & \mathbb{C}_{C_b} & \mathbb{A}_{C_b} \end{pmatrix},$$

$$\begin{pmatrix} \mathbb{B}_T + \Delta t \mathbb{A}_T^{(n)} & 0 & \mathbb{C}_T \\ 0 & \mathbb{B}_{C_a} + \Delta t \mathbb{A}_{C_a}^{(n)} & \mathbb{C}_{C_a} \\ 0 & \Delta t \mathbb{C}_{C_b}^{(n)} & \mathbb{B}_{C_b} + \Delta t \mathbb{A}_{C_b}^{(n)} \end{pmatrix} \begin{pmatrix} T^{(n+1)} \\ C_a^{(n+1)} \\ C_b^{(n+1)} \end{pmatrix} = \tilde{R}^{(n)}. \tag{9}$$

$$X = \begin{pmatrix} T \\ C_a \\ C_b \end{pmatrix},$$

$$R = \begin{pmatrix} R_T \\ R_{C_a} \\ 0 \end{pmatrix}.$$

Values D_a, K, γ in n^{th} time step were implicitly chosen by substituting the guess $\hat{X}^{(n+1)}$ in time step $(n + 1)$. Matrix \mathbb{D} and right hand side R are time-dependent, more accurately, they depend on values $c_v, \chi, \lambda, D_a, C_a^{100}, \gamma$ and φ which consist in X . For n^{th} time step, they are having form

$$\tilde{\mathbb{D}}^{(n)} = \tilde{\mathbb{D}}(\hat{X}^{(n+1)}),$$

$$R^{(n)} = R(\hat{X}^{(n+1)}).$$

Consequently, the problem

$$\mathbb{D} \frac{X^{(n+1)} - X^{(n)}}{\Delta t} + \tilde{\mathbb{D}}^{(n)} X^{(n+1)} = R^{(n)},$$

was solved and the variation between the solution $X^{(n+1)}$ and the guess $\hat{X}^{(n+1)}$ was watched. For the large variation, the solution $X^{(n+1)}$ was used as new estimate $\hat{X}^{(n+1)}$ (for first iteration we used $\hat{X}^{(n+1)} \equiv X^{(n)}$). This process was repeated several times until only small variation are reached. Then the new initial problem for time step $(n+2)$ was solved. In particular iteration in n^{th} time step the linear system is solved,

$$(\mathbb{D} + \Delta t \tilde{\mathbb{D}}^{(n)}) X^{(n+1)} = R^{(n)} \Delta t + \mathbb{D} X^{(n)}.$$

If denoted

$$R^{(n)} \Delta t + \mathbb{D} X^{(n)} = \tilde{R}^{(n)},$$

the linear system can be written in block structure,

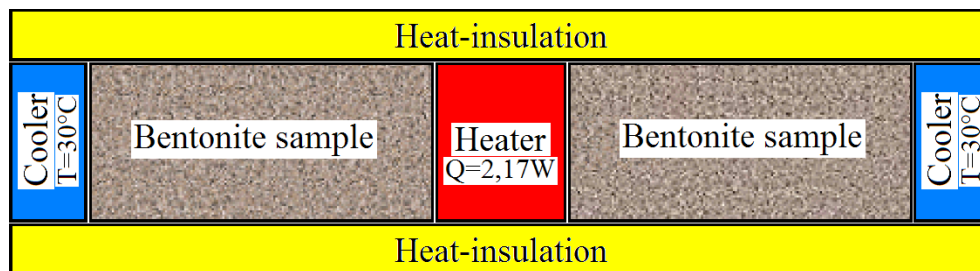


Figure 2: Design of experimental device

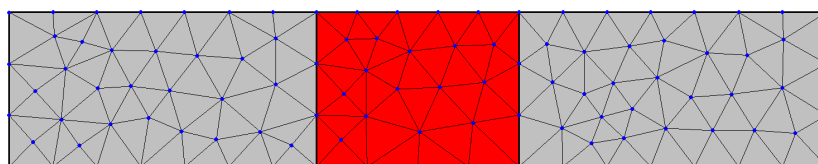


Figure 3: Model mesh of experimental device

4 Experiment BenchMark 1.3

4.1 Design of experimental device

Experimental device and results obtained by this device were published in [5]. Scheme of experimental device is shown on figure 2.

The two cylindrical specimens (38 mm diameter and 76 mm height) are placed in the device, grey color on the scheme.

The heater is located between them, red color on the scheme. The heater is a copper cylinder (38 mm diameter and 50 mm height) with electrical resistor inside. The resistor is connected to an adjustable source of direct current, so that output heating power is kept on level $Q_{heater} = 2,17$ W.

The temperature of other bases of specimens are controlled by stainless steel collars on value $T_{cooler} = 30$ °C, blue color on the scheme.

The ensemble is contained in a perspex tube. This tube minimalize water vapor losses and heat losses.

The initial temperature of all parts of experimental device is $T_{initial} = 22$ °C. The initial saturation of bentonite specimens responds to ambient humidity $RH_{initial} = 50\%$.

In time $t = 0$ s the heater and the coolers were switched on. Experiment was performed till the moment, when the temperatures in each measured points were stabilized. Final time was 175 hours.

4.2 Measured values

The evolution of temperature was measured in points which lays on the center line of cylindrical specimens, in distances ($x = 0, 20, 38, 60, 76$ mm) from heater. The temperature was measured continuously.

The moisture was measured on the end of experiment. The apparatus was dismantled without delay. Each specimen was cut into six small cylinders and the water content of each cylinder was determined.

4.3 Model of experimental device

Scheme of model of experimental device is shown on figure 3. Mesh has 107 nodes and 169 triangles. Third virtual dimension of mesh, thickness of elements, is 29,85 mm. So, volumes and masses of specimens and heater are the same as volume and mass of parts in original apparatus.

Material parameters of bentonite are taken from [1], [2]. Values of parameters are:

- $c_v = \frac{4200}{1630}Cb + (1.38T + 732.5)$ [$\text{Jkg}^{-1}\text{K}^{-1}$]
- $\lambda = \left(\frac{0.57-1.28}{1+e^{\left(\frac{Cb}{350}-0.65\right)/0.1}} \right) + 1.28$ [$\text{Wm}^{-1}\text{s}^{-1}$]
- $\chi = 0$ [Jkg^{-1}]

- $D_a = 1.4e - 5 \text{ [m}^2\text{s}^{-1}\text{]}$
- $\gamma = 2.5e^{-3} \text{ [1]}$
- $\varphi = \frac{C_b}{350} \text{ [1]}$
- $\tau = 6 \text{ [1]}$
- $\epsilon = 0.45 \text{ [1]}$
- $\rho = 1630 \text{ [kgm}^{-3}\text{]}$

Material parameters of copper are taken as standard. Values of parameters are:

- $c_v = 392.9 \text{ [Jkg}^{-1}\text{K}^{-1}\text{]}$
- $\lambda = 385 \text{ [Wm}^{-1}\text{s}^{-1}\text{]}$
- $\chi = 0 \text{ [Jkg}^{-1}\text{]}$
- $D_a = 0.0 \text{ [m}^2\text{s}^{-1}\text{]}$
- $\gamma = 0.0 \text{ [1]}$
- $\varphi = 0.0 \text{ [1]}$
- $\tau = 1 \text{ [1]}$
- $\epsilon = 0.0 \text{ [1]}$
- $\rho = 8930 \text{ [kgm}^{-3}\text{]}$

The initial conditions (temperature, saturation) are applied by equation (5).

In time $t = 0$ s, the Neumann boundary condition (3) is applied on part of mesh, which represented heater, red part on figure 3. Cooler and heat losses through heat-insulation, yellow part on figure 3, are represented by the Newton boundary condition (4). Parameter σ_T is chosen as the best guess and was improved by iterations.

All boundaries are closed for water vapors, the Neumann boundary condition (3) $q = 0$ is applied.

The model solve this task for time interval 175 hours.

The temperatures in measured points are shown in graph on figure 4. For the better illustration, the graph is reduced only to 100 hours.

The water content of specimens, on the end of scenario ($t = 175$ hours), is shown in graph on figure 5.

5 Comparison of results and conclusion

The graphic comparison of experimental data, collected during experiment *Benchmark 1.3*, and data obtained by model, see figure 4 and 5, shown that the model, described in this article, solve the task of coupled heat and moisture transport with diffusion on porous media with good accuracy.

On the other hand it has to be said that this model itself is not able to fully describe the processes in a "real life" bentonite buffer. What is missing is a model, which will have to be done in the future, which will deal with liquid underground water. This water which is obviously present everywhere in the natural conditions and has to be taken in consideration in future research.

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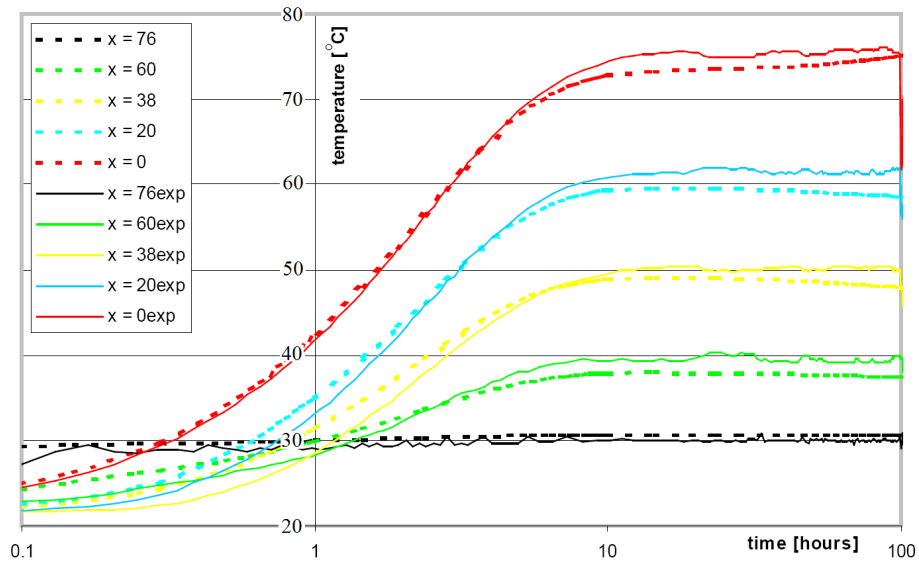


Figure 4: Evolution of temperature

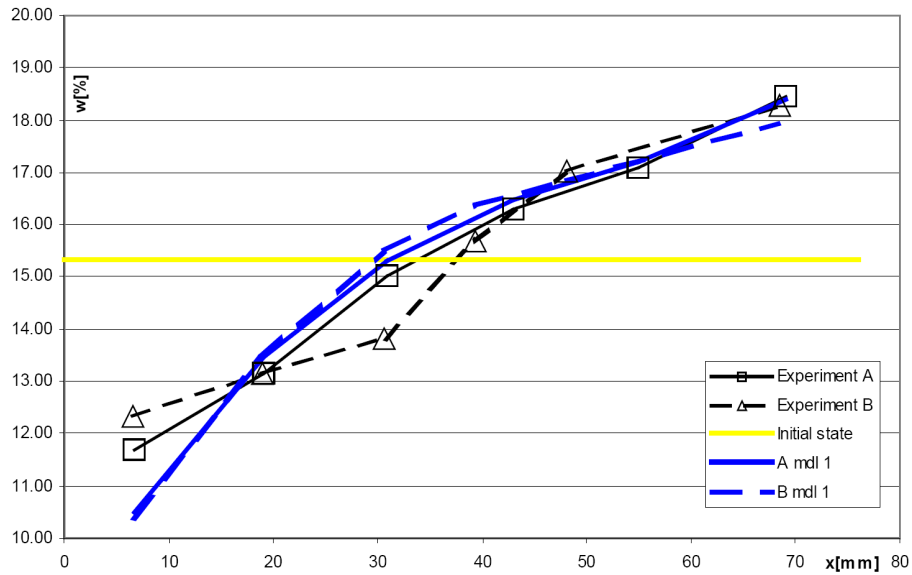


Figure 5: Water content profiles on the end ($t_{end} = 175$ hours)