MODELLING THE IMPACT OF BACK-FILLING MATERIAL ON THE WATER QUALITY OF FLOODED IRON MINES (LORRAINE, FRANCE)

HAMM Virginie[1], FABRIOL Robert[1], VAUTE Laurent[2], FOURNIGUET Guy[2], BAUBRON Jean-Claude[2]

[1] BRGM GISOS, Rue du Doyen Marcel Roubault – BP 40 – 54501 Vandoeuvre-lès-Nancy Cedex - France.
v.hamm@brgm.fr; r.fabriol@brgm.fr

[2] BRGM SGR/LOR, 1 av. du parc de Brabois - 54500 Vandoeuvre-lès-Nancy Cedex - France.
g.fourniguet@brgm.fr; l.vaute@brgm.fr; jc.baubron@cegetel.net

ABSTRACT: Residual voids located in the collapsed zones of mine workings present a risk of subsidence for overlying buildings. In the Lorraine iron basin (France), filling of the voids above the "Cité du Stock" with a slurry made of cement and fly ash has been envisaged. The impact of the back-filling material on the underground water quality of the flooded mine used for drinking water has been decided. Laboratory leaching tests were carried out and a model of the mining reservoir was built with a PNS ("Pipe Network System"). The results of leaching tests in columns allowed us to identify the mixture of cement and fly ash with the lowest contaminant capacity. Flow and transport modelling in galleries has shown that the concentrations of chemical elements at the supply well are within the limit concentrations for drinking water.

KEYWORDS: Flooded mine, back-filling material, leaching test, pipe network system, transport of chemical elements

RESUME : Les vides résiduels dans les zones foudroyées des travaux miniers présentent un risque d'affaissement pour les habitations sus-jacentes. Dans le bassin ferrifère lorrain (France), un comblement des vides par un coulis de ciment avec des cendres volantes a été envisagé sous la "Cité du Stock". L'impact du remblayage sur la qualité de l'eau du réservoir minier exploitée en aval pour l'A.E.P a été évalué. Des tests de lixiviation en laboratoire sur différents types de coulis ont été réalisés et un modèle du réservoir minier ennoyé a été construit sous forme d'un RDT (« Réseau De Tubes »). Les résultats des tests de lixiviation en colonne ont permis d'identifier le mélange ciment et cendres volantes le moins polluant et la modélisation des écoulements et du transport dans le réseau de galeries a montré que les concentrations en éléments chimiques à l'A.E.P sont très inférieures aux concentrations limites pour l'eau potable.

MOTS-CLEFS : Mine ennoyée, matériau de remblayage, test de lixiviation, réseau de tubes, transport d'éléments chimiques

1. Introduction

Closure of iron mines in the Lorraine basin (France) has left a dense network of underground galleries. Residual voids located in the collapsed zones present a risk of subsidence for overlying buildings. In June 2002, open mining voids were identified under the "Cité du Stock" (Thil, France). This zone was assumed to have collapsed but wide open spaces were detected (10,000 m^3) 60 m below the surface. A preliminary study was carried out to determine the possible impact of back-filling material on the underground water quality of the Godbrange flooded mine located downstream of the zone (Baubron et al. 2003). Water from the reservoir is used for drinking water supply from the Hussigny-Godbrange pumping well.

This paper presents the main results of the study: the chemical model resulting from the leaching tests on 6 mixtures of cement and fly ash, and the construction of a pipe network system for representing hydraulic and transport behaviour in flooded galleries and its application to the Godbrange mining reservoir.

2. Chemical evolution of water in contact with the back-filling material: experimental approach

2.1. The back-filling material tested

Several cement - fly ash slurries were tested in order to obtain an optimal back-filling material. The material must present a mechanical resistivity greater than 2.5 MPa, a low chemical dissolution rate in contact with water and a low cement content to reduce the cost. Two types of cement were used; CEM I and CEM III. The fly ashes were sampled from the storage of an electrical power plant (EDF, La Maxe). The slurry compositions tested are presented in table 1. Once the cement, fly ash and water had been mixed in a stirred tank, the resulting slurry was used to prepare cylindrical samples. After 90 days setting, the samples were crushed into particles to fill the leaching columns.

Table1. Composition of six cement – fly ash slurries tested

Reference	Cement (weight %)	Fly ash (weight %)	Slurry (weight in g)	Dogger water (weight in g)
CEMI 3-7	30% CEM I 52.5	70% FA	10608	7497
CEMI 2-8	20% CEM I 52.5	80% FA	9888	7772
CEMI 1-9	10% CEM I 52.5	90% FA	10164	7572
CEMIII 3-7	30% CEM III C 32.5	70% FA	9616	7617
CEMIII 2-8	20% CEM III C 32.5	80% FA	10116	7518
CEMIII 1-9	10% CEM III C 32.5	90% FA	9895	7797

2.2. Column leaching tests

Each of the six columns of the leaching system was filled with 10 kg of ground material and 7.5 kg of Dogger water (table 1). The particle size distribution was identical in each column (35 mm< S <40 mm). In a first step, the system remained closed for 14 days to approach water-material chemical equilibrium. In a second step, the system was opened to observe any chemical changes in the leaching conditions. Each column was supplied at its base with a flow of 300 ml/day of Dogger water. The water overflowed through an opening at the top of the column, where it was sampled for chemical analyses. The water conductivity and temperature in the column were measured continuously (figure 1).

Continuous monitoring of conductivity is a reliable indicator of the bulk composition of the water. The mineralisation observed after 14 days decreased gradually until 125 days of leaching, when similar chemical concentrations were obtained at the inlet (Dogger water) and outlet of the CEM III columns.

The slurries made of CEM I contaminated water more than the slurries made of CEM III. The concentration of chemical elements in the water results from the dissolution of fly ash as well as from cement. At this stage of the work, we are not able to identify the source of each contaminant in the slurry. The results of the 6 leaching tests indicate the CEM III 1-9 (10% CEM III and 90% of fly ash) is the most appropriate among the slurries tested. The chemical concentrations in water are lower than the limits defined by the French administration for drinking water supply ("*Livre III du Code de la Santé Publique*"), except for sulphate and boron at the beginning of the leaching test, and for aluminium and selenium during the leaching test period. The CEM III 1-9 is considered the more favourable among the six back-filling materials tested, and used hereafter for the chemical model.

Figure 1. Experimental device with the six leaching columns

2.3. Chemical interaction model

The column with an ascending flow may be represented as a perfectly stirred reactor described by the following mass-balance equation (Villermaux, 1993):

$$QsCs = QeCe + V(t)\ r(t) \ - \ \frac{d(V(t)C(t))}{dt} \qquad \text{(a)}$$

where:
Qs = outflow rate (l/s)
Qe = inflow rate (l/s)
Cs = outflow concentration (mol/l)
Ce = inflow concentration (mol/l)
C(t) = concentration in the reactor (mol/l)
V(t) = volume of water in the reactor (l) at time t
r(t) = production or consumption rate of a given chemical element or compound (mol/l/s)

Consider the dissolution-precipitation reaction of a mineral A_nB represented by the following equation:

$$A_nB \leftrightarrow nA^+ + B^{n-} \qquad \text{(b)}$$

The rate of this reaction is the difference between the dissolution rate and the precipitation rate:

$$r(t) = [k_{diss}\ (A_nB)] - [k_{pcp}\ (A^+)^n\ (B^{n-})] \qquad \text{(c)}$$

where:
k_{diss} = dissolution constant
k_{pcp} = precipitation constant
(A_nB) = mineral activity (1 for a pure solid)
$(A^+)^n\ (B^{n-})$ = dissolved species activity

Taking into account the relationship between the equilibrium constant of the reaction K and the kinetic constants, $K = k_{diss}/k_{pcp}$, the total reaction rate may be expressed as:

$$r(t) = k_{pcp}\ [K - (A^+)^n\ (B^{n-})] \qquad \text{(d)}$$

The conductivity of water C_w (µS/cm) is an overall function of the activity of ions. On the basis of equation (d), we assume a global reaction rate $r_g(t)$ function of C_w and empirically expressed as:

$$r_g(t) = [k_g\, M(t)^n]\, [K_g - C_w\,(t)] \qquad \text{(e)}$$

where:

k_g = overall kinetic precipitation constant

K_g = overall equilibrium constant

M(t) = total mass of chemical elements in the solid, expressed in µS/cm

n = overall order of reaction

The constants for the CEM III 1-9 kinetic model are obtained by fitting with the data from leaching column experiments.

Table 2. Constants for the CEM III 1-9 slurry

	CEM III 1-9
log K_g	+ 0.4
log k_g	- 11.75
M (µS/cm)	56.25
n	8.25

2.4. Chemical simulator

The numerical simulator was built using Matlab® software and its graphic interface, Simulink®. This interface enables the modelling and simulation of dynamic systems using a graphic representation of the block-diagram type based on bond-graphs formalism (Maschke 1992; Auroy 2000, Bequette 1998; Kervévan et al. 1998; Collon 2003, Collon et al. 2004).

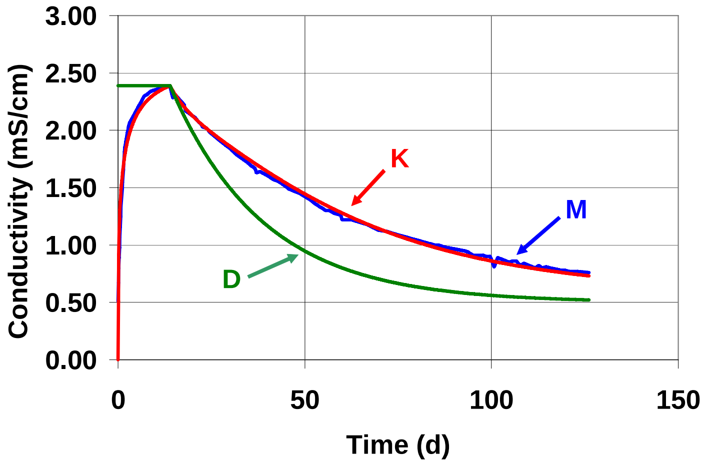

Figure 2. Calculated and measured water conductivity obtained with the CEM III 1-9 slurry. (M: measurements; D: perfectly stirred tank; K: perfectly stirred tank with kinetic)

The simulated conductivities (figure 2) are in good agreement with those measured. The kinetic effect is highlighted by comparing it with the results obtained with a perfectly mixed tank without any kinetic effect. The difference is due to the progressive dissolution of contaminant from the slurry. When comparing the change over time of contaminant concentration and water conductivity, both slopes appear similar. Consequently, the concentration is empirically calculated from the conductivity of water. This relation is used to simulate the evolution of the contaminant concentration in water. For each chemical element a proportionality coefficient is defined. Figure 3 represents the calculated and analysed selenium concentration in water. The chemical simulator built is used as a point source of contaminant in the transport model described hereafter.

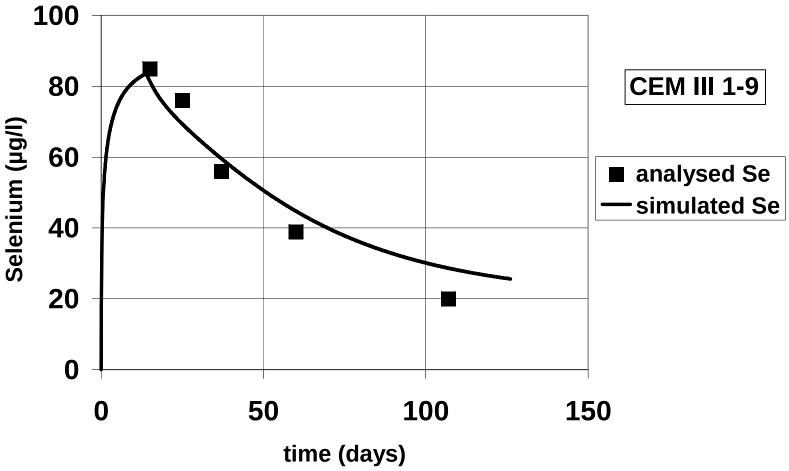

Figure 3. Calculated and analysed selenium concentration in water at the leaching column outlet

3. A Pipe Network System approach for modelling the flow and transport of chemical elements in the flooded iron mines

3.1. Pipe Network System principles: hydraulic behaviour and transport of chemical substances

The mine galleries can be modelled as connected pipes forming a pipe network system where pipes are channels used to convey water and chemical substances from one location to another. The physical characteristics of such a network include the length of the pipes, their inside diameter and their roughness coefficient.

When water is conveyed through a pipe, hydraulic energy is lost due to the friction between the moving water and the stationary pipe surface. This friction loss is a major energy loss in pipe flow and is a function of the flow rate, pipe length, hydraulic diameter, roughness coefficient and fluid properties (Sellens 1996).

The head loss associated with flow through a pipe can be expressed in a general fashion as:

$$h_L = a.q^b \qquad \text{(f)}$$

where:
h_L = head loss, m
q = flow rate, m^3/s
a = resistance coefficient
b = flow exponent

The values of the resistance coefficient and flow exponent depend on which head loss law is applied. A widely used law is the Darcy-Weisbach formula where the resistance coefficient “a” depends on a friction factor, the pipe length, the hydraulic section and diameter and the flow exponent “b” equals 2, as shown below:

$$\mathrm{h_L} = \lambda(\varepsilon, \mathrm{Re}).\frac{L}{D_h}.\frac{1}{2gS_h^{\;2}}.q^2 \qquad \text{(h)}$$

where:
λ = a friction factor that depends on the roughness coefficient ε and Reynolds number Re
L = pipe length, m
D_h = hydraulic diameter, m
S_h = hydraulic section, m
g = gravity, m/s^2
q = flow, m^3/s

The expression of the friction factor depends on the flow; indeed, for turbulent flow observed during flooding, the friction factor depends only on the relative roughness coefficient but for the laminar flow observed generally after flooding, the friction factor becomes inversely proportional to the Reynolds number (λ=64/Re) and the head loss in a pipe can be expressed as a linear function of the flow:

$$h_L = \frac{32\nu L}{gS_h D_h^{\;2}}.q \qquad \text{(i)}$$

where:
ν = kinematic viscosity, m^2/s

Equation (i) can also be expressed as:

$$q_{ij} = k_{ij}.(h_i - h_j) \qquad \text{(j)}$$

where:
k_{ij} = conductance of a pipe [m^2/s]. This represents the flow per unit of pipe length and depends only on the geometric characteristics of the pipe and on the flow, kinematic viscosity and gravity,
q_{ij} = flow between nodes i and j, positive for flow entering the node j ($h_i > h_j$) and negative for flow leaving the node j ($h_i < h_j$).

A second set of equations that must be satisfied is the flow continuity at each pipe junction:

$$\sum_i q_{ij} + q_{imposé,j} = 0 \qquad \text{(k)}$$

where $q_{imposé,j}$ is a possible incoming or outcoming flow from external conditions.

The combination of equations (j) and (k) leads finally to:

$$h_j = \frac{\sum_i h_i k_{ij} + q_{imposé,j}}{\sum_i k_{ij}} \qquad \text{(l)}$$

The hydraulic head is calculated at each node of the pipe network system and its value determines the value and direction of flow in the network. In order to solve these two sets of equations, the user must define initial and boundary conditions on hydraulic head and flow.

The transport through the pipe network is considered non reactive. This means that a substance which travels through a pipe will not react with the surrounding rock material. A dissolved substance will travel down a pipe by advection and will mix completely and instantaneously at junctions and be scattered in the entire network.

Two sets of equations describe these mechanisms: the first is the advection through a pipe, which implies a delay in the concentration of the downstream node:

$$C_{downstream}(t+\Delta t) = C_{upstream}(t-(\tau-\Delta t)) \qquad \text{(m)}$$

where:

t = actual transport time, s

Δt = time step, s

τ = transport delay through the pipe, τ = v/q where v represents the volume of the pipe (m^3) and q the flow in the pipe (m^3/s)

This equation requires that the time step is shorter than the time delay to pass through a pipe.

The second set of equations describes mixing at pipe junctions; the concentration of a substance leaving the junction is simply the flow-weighted sum of the concentrations from the inflowing pipes:

$$C_{j/x=0} = \frac{\sum_i q_{ij} C_{i/x=L} + q_{j,ext} C_{j,ext}}{\sum_i q_{ij} + q_{j,ext}} \qquad \text{(n)}$$

where:

$C_{j/x=0}$ = concentration at node j, mg/L

$C_{i/x=L}$ = concentration at the end of the link of node i, mg/L

$C_{j,ext}$ = concentration of the external flow entering the network at node j, mg/L

q_{ij} = flow through the link ij, m^3/s

$q_{j,ext}$ = external source flow entering the network at node j, m^3/s

3.2. Building the Pipe Network System: description and advantages of this approach

The software used for building the pipe network was Matlab® and its graphical interface Simulink®. The principle was to build the flow and chemical sets of equations described above by using block features provided by Simulink®. A pipe network can be represented as the repetition of a basic pattern conceived of a junction and its two connected pipes. Figure 4 below shows how pipes and nodes are modelled with Simulink® block diagrams. The node or junction in a network is represented by two blocks, one for the hydraulic head equations and the other for the water quality equations. The conductance of a pipe and the flow rate are calculated at the link between two nodes. The entry parameters for each pipe or gallery are the length, the hydraulic section, the inside diameter and a roughness coefficient for non laminar flow. This approach does not require other parameters, in contrast with porous-medium models where the user must define the intersticial permeability and a storage factor to solve hydraulic equations and the porosity and a dispersivity coefficient for the transport of chemical substances (Lanini et al. 2000). These parameters can be estimated for a porous aquifer but not for wide mining spaces. The pipe network system approach is interesting for observing what occurs inside the galleries and at the pipe wall, without modelling the rock surrounding the galleries, which is not open to water except at the collapsed zones, which are considered as refill zones.

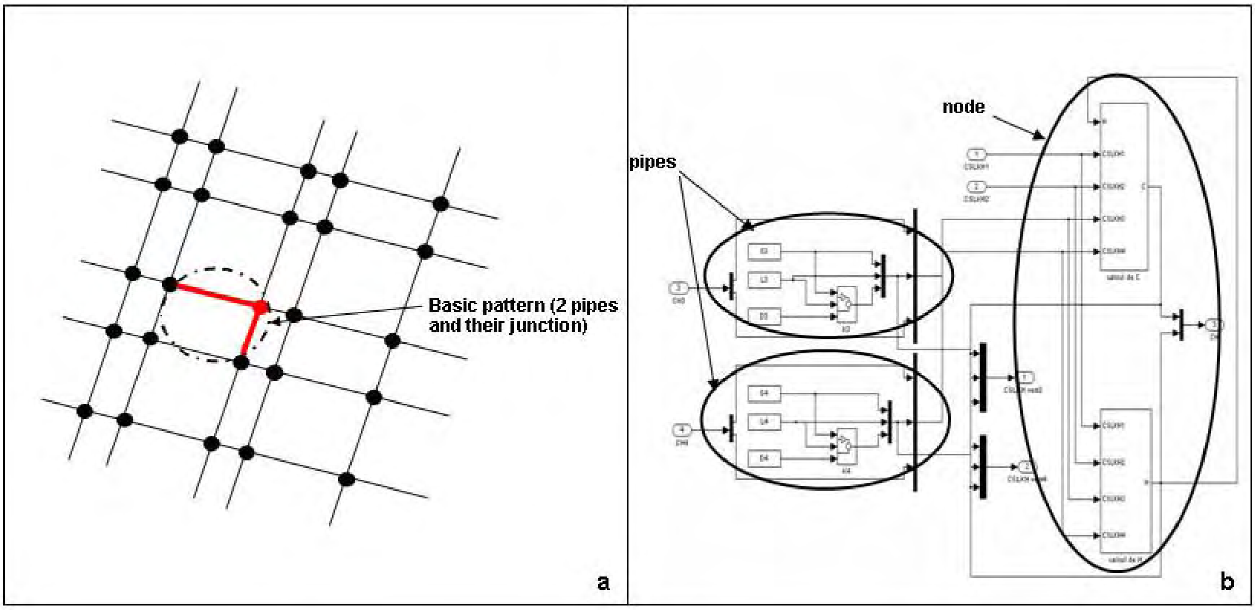

Figure 4 : Example of a basic pattern in a network (a) and its representation with Simulink® block features (b)

4. Modelling the impact of the “Cité du stock” cavity filling on the Godbrange reservoir

4.1. Godbrange mining basin: location and description

The Godbrange iron mine is located between the north part of the Lorraine iron basin and the Luxembourg border. It covers the mining concessions of Hussigny-Godbrange, Tiercelet, Brehain, Villerupt and Crusnes (figure 5). The iron-bearing formation is composed of alternating marly levels and ferruginous limestone. It is overlain by a micaceous marl layer and followed by a Dogger Bajocian limestone succession that constitutes a major karstic aquifer (Collon et al, 2002). Its floor area is estimated to be 13 km^2 and the mine water reserves are estimated at around 11,600,000 m^3. The reservoir has been flooded since 1987, due to the end of dewatering. The water level reaches the overflow point at 337 m in the north western end of the reservoir and flows into the Moulaine River. The water of the reservoir is used for drinking water at the Hussigny-Godbrange supplying well located north west of the basin.

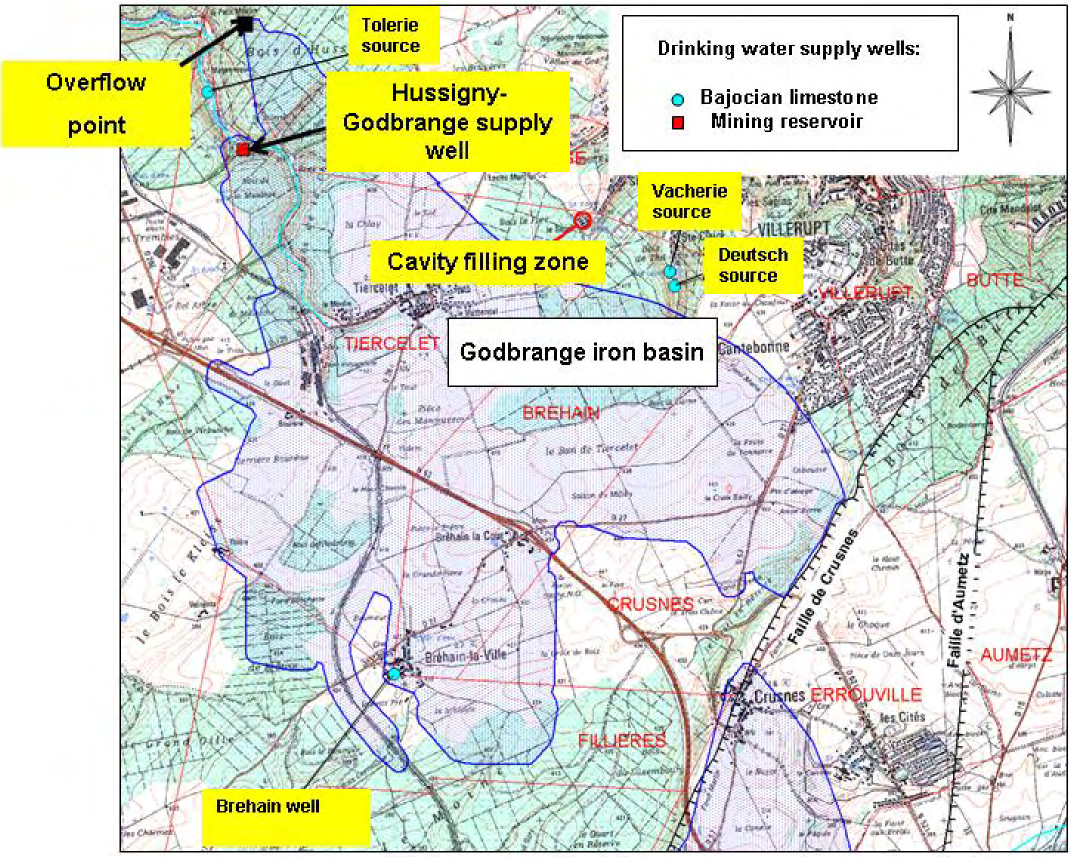

Figure 5 : Geographical location of the Godbrange iron basin

4.2. Building the Pipe Network System of the Godbrange mining reservoir

The pipe network system was built using digital planes of the Godbrange mine. The main galleries conveying water from the cavity zone to the drinking water supply well and to the overflow point were digitalized and reproduced in the conceptual network. The collapsed zones were modelled as external source flows coming from the above Dogger aquifer (refill zones). The boundary conditions of the Godbrange reservoir are represented by incoming flows due to refill zones, the values of which are proportional to the refill areas, an outcoming flow of 175 m^3/d at the pumping well and a fixed head of 337 m at the overflow point. Figure 6 below shows the conceptual model and an extract of the pipe network as it appears in the Simulink® model.

The chemical simulator described in paragraph 2.4 has been introduced in the pipe network system as a point but time varying chemical source entering in the network by the cavity filling zone.

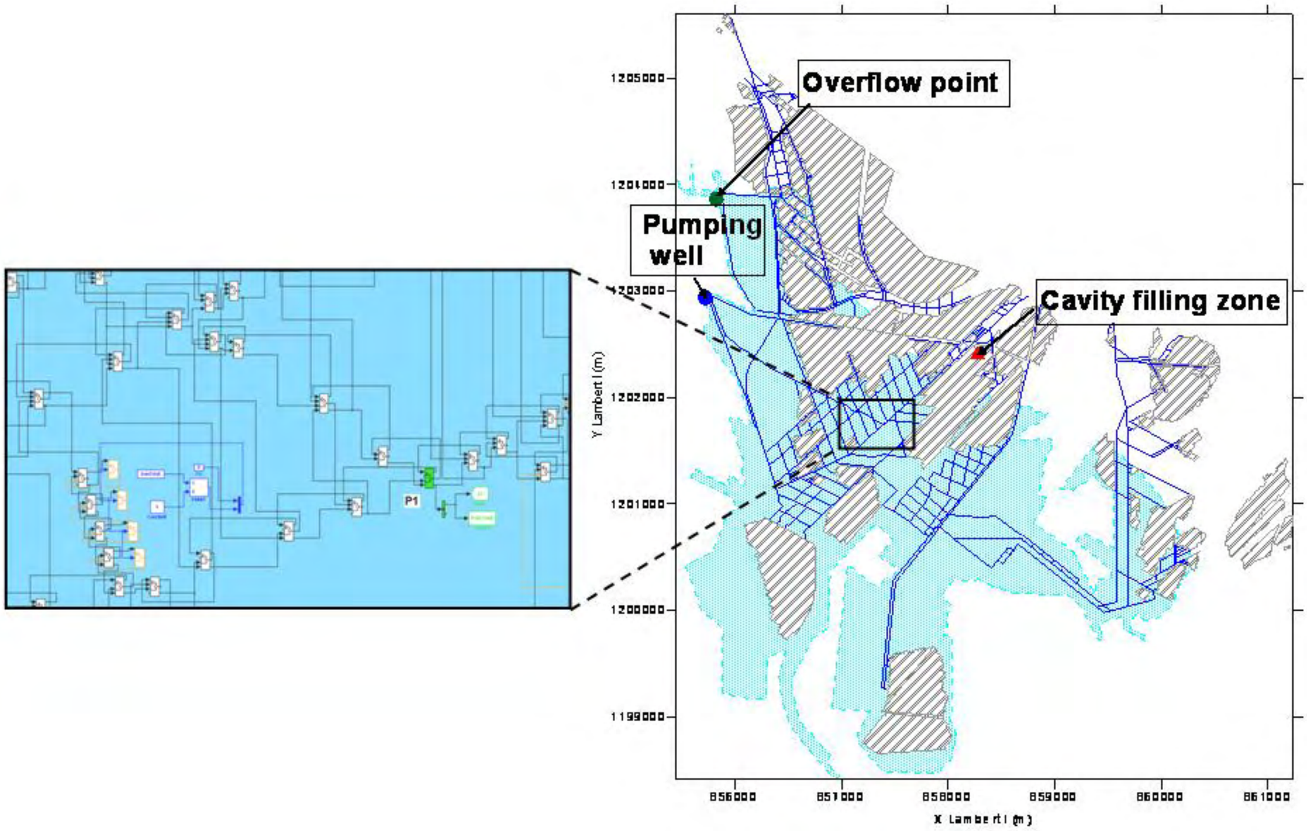

Figure 6 : The conceptual model of the Godbrange reservoir and an extract of the network in Simulink® (*dark blue: pipe network, light blue: flooded zone, striped grey: collapsed zones*)

4.3. Transport of contaminants from the cavity filling material

The simulation was performed for a total duration of 16 years after the filling of the cavity. As explained in paragraph 2.4, the proportionality between the water conductivity measurement and the contaminant concentration was used to determine the concentrations of all elements released from the filling material. Figure 7 below gives an example of the simulation for the element selenium. The concentration in the entire network was at a maximum after 1 year of simulation, and then decreased very slowly without reaching the initial state after 16 years of simulation. The limit value of selenium for drinking water was set at 10 µg/L. The simulated Se concentration exceeds this value the first year near the cavity filling, and then decreases below this limit concentration. The model shows that the pumping well for drinking water supply and the overflow point are not contaminated by any substance coming from the filling material because the contaminant travelling through the network is diluted by water refill zones with a high dilution factor.

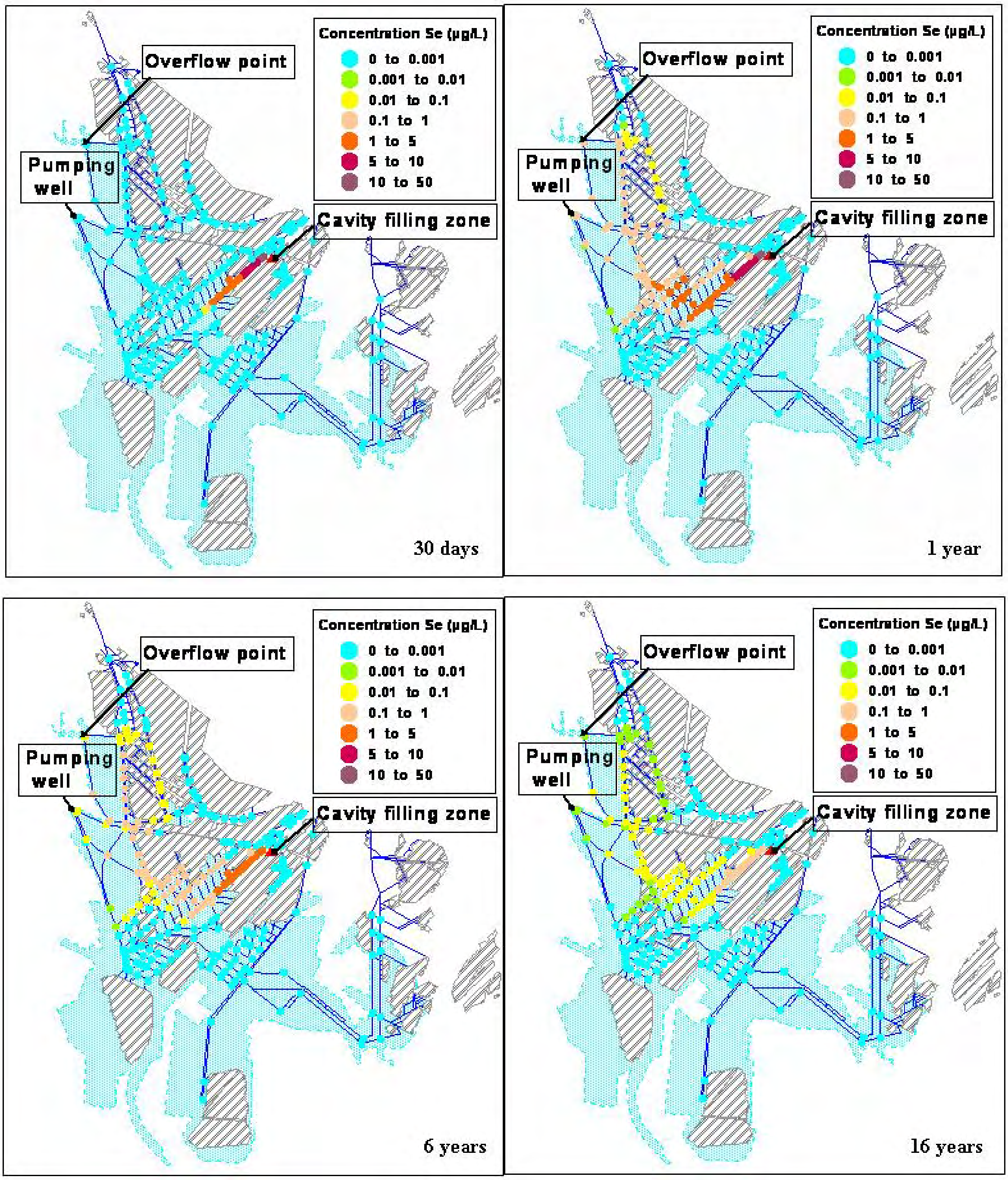

Figure 7 : Computed concentration of selenium (µg/L) in the pipe network for 30 days, 1 year, 6 years and 16 years of simulation after cavity filling.

5. Conclusions

Leaching tests performed on six different mixtures of cement and fly ash indicated that the concentration of substances dissolved from the mixture CEM III 1-9 (10% of cement and 90% of ash) were below the limit concentrations for drinking water, except for the elements boron, sulphate, aluminium and selenium. This mixture was used with its kinetic model as the cavity filling source in the pipe network system of the Godbrange mining reservoir. The results of the hydraulic and transport modelling in the flooded galleries show that the concentrations of chemical elements at the supply well located in the north west are one thousand to one million times lower than the limit concentrations for drinking water. Furthermore, substances coming from the cavity filling material are immediately diluted after entering the reservoir and the concentrations become rapidly in line with drinking water standards.

6. Acknowledgements

This work forms part of the research programme carried out by GISOS (Research Group for the Impact and Safety of Underground Works).

7. References

Auroy M., (2000) - *Élaboration des schémas de procédés industriels*. Techniques de l'Ingénieur, traité Génie des procédés, J 6 018, 17 p.

Bequette B. W. (1998) – *Process dynamics Modeling, Analysis and Simulation.* Prentice Hall PTR, 622 p.

Baubron J-C., Fabriol R., Fourniguet G., Hamm V., Vaute L. (2003) – *Modélisation hydrogéologique du réservoir minier de Godbrange et influence d'un comblement partiel à cendres volantes.* Rapport brgm/RP-52802-FR, 73 p., 22 fig., 13 tab., 6 ann.

Collon P., Fabriol R., Buès M., (2004) - *Ennoyage des mines de fer lorraines : impact sur la qualité de l'eau - Flooding of the iron mines of Lorraine: impact on the water quality,* Comptes Rendus des Géosciences 336, p. 889-899

Collon P., (2003) - *Evolution de la qualité de l'eau des mines abandonnées du bassin ferrifère lorrain. De l'expérimentation en laboratoire à la modélisation in situ*. Thèse INPL, Octobre 2003. 247 p.

Collon P., Fabriol R., Vaute L., Pinault J-L., Buès M., (2002) - *Modelling changes in sulphate concentrations in discharge water from the abandoned iron mine of Saizerais*. Proceedings SWEMP 2002.pp 917-924.

Kervévan C., Thiéry D., Baranger P., (1998) - *SCS: Specific Chemical Simulators dedicated to chemistry-transport coupled modelling Part lll - Coupling of SCS with the hydro-transport modelling software MARTHE.* Goldschmidt Conference 1998, Toulouse, Mineralogical magazine, Vol 62 A, p. 773-774

Lanini S., Fabriol R. (2000) - *Différentes approches de modélisation des écoulements et du transport dans un réseau de tubes – comparaison avec des résultats expérimentaux*. Rapport BRGM/RP-50154-FR, 48 p., 10 fig., 1 tab., 3 ann.

Maschke B. (1992) - *Approche systémique des systèmes continus. Graphes de liaison.* Journées EEA-Electrotechnique, 26 et 27 mars 1992, Laboratoire d'Automatisme Industriel, Conservatoire National des Arts et Métiers, 35 p.

Sellens R. (1996) - *Losses in Piping*. Course of Queen's University at Kingston, Mechanical Engineering (http://me.queensu.ca/courses/MECH451/losses.htm)

Villermaux J. (1993) – *Génie de la réaction chimique, conception et fonctionnement des réacteurs*. TEC & DOC – Lavoisier, 448 p.