

## **Modelling the Remediation of Contaminated Groundwater Using Zero-Valent Iron Barrier**

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### **ABSTRACT**

This paper presents results of modelling studies on remediation of groundwater contaminated with uranium using a zero-valent iron permeable reactive barrier (ZVI PRB) at the U.S. Oak Ridge Y-12 site that are used to establish modelling techniques that are of value to other sites such as in the UK. A systematic modelling methodology has been developed to study the problem by using a suite of modelling tools. Firstly a conceptual basis of the main chemical processes representing the remediation of uranium by the ZVI PRB is developed. Two main effects involving reduction and corrosion have been identified as being relevant for the remediation processes. These are then formulated and implemented using the reactive chemical model PHREEQC to provide underpinning chemical input parameters for subsequent reactive solute transport modelling using the TRAFFIC and PHAST codes.

Initial results shows that modelling can be a very cost-effective means to study the hydrogeological and geochemical processes involved and to aid understanding of the remediation concept. The modelling approaches presented and lessons learnt are thought to be relevant to other cases of contaminated land study and are likely to be of value to site management concepts which consider on-site disposal of contaminated soils and materials.

### **INTRODUCTION**

Modelling represents a very cost-effective and risk-beneficial solution for the increased understanding of issues and uncertainties associated with liabilities and site end-point decision making in general. Modelling may also be used to evaluate the need for a technology or part of a Best Practical Environmental Option (BPEO). Modelling provides important tools for the assessment of environmental risks and supports the evaluation of performance issues and uncertainties, such as those relating to the likely success of remediation strategies, and other issues underpinning end point decisions. The costs involved in modelling can often be offset against the costs of large-scale pilot plants. Additional to the cost benefits, there are conventional safety benefits, reduced worker doses, energy, material and waste savings, and no decommissioning requirements. Therefore the application of modelling techniques, and the development of underpinning methodologies, is highly cost-effective and risk-beneficial.

This paper presents results of modelling studies on remediation of groundwater contaminated with uranium using a zero-valent iron permeable reactive barrier (ZVI PRB) at the U.S. Oak Ridge Y-12 site. The modelling work covers the deployment of a suite of models including underpinning geochemical models, groundwater flow and coupled reactive transport models and an assessment level model, using different time and spatial scales. The coupled reactive transport models, using the underpinning codes TRAFFIC (TRansport And Fluid Flow Including Chemistry) and PHAST (PHREEQC And HST3D), consider a local scale (approximately 100 m), whilst the assessment level model using the assessment level code SimER (Simulation of Environmental Risks) considers a larger kilometre scale area (approximately 5 km). The predictions from the models are compared against field measurements to establish the validity and confidence of the modelling approaches.

The primary focus of the modelling work concerns the establishment of approaches in formulating the conceptual model through to detailed modelling of site hydrology, reactive contamination transport and remediation of an evolving contaminant plume by a permeable reactive barrier. In particular, remediation of

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uranium in groundwater at the Y-12 site by a PRB has been studied at different levels of detail using a number of geochemistry and reactive transport codes.

## **CODE DESCRIPTIONS**

A number of codes have been used in the present modelling exercise, including an underpinning geochemistry code PHREEQC; underpinning reactive transport codes TRAFFIC and PHAST; and a new Performance Assessment (PA) code SimER.

### **PHREEQC**

PHREEQC (1) performs both speciation-solubility calculations and reaction path calculations. PHREEQC calculates concentrations of elements, molalities and activities of aqueous species, pH, pe, saturation indices, and mole transfers of phases to achieve equilibrium as a function of specified reversible and irreversible geochemical reactions.

This computer program is based on equilibrium chemistry of aqueous solutions interacting with minerals, gases, solid solutions, exchangers, and sorption surfaces, but also includes the capability to model kinetic reactions with rate equations that are completely user-specified in the form of BASIC statements. The modelling capabilities of PHREEQC v2 include:

- Aqueous, mineral, gas, surface, ion-exchange, and solid-solution equilibria
- Kinetic reactions (which can represent processes of microbial activity, metal corrosion, mineral reaction and radioactive decay).
- 1D diffusion or advection and dispersion with dual-porosity medium
- A powerful inverse modelling capability allows identification of reactions that account for the chemical evolution in observed water compositions
- Extensive geochemical databases

### **TRAFFIC**

TRAFFIC (2, 3) is a three-dimensional finite element (FE) reactive multicomponent transport model under variably saturated media. It has been developed by the Research and Technology area of BNFL, now Nexia Solutions. The code couples the transport simulator and the geochemical modelling code PHREEQC based on the sequential operator splitting solution strategy. Through the flexible, generic nature of PHREEQC, TRAFFIC can handle a broad range of geochemical reactive transport processes, including aqueous surface complexation, mineral precipitation / dissolution and ion exchange. TRAFFIC includes the following features:

- Coupled groundwater flow, transport of multiple chemical species and heat transport;
- Flow and transport using FE;
- Eulerian-Lagrangian module for transport;
- Equilibrium sorption or kinetic, reversible and irreversible sorption;
- Radioactive decay of multiple chains of radionuclides;
- Equilibrium chemical speciation (PHREEQC);
- Comprehensive electrokinetic transport;
- A colloidal transport module.

## **PHAST**

PHAST (4) is a program for simulating groundwater flow, solute transport, and multicomponent geochemical reactions in three-dimensional saturated conditions. It is a versatile ground-water flow and solute-transport simulator with capabilities to model a wide range of equilibrium and kinetic geochemical reactions. The flow and transport calculations are based on a modified version of HST3D (5, 6) that is restricted to constant fluid density and constant temperature. The geochemical reactions are simulated with the geochemical model PHREEQC v2.11 (1) which is embedded in the PHAST code.

Chemical reactions in PHAST include: (i) homogeneous equilibria using an ion-association thermodynamic model; (ii) heterogeneous equilibria between the aqueous solution and minerals, gases, surface complexation sites, ion exchange sites, and solid solutions; and (iii) kinetic reactions with rates that are a function of solution composition. The aqueous model (elements, chemical reactions, and equilibrium constants), minerals, gases, exchangers, surfaces, and rate expressions may be defined or modified by the user. PHAST has an advantage over the TRAFFIC code in that the full kinetic functionality of PHREEQC is enabled allowing the modelling of microbial and corrosion processes.

The PHAST code can be run on most computer systems, including computers with Windows, Linux and Unix operating systems. The numerical simulation of groundwater flow and solute transport with geochemical reactions often requires the use of large amounts of memory for execution and long processing times. The chemical calculations are the most computationally intensive part of the reactive transport simulations with PHAST. However, operator splitting between the chemical reaction step and the flow and transport steps leads to a natural parallelisation of the calculations. To reduce the long processing times of PHAST calculations, a parallel version of PHAST has been developed by using the Message Passing Interface (MPI) that runs on a multiprocessor computer or on a collection of computers that are networked. In addition, the PC installation includes a 3D graphical viewer which can provide very effective illustration of model configuration and results.

## **SIMER**

SimER (7) is a Performance Assessment code developed by BNFL (now Nexia Solutions) for application both to contaminated land issues and the disposal of low-level waste (LLW). It enables the source-pathway-receptor analysis for contaminants of interest to be assessed in detail. SimER's main capabilities can be summarised as follows:

- The whole system can be modelled in a single code. This avoids the need for the output from one code to be used as the input to another. This greatly speeds up the assessment process and has major quality assurance advantages.
- Ability to represent climate changes, evolving landscape and engineered features.
- SimER has a multiple run capability. This enables both sensitivity studies and probabilistic assessments to be undertaken.
- Both radioactive and non-radioactive contaminants can be modelled.
- The code includes a groundwater flow model, enabling groundwater flows to be calculated within the code rather than having to be imposed (as is generally required in performance assessment (PA) codes).
- The code includes a model for the chemical evolution of the system. This enables the variation of contaminant transport parameters with time to be calculated.
- All model parameters have associated units, and SimER checks that all expression and algorithms that are used have consistent units. This checking provides a major contribution to the quality assurance of SimER calculations.
- The code uses file-based input and output and enables libraries of input parameters to be included in code input.

## Y-12 SITE DESCRIPTIONS

The Y-12 Pathway-2 PRB field trial experiment, conducted at the US Oak Ridge Reservation (ORR), concerning the remediation of a uranium contaminant plume within the ORR was selected for the modelling study. Pathway 2 is a shallow pathway, carrying contaminants west, to the upper reaches of Bear Creek. The area around the PRB is designated as Area 2 and contains many boreholes for the monitoring of groundwater. This pathway is contaminated primarily with low levels of uranium and nitrate. Extensive field and geochemical data, with comprehensive quality assurance control, were available for this site in the public domain (8). The field trial experiment covered many aspects of a typical radioactive contaminant plume remediation technology. The contaminants and environmental conditions are considered to be of sufficient comparability to UK nuclear sites to contribute to the development of high confidence tools for assessing potential liabilities and risks, and on the selection of remedial techniques.

## MECHANISTIC LEVEL MODELLING OF THE Y-12 SITE

Detailed mechanistic modelling using 3D groundwater flow and biogeochemical reactive transport models have examined in detail the processes of uranium removal from groundwater in the vicinity of the zero-valent iron barrier and gravel trench PRB at the Y-12 site.

### Geochemical modelling using PHREEQC

Corrosion is the main process that forms the conceptual geochemical basis to be exploited in ZVI PRBs. Two effects are apparent, which may immobilise contaminants such as uranium and technetium.

1. Mediation of reducing conditions in the environment of the PRB through the utilisation of electron acceptors in the groundwater and adjacent sediments and soils. Once the system becomes sufficiently anaerobic through the reduction of dissolved oxygen and nitrate oxidised contaminant species such as the radionuclides U(VI) and Tc(VII) will be reduced to their less soluble forms (U(IV), Tc(IV)). Microbial processes may in part mediate the intrinsically slow process of corrosion and the coupled reduction reactions.
2. Iron corrosion products such as Fe(III) hydroxide are good sorbers of radionuclides, in particular anion species will tend to sorb under the alkaline conditions that result through anaerobic corrosion. Iron corrosion products will tend to be present in a zoned distributed, where more oxidised products are formed away from the metallic ZVI.

In the case of the behaviour of uranium at the Y-12 site the reductive process is considered to be the dominant process (9). In addition the reduction of  $\text{NO}_3^-$ , which is a feature of the plume contamination may also be affected by the ZVI.

### Representation of corrosion processes with PHREEQC

The kinetic processes of corrosion that limit the rate at which metallic iron is reacted with groundwater and associated minerals and corrosion products can in principle be represented using the functionality of PHREEQC version 2 (1). A representation of the processes of removal of uranium from groundwater through reduction of U(VI) and solubility control of U(IV) was made using the PHREEQC code as this is thought to be the primary mechanism (9). A simple speciation model based on equilibrium chemistry using PHREEQC has been developed to model precipitation within the zero-valent iron reactive barrier.

The geochemical environment within the PRB is driven principally by the corrosion of  $\text{Fe}^0$ . In the presence of dissolved oxygen, this corrosion occurs via the following reaction,

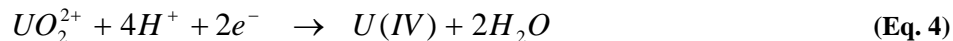


However, under anaerobic conditions, corrosion occurs according to,



While the groundwater within Area 2 is generally aerobic, the current approach does not model corrosion explicitly but only considers the secondary anaerobic corrosion products formed.

The rate at which corrosion occurs will be dependent on a number of factors, including pH and the concentrations of aqueous species. It is postulated (9) that the primary mechanism for remediation of uranium by the PRB involves reductive precipitation, whereby the influent U(VI) is reduced to U(IV) which readily forms precipitates, leading to a significant reduction in aqueous uranium. Reduction of U(VI) occurs according to the following stoichiometric reactions,



A simpler representation of the chemical features of the ZVI is proposed which considers only the typical anaerobic corrosion products that will form. The PHREEQC modelling presented here investigates the chemical conditions that are established by buffering reactions of the mineral corrosion products which are considered to form by anaerobic corrosion. The PHREEQC model of the barrier is thus defined by these buffering mineral phases. This approach considers a homogeneous distribution of corrosion products and cannot be used to investigate the temporal variation in the properties of the ZVI PRB that result from corrosion. The model will however be able to consider the temporal changes in geochemistry and mineralogy that result from the flow of groundwater and this may be of relevance to processes such as clogging of the barrier such as due to the precipitation of carbonates in the high pH vicinity of the PRB.

In addition to reactions involving iron and uranium, numerous other aqueous phase and precipitation reactions are included in the current modelling. The most significant aqueous phase reactions include redox reactions involving reduction of nitrate and sulphate and complexation of the various master species. In addition to uraninite and calcite, other mineral phases included siderite, troilite, Fe(OH)<sub>2</sub> and amorphous UO<sub>2</sub> to simulate the effect of the zero-valent iron corrosion products in the PRB. All mineral phases are allowed to precipitate freely according to thermodynamic equilibrium.

As outlined above the conceptual geochemical speciation model implemented within PHREEQC V2.8 (1) has been adopted. It is assumed in the current modelling that the resulting U(IV) may precipitate as uraninite or amorphous UO<sub>2</sub>. Equilibrium conditions are assumed for all reactions.

The PHREEQC model developed considered the effect of typical iron corrosion products observed at the site (9) which act to buffer the pH, Eh (pe) and carbonate concentration which are key variables that affect the speciation of uranium. These initial calculations considered a mineral assemblage of Fe(OH)<sub>2</sub>, troilite (FeS) and siderite (FeCO<sub>3</sub>) which resulted in the establishment of alkaline (pH 10) and reducing chemical conditions where the solubility of uranium was lowered from 4.2e-06 mol/l (~1mg/l representative of contaminated groundwater) to 5.9e-08 mol/l within the PRB. PHREEQC speciation calculations were undertaken to provide data for different models and inputs for the TRAFFIC and PHAST models representing,

- the geosphere groundwater, and
- the reduced groundwater present in the ZVI PRB

The two groundwater compositions are defined by the mineral buffers present in the PHREEQC model. The geosphere region contains solely calcite (CaCO<sub>3</sub>), whereas the ZVI contains the corrosion products; Fe(OH)<sub>2</sub>, troilite (FeS) and siderite (FeCO<sub>3</sub>). An excess of 10 moles/litre of porewater of each of these minerals was included in the model. In addition, within the ZVI region the uranium (IV) phase UO<sub>2</sub>(am) was allowed to precipitate in the model, where it becomes oversaturated and in this way the precipitation and immobilisation of uranium can be simulated.

## Hydrogeochemical modelling using TRAFFIC

The speciation model described above is implemented within the chemical model for the transport and speciation modelling using the BNFL in-house code TRAFFIC (2, 3).

TRAFFIC model with and without the remediation effects of the Y-12 zero-valent PRB on uranium has been run for a 5 year period. Figure 1 shows the uranium concentrations after 5 years, with the PRB in place, showing reduced concentrations downgradient of the PRB. Comparisons of computed uranium concentrations with measurements up- and down-gradient of the PRB are shown in Figure 2. As expected at upgradient locations predictions from models with and without reactive chemistry are essentially the same, while the effect of uranium reduction downgradient of the PRB is apparent. However it is apparent that the equilibrium geochemical model used here does tend to over-predict removal of uranium within the barrier.

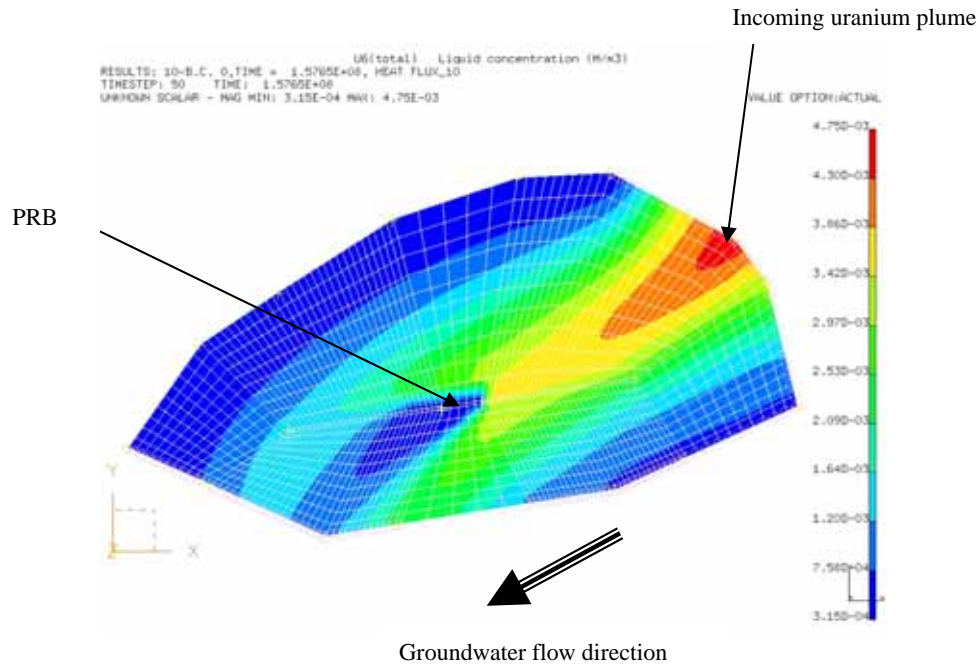
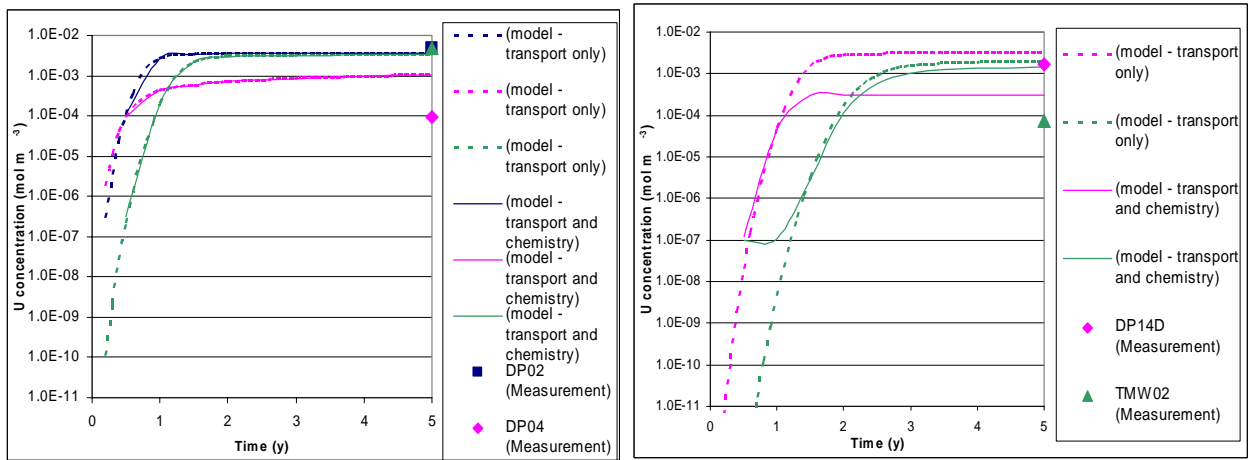


Fig. 1 Uranium concentration from the TRAFFIC model after 5 years.

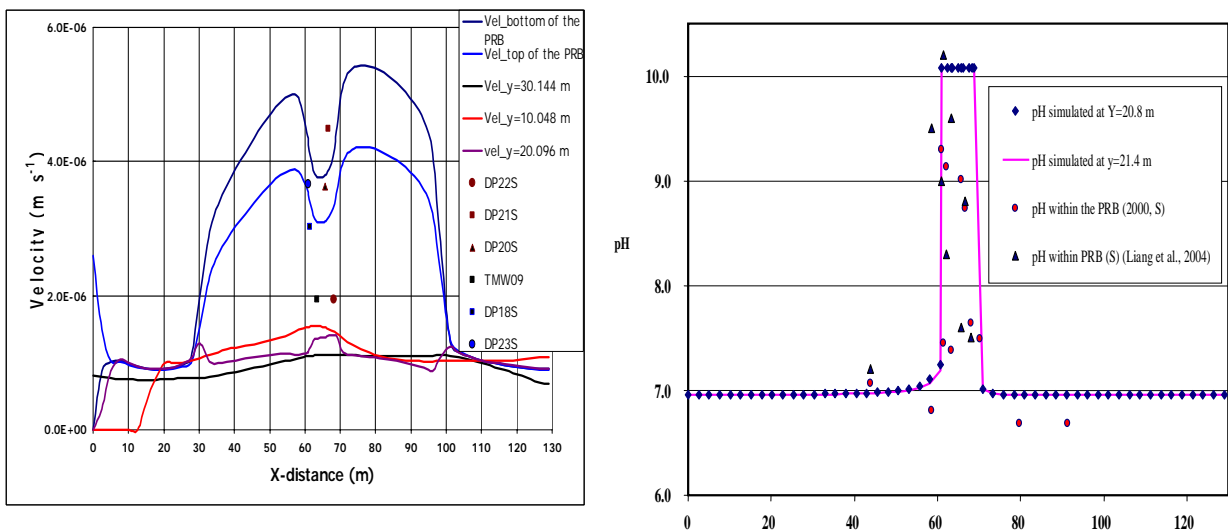


**Fig. 2 Comparison of computed uranium concentrations with measurements: upgradient (left) and downgradient (right) of PRB.**

### Hydrogeochemical modelling using PHAST

A PHAST model was used to model the remediation of groundwater contaminated with uranium using a permeable reactive barrier at the Y-12 site. PHAST uses rectangular finite difference meshes, the irregular domain shape used in the TRAFFIC model is implemented within a rectangular domain with boundary conditions applied at the nearest neighbouring nodes. The resulting model domain is 129 m long by 62.8 m wide by 9 m deep such that its length is oriented parallel to the permeable reactive barrier. Finer grids are used at the reactive core to resolve the expected rapid changes in the region.

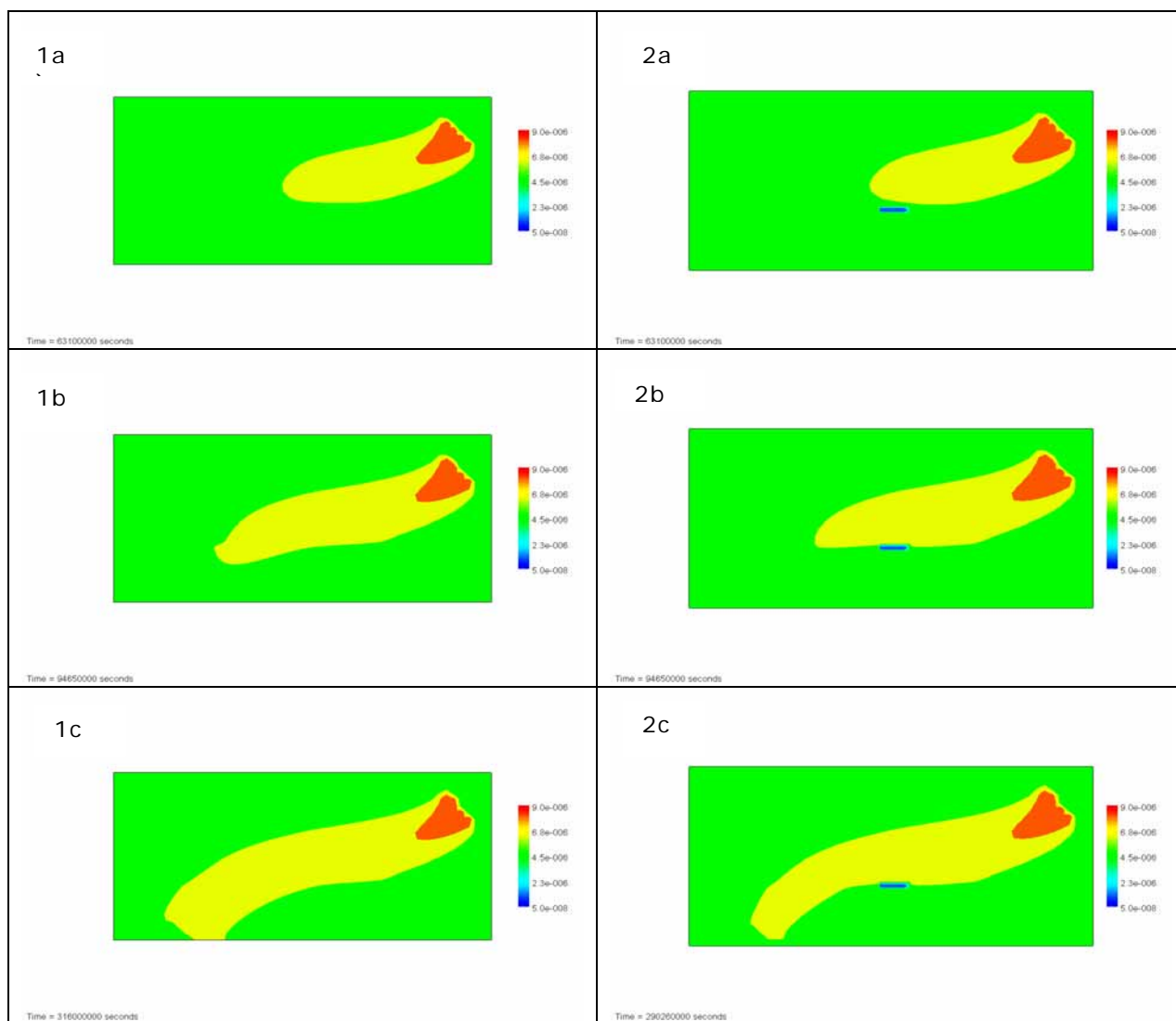
Comparison of velocity magnitudes from PHAST and field data are made at  $y=10.048$  m,  $20.096$  m,  $20.8$  m (PRB bottom or downgradient),  $21.4$  m (PRB top or upgradient) and  $30.144$  m (Figure 3). The rise in groundwater velocity within the gravel trenches and the iron core of the PRB can be clearly seen. The predicted velocities compare well with the observed velocities. Also shown are the calculated pH values indicating a pH increase in  $Fe^0$  media which generally agrees with observation over the 5 year period (10). The model predicts a pH of 6.75 upgradient of the barrier, increasing quickly to a maximum value of 10.1 within the iron core reactive, which is in good agreement with observations.



**Fig. 3 Comparison of modelled results from PHAST to measurements (10): i) groundwater velocity (left); and ii) pH (right), across the PRB .**

Figure 4 shows the spatial development of the predicted total uranium plume over a 10 year period with and without the PRB included, with green denoting the low background concentrations and yellow to red the

higher concentrations. Both simulations give similar results at the end of 1 year when the plume is about to reach the reactive core region. Removal of uranium by the barrier can be clearly seen after 3 years. Computed results indicate that uranium is effectively removed by the barrier and that uranium concentrations in the PRB are generally very low ( $<5 \times 10^{-8} \text{ mol l}^{-1}$  denoted blue) with over two order of magnitude reduction. It is noted (9) that uranium immobilised on entering the PRB, an observation supported by the relative abundance of uranium within core samples from the upgradient end of the barrier. This suggests that the reactive barrier is performing well in removing or retaining uranium from the contaminated groundwater. The PHAST model also shows that the plume is not completely ‘captured’ by the PRB and that parts of the plume ‘spill over’ downgradient of the gravel trench. This prediction by the present model is supported by the subsequent need to extend the downgradient gravel trench at the Y-12 site. This clearly demonstrates the value of modelling as a cost-effective means to assess and guide design at an early stage.



**Fig. 4 Computed uranium concentration ( $\text{mol l}^{-1}$ ) from two calculations (i.e. without PRB (1); and with PRB (2)) at different time (a=1 year; b=3 years and c=10 years).**

#### Related Modelling work

SimER has been used to investigate the hydrogeological features of the PRB using a kilometre scale model. In addition, the code was used to model the specific time evolution and events which are essential to fully



understand and compare the effect of a remediation technology. The SimER model assumes, on the basis of the above underpinning modelling that within the remediation zone uranium is solubility limited to a concentration of  $1\text{e-}4 \text{ mol m}^{-3}$ . Elsewhere, uranium migration is affected by sorption processes which are represented by sorption coefficients typical of the soils and underlying saprolite rock. Initial results from the SimER models are presented in a separate paper (11) for this conference.

## DISCUSSIONS

The present model demonstrates the validity of the approach to model the temporal changes in geochemistry and mineralogy, and in modelling remediation of contaminated groundwater by the PRB. Good agreement have been obtained between the model and measurements for groundwater flow, and the modelled contaminated uranium plume concentrations broadly in agreement with field data. A number of issues have been identified from the present modelling exercise related to undertaking assessments with limited or no site-specific information, which directly affecting model data input implementation:

- Hydrogeological data: The Y-12 site is known to contain non-homogeneous geological media and structures at the top soil layer, with fractured rocks underneath potentially influencing groundwater flow behaviour. Obviously this presents a difficult situation for any model to represent the complex geology in detail even if field data exist, which are highly unlikely in practice. The approach adopted here is to use the field hydraulic head as model input to derive the overall groundwater flow. Averaged characterisation parameters for soil (e.g. hydraulic conductivity, porosity etc.) and groundwater (e.g. species concentration) are used in model implementation. This should produce an overall flow that is broadly consistent with the groundwater flow in the region, although localised changes due to heterogeneity will not be represented.
- Plume data: Plume input data (i.e. both the maximum and concentration gradient values) for the present modelling is estimated from selected borehole measurements upgradient of the PRB. These are assumed to be time invariant, while in reality they may be time varying. Hence the present modelling only provides a broad representation of plume input, but is useful to demonstrate the capability of the modelling approach to remediation assessment. Ideally temporal concentration data of sufficient spatial resolution are available to represent the plume to provide model solute boundary conditions. It is also important that hydrogeological data covering the same period are available to provide a consistent model, such that detailed comparison of plume development against field measurements can be assessed.
- Chemistry data: No attempt has been made in the current modelling to include the effects of uranium sorption on to corrosion products, even though this has been postulated as being a possible (but less probable) route for uranium retention within the barrier. If suitable data could be found, such a mechanism could be included in the speciation model in order to gauge the relative significance of the two remediation mechanisms, thus gaining valuable insight into the likelihood for future remobilisation of uranium.
- Chemical speciation modelling: The present model clearly demonstrate the validity of the approach to model the temporal changes in geochemistry and mineralogy, and in modelling remediation of contaminated groundwater by the PRB. However it is apparent that the equilibrium geochemical model used here does tend to overpredict removal of uranium within the barrier. The overprediction of its removal from solution points to the need for consideration of kinetic descriptions for precipitation of  $\text{CaCO}_3$  and/or siderite. Such kinetic processes can be readily incorporated into PHREEQC, subjected to site-specific data being available.

## **CONCLUDING REMARKS**

The present model demonstrates the validity of the approach to model the temporal changes in geochemistry and mineralogy, and in modelling remediation of contaminated groundwater by the PRB. Comparisons are made between the modelled results and field measurements. Very good agreement between the model and measurements has been obtained for groundwater flow. The modelled contaminated uranium plume concentrations are broadly in agreement with field data.

It has been shown that the conceptual model suggested for the geochemical reactions occurring within the PRB can reasonably reproduce field measurements for the remediation of uranium. In particular, the use of an equilibrium model for uranium speciation appears to broadly represent the dominant reductive process from U(VI) to U(IV) (9), although the use of equilibrium geochemical model tends to overpredict removal of uranium within the barrier.

The need for the various site characterisation data covering the modelled period has been highlighted to enable detailed assessment of the modelled results against field measurements.

## **ACKNOWLEDGEMENTS**

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