International Journal of GEOMATE, Nov., 2018 Vol.15, Issue 51, pp.9-15
Geotec., Const. Mat. & Env., DOI: https://doi.org/10.21660/2018.51.7260
ISSN: 2186-2982 (Print), 2186-2990 (Online), Japan

GEOCHEMICAL REACTIVE MODELING OF FLOW AND TRANSPORT PROCESS AT A MINE SITE IN NORTHERN TERRITORY, AUSTRALIA

*Michael Hayford[1], Bithin Datta[1,2]

[1]College of Science and Engineering, James Cook University, Australia; [2] CRC-CARE, University of New Castle, Australia

*Corresponding Author, Received: 12 June 2017, Revised: 13 April 2018, Accepted: 14 May 2018

ABSTRACT: Subsurface contamination of metals and radionuclides are common problems in abandoned mine sites. Metals and radionuclides are likely to occur in the mobile phase or may bind to soil particles and geologic framework. The transport process for reactive contaminants also include chemical reactions in groundwater aquifers, which are complex physical and biogeochemical processes. Accurate tools to reliably predict the movement and changes in concentration of metals and their daughter products (radionuclides) formations in the subsurface environment is important for decision makers to analyze impact, and to evaluate the effectiveness of remediation strategies. An advanced numerical model can provide such useful direction of predictions. A 3-D contaminant transport model for fluid flow, hydrogeologic transport, and biogeochemical kinetic and equilibrium reactions in saturated and unsaturated media was calibrated, validated and implemented to model the hydrogeochemical processes that occur in the subsurface at such a complex contaminated site, consisting of waste rock dumps and flooded mine pits of this abandoned uranium mine site. The developed flow and transport simulation model for no longer in use mine site in Northern Territory of Australia is discussed.

Keywords: Reactive Transport, Flow and Transport Modelling, Groundwater Contamination, Mine site

1. INTRODUCTION

Groundwater plays an important role in human history and groundwater contamination has attracted intensive investigations in the past 30 years. Contaminants in the aqueous environment undergo changes in concentration resulting from physical, chemical, biological processes, and a capability to understand and model these processes is at the core of assessing the efficacy of environmental remediation strategies. The modeling and prediction of the complex multiple species reactive transport process in mine sites is particularly challenging. The development of an accurate model capable of predicting the transport scenario is very challenging, but a necessity for designing future remediation strategies. Therefore, a robust numerical multiple species reactive transport simulation model HYDROGEOCHEM 5.0 [1] has been calibrated and validated for a study area comprising of an underlying aquifer in a no longer operational mine site in the Northern Territory of Australia. Acid rock drainage (ARD) and heavy metal mobilization at the former mine site have led to significant environment impacts on local groundwater and in the East Branch of the Finniss River [2].

Acid and metalliferous drainage (AMD) also referred to as acid mine drainage or acid rock drainage occurs when sulphidic minerals, such as arsenopyrite, chalcopyrite and pyrite, are exposed to oxygen and water. Although AMD occurs naturally, the majority of AMD is anthropogenic and originates from reactive sulfide minerals stored in waste rock dumps (WRDs), mine pits, and tailings. Notwithstanding the fact that extensive rehabilitation studies at the mine site exist, a well-defined reactive transport model, with a comprehensive suite of reaction types and reaction rates incorporated into a model aimed at the remediation and rehabilitation of the site, has not been implemented yet. These reaction types include but not limited to aqueous complexation, adsorption-desorption, ion exchange, oxidation-reduction, precipitation-dissolution, acid-base reactions, and microbial mediated reactions.

The main sources of pollutants to the environment are the open pit, the waste rock damps and the tailings dam. The waste rock damps and tailing dams most often consist of several minerals and geological matrix, which are exposed to weathering conditions. Pyrite oxidation is the principle reaction in the leaching of metal and radionuclides into the environment. Accurate prediction of the release rate of metal and radionuclides from these sources and their transport into the subsurface environment is a critical factor to the assessment of environmental impacts and to the development of effective remediation strategies. To produce a realistic

representation of the system under study with the existing real complex problems, sophisticated models are required. Reactive transport codes are powerful tools in the evaluation of coupled hydrological chemical processes and in the prediction of the long-term performance of remediation strategies.

A presentation of the application of a reactive-transport model to simulate the transport of reactive multi-species contaminants in heterogeneous, anisotropic, saturated-unsaturated waste rocks damp and mine open pits of the first uranium mine site in Australia is illustrated in this paper. The computer program used to describe the complex hydrogeochemical processes that control the generation of acid drainage and the distribution of solutes between groundwater and waste rock damp is the hydrogeochemical transport model, HYDROGEOCHEM 5.0. This numerical code has been earlier used for modeling a mine site in Queensland Australia [3]. The computer program is a 3-D numerical model of fluid flow, thermal, hydrologic transport, and biogeochemical kinetic and equilibrium reactions in saturated and unsaturated media.

The purpose of this paper is to focus on the solution results of reactive transport geochemical modeling to predict the important complex processes that control the movement and distribution of contaminants in groundwater. It is also likely that this calibrated and validated transport model will be utilized for a contamination source characterization study for this contaminated aquifer site. Information gained from these investigations should also provide insight into processes that occur at other sites similarly contaminated.

2. NUMERICAL SIMULATION

2.1 Flow Model

The general equations for flow through saturated-unsaturated media are obtained based on following fluid continuity, solid continuity, fluid movement (Darcy's law), stabilization of media, and water compressibility [1]

$$\frac{\rho}{\rho_o} F \frac{\partial h}{\partial t} = -\nabla \cdot \left[K. \left(\nabla h + \frac{\rho}{\rho_o} \nabla z \right) + \frac{\rho^*}{\rho_o} q \right] \qquad (1)$$

Where F = generalized storage coefficient (1/L) defined as

$$F = \alpha' \frac{\theta}{n_e} + \beta' \theta + n_e \frac{dS}{dh} \qquad (2)$$

$$\boldsymbol{K} = \frac{\rho g}{\mu} \quad K = \frac{\rho / \rho_O}{\mu / \mu_O} \frac{\rho_o g}{\mu_O} \quad K_s K_r = \frac{(\rho / \rho_o)}{(\mu / \mu_o)} K_{so} k_r \qquad (3)$$

and V = Darcy's velocity (L/T), described as:

$$\mathbf{V} = - \boldsymbol{K} \cdot \left(\frac{\rho_o}{\rho} \nabla h + \nabla z \right) \qquad (4)$$

Where: $\boldsymbol{\theta}$: effective moisture content (L^3/ L^3); h: pressure head (L); z is the potential head (L), t: time (T); q: source or sink of fluid [(L^3/ L^3)/T]; $\boldsymbol{\rho_0}$: fluid density without biochemical concentration (M/ L^3); $\boldsymbol{\rho}$: fluid density with dissolved biochemical concentration (M/ L^3); ρ*: fluid density of either injection (= ρ*) or withdraw (= ρ) (M/ L^3); $\boldsymbol{\mu_o}$: fluid dynamic viscosity at zero biogeochemical concentration [(M/L)/T]; μ: the fluid dynamic viscosity with dissolved biogeochemical concentrations [(M/L)/T]; α': modified compressibility of the soil matrix (1/L); ß: modified compressibility of the liquid (1/L); ne: effective porosity (L3/L3); S: degree of effective saturation of water; G: is the gravity (L/T2); k: permeability tensor (L2); ks: saturated permeability tensor (L2); Kso: referenced saturated hydraulic conductivity tensor (L/T); kr: relative permeability or relative hydraulic conductivity (dimensionless).The finite element method was used to solve Eq. (1), (2), (3) and (4), and the constitutive relationships among the pressure head, degree of saturation, and hydraulic conductivity tensor, together with the appropriate initial conditions and boundary conditions. The temporal-spatial distributions of the hydrological variables, including pressure head, total head, effective moisture content, and Darcy's velocity are simulated.

2.2 Reactive Transport Model

Governing equations for the reactive transport of the reactive biogeochemical system is discussed below. The equations for transport were derived based on the continuity of mass and Fick 's flux laws. The main transport and fate processes are advection, dispersion/diffusion and biogeochemical reactions (including radioactive decay). The general transport equation governing the temporal-spatial distribution of any biogeochemical species in a reactive system is described below [1]:

$$\frac{\partial \theta C_i}{\partial t} + \theta \alpha' \frac{\partial h}{\partial t} C_i = L(C_i) + \theta r_i + M_i, i \notin \{M\} \qquad (5)$$

Where L is the transport operator denoting

$$L(C_i) = -\nabla \bullet (V C_i) + \nabla \bullet \left[\theta D \bullet \nabla C_i \right] \qquad (6)$$

where Ci is the concentration of the i-th species in units of chemical mass per water volume [M/L^3]; r_i is the production rate of the i-th species because of biogeochemical reactions in chemical mass per water volume per unit time [$M/L^3/T$];

{M} = {1,2,…,M} in which M is the number of biogeochemical species; D is the dispersion coefficient tensor [L^2/T]; and M_i is the source/sink of the i-th species in chemical mass per unit volume of media [M/L^3/T].

3. STUDY AREA DESCRIPTION

The former mine site is located approximately 105km by road south of Darwin in the headwaters of the East Branch of the Finniss River, near Batchelor in the Northern Territory, Australia. This former mine was one of Australia's first major uranium mines and produced approximately 3,500 tonnes of uranium and 20,000 tonnes of copper concentrate between 1954 and 1971 [4]. Uranium was mined from White's and Dyson's open pits from 1954 to 1971, while copper was extracted from the Intermediate pit in 1964. The site underwent rehabilitation from 1983 to 1986 at a total cost of $18.6 million. Several rehabilitation works have taken place since 1986 and rehabilitation is still on going. [4].

3.1 Geological and Hydrogeological Characterization

The mine areas consist of east branch of the Finniss River about 8.5 km upstream of its confluence with the West Branch of the Finniss River. Surface water enters the mine site from the east via the upper East Branch of the Finniss River and from the southeast via Fitch Creek. River flows vary in response to intra annual variability in rainfall and changes over the course of a year [5]. The mine area mineral field contains polymetallic ore deposits, such as the Ranger and Woodcutters ore deposits. The mine site includes the Giant's reef fault. The main lithological units are the Rum Jungle Complex and meta-sedimentary and subordinate meta-volcanic rocks of the Mount Partridge Group. The Rum Jungle Complex consists mainly of granites and the Mount Partridge Group consists of sedimentary units: Geolsec formation, the Whites formation, the Coomalie Dolostone, and the Crater formation [6].

Fig. 1 Satellite view of the study site

3.2 Conceptualization and Model Discretization

A numerical groundwater flow model is constructed to simulate variations in the groundwater flow system at the mine site from December 2010 to November 2014. This numerical flow model is a mathematical representation of a conceptual model that enables a quantitative representation of real field features. The numerical representation is based on the following assumptions: The aquifer system at the former mine site can be subdivided into hydrostratigraphic units that represent either waste rock damps or the naturally occurring bedrock aquifer, each hydrostratigraphic unit is represented as a single model layer with representative hydraulic properties and recharge is estimated as a proportion of incident rainfall, mine waste is represented by a single model layer of variable thickness of the waste rock damps and Dyson's (backfilled) open Pit, whereas the geological aquifer units are represented by model layers with constant thicknesses across the model domain, water movement in the hydrostratigraphic units follows Darcy's law, the flooded open pits is represented by specified head boundary condition that is equivalent to observed water levels in the pits during the simulation period.

3.2.1 Numerical method

The finite element method is used for temporal and spatial discretization of the governing partial differential equations in the flow module. The Galerkin finite element method was used for spatial discretization of the modified Richards equation that governs the distribution of pressure fields. For this model, the critical assumption of this approach is that every element within a particular section of a layer is assigned a set of hydraulic properties to account for heterogeneity. The layers are discretized horizontally into triangular wedge elements but each layer of a finite element mesh is one element thick. The flow of the groundwater is simulated for the observation period 2010 and 2014. Two case scenarios of both steady state and transient flow is simulated for the flow.

3.2.2 Spatial discretization

The numerical model domain was spatially

discretized into a 3-dimensional mesh with a triangular wedge mesh. In planar view, each element is a triangular wedge, whereas the thickness of the elements depends on the number of layers used to vertically discretize the model domain. The thickness of the elements varies depending on lithology. The model is made up of 6 layers and covers a maximum elevation of about 110 m. Surface topography from a recent terrain elevation data was used to define the top of layer 1, including the WRDs and Dyson (backfilled) pit. Figure 2 shows a plan view of finite element model domain. Layer thicknesses are allocated in this order. Layer 1 which consist mainly of waste rock damps is assigned variable thickness, layer 2 thickness is from 0 to 7.5 m, layer 3m is 7.5m to 15 m, layer 4 is 15m to 45 m, layer 5 is 45m to 105 m, and layer 6 is 105m to 150 m. The tops and bottoms of layers 3 to 6 are set to thicknesses values listed above which is fixed throughout the model.

3.2.3 Boundary conditions

Specified heads boundary conditions are assigned to elements from layers 1, 2, 3, and 4 that intersects the perimeters of the flooded pits which includes main, intermediate and brown oxide pits. The elements represent the bedrock aquifer that is in contact with standing water within the pits and was assigned a head that is equal to the measured water level in the pits. Flows within the flooded open pits themselves are not simulated by the model so elements within the head boundary are set to be inactive. Pit water levels and groundwater levels at monitoring locations close are used to represent the open pit as a head boundary. The browns oxide open pit is not completely flooded so heads were only specified for elements in layers 3 and 4. Time varied constant head nodes are set that simulate water level changes in the main, intermediate and brown's pits and the river. The aquifer parameters imputed to the model area is shown in Table 1

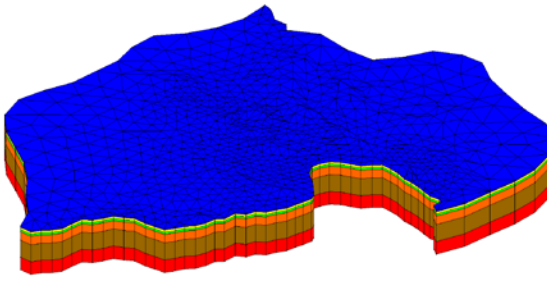

Fig. 2 Three dimensions finite element representation of the study site

To model and simulate the reactive transport processes, equations of either the conventional finite element methods or the hybrid Lagrangian–Eulerian finite element methods were used for spatial discretization. The chemical equilibrium equations were solved by the Newton–Raphson method or Picard method. This study considers the possible hydrogeochemical transport of 6 components, including OH-, $Cu^{2+,}$ Fe^{2+}, Fe^{3+},Mn^{2+}, UO_2^{2+} and 20 species are considered in this study. These selected sets of components are based on the fact that the primary mining commodities were uranium and copper. Table 2 shows some chemical reactions incorporated in the model.

Table 1 Aquifer hydrogeologic properties

Aquifer parameter	Value
Number of nodes	6587
Number of elements	10704
Horizontal hydraulic conductivity, K_x K_y	m/d
Layer 1	0.01728 - 5.0112
Layer 2	0.00864 - 0.7776
Layer 3	0.00864 - 0.8467
Layer 4	0.00864 - 0.8467
Layer 5	0.00864 - 0.0259
Layer 6	0.0001728 - 0.0648
Vertical hydraulic conductivity, K_z	m/d
Layer 1	0.00864 - 0.2419
Layer 2	0.00432 - 2.4192
Layer 3	0.00864 - 0.8640
Layer 4	0.00864 - 1.2960
Layer 5	0.00864 - 0.2074
Layer 6	0.00864 - 0.0648
Effective porosity, θ	0.28
Longitudinal dispersivity, αL	10 m/d
Transverse dispersivity, αT	0.1 m/d
Vertical dispersivity (αV):	0.01
Average rainfall	2372 mm/year

Table 2 Chemical reactions involved in transport

Chemical reactions	Constant rate (log K)
$H_2O(aq) \rightarrow H^+ + OH^-$	-13.99
$H^+ + SO_4 \rightarrow HSO_4^-$	1.99
$Cu^{2+} + H_2O \rightarrow Cu(OH)^+ + H^+$	-9.19
$Cu^{2+} + SO_4^{2-} \rightarrow CuSO_4$	2.36
$Cu^{2+} + 2H_2O \rightarrow Cu(OH)_2 + 2H^+$	-16.19
$Cu^{2+} + 3H_2O \rightarrow Cu(OH)_3^- + 3H^+$	-26.9
$Fe^{2+} + H2O \rightarrow H^+ + FeOH^+$	-9.50
$Fe^{2+} + SO_4^{2-} \rightarrow FeSO_4$	2.20
$Fe^{2+} + 2H_2O \rightarrow 2H^+ + Fe(OH)_{2\ (aq)}$	-20.57
$Fe^{2+} + 3H_2O \rightarrow 3H^+ + Fe(OH)_3^-$	-31.00
$Fe^{2+} + 4H^2O \rightarrow 4H^+ + Fe(OH)_4^{2-}$	-46.00
$Mn^{2+} + SO4^- \rightarrow MnSO_4$	2.26
$Mn^{2+} + H_2O \rightarrow MnOH^+ + H$	-10.59

$Mn^{2+} + 3H_2O \rightarrow Mn(OH)_3^- + 3H$	-34.08
$UO_2^{2+} + SO_4^{2-} \rightarrow UO_2SO_4$	3.15
$UO_2^{2+} + SO_4^{2-} \rightarrow UO_2(SO4)_2^{2-}$	4.14
$UO_2^{+2} + 2H_2O \leftrightarrow UO_2(OH)_2\ aq + 2H^+$	12.15

3.2.4 Conceptual modeling

The transport model for the mine site was built upon the flow model by including the necessary transport conditions, which includes the transport boundary conditions, the total number of components and the species to be simulated. The assumption is that the transport of the contaminants is based solely on the flow fields simulated whereby there is a possible rock interaction or mineral interactions with the aquifer rock bed and formation of daughter products or additional species.

It is assumed that the oxidative dissolution of pyrite is the driving force in the discharge rate of metal and radionuclides from waste rock dumps. The reactive system is completely defined by geochemical reactions through a network of reactions specifying chemical reactions and the total number of chemical species involved in the reaction. The standard equilibrium expression with a suitable equilibrium constant is used to represent all the fast reactions, such as the aqueous complexation reaction and precipitation of secondary phases. The slow reactions were represented by the kinetic expression and associated rate constants to address the dissolution reactions of key minerals present in the waste rock dumps and pits. The reaction network that describes the evolution of the geochemical system is shown in Table 2.

4. RESULTS AND DISCUSSION

The flow model calibration is carried out by running the forward simulation repeatedly and manually adjusting the input parameters selected for calibration, including those used to define initial and boundary conditions, at each time, until a satisfactory match between model results and field data is achieved. The trial and error procedure is used in this study. This is adopted because it is conceptually straightforward and requires nothing more computationally than making multiple forward simulations runs. This procedure is flexible allowing any adjustment in parameter values and parameter structures, including changes in mesh designs and the representation of the geologic framework

The main aim of calibration was to obtain the estimation of hydraulic conductivity and groundwater levels for the aquifer based on limited field measurement data. Hydraulic head measurement data from 20 monitoring locations distributed across the mining area were used for the simulation model calibration. The hydraulic head data used for model calibration and validation were recorded in 2010 and 2014. A portion of the average rainfall intensity per year was specified as a recharge for calibration of the simulation model also using measured head data from monitoring locations. Calibration targets for the developed model were set to be within 2m intervals of the observed hydraulic head value in monitoring locations with a confidence level of 90%. Exact boundary conditions are difficult to determine with very limited observed measurement data. Hence, realistic boundary conditions need to be specified that matches the site conditions. In the calibration process, one of the most difficult issues is to assign appropriately the correct boundary conditions. Therefore, the boundary conditions need to be determined based on preliminarily calibration results as well. The model's boundary conditions are adjusted manually to achieve the calibration targets. The measured and simulated heads were compared at selected points.

The developed model was calibrated for flow using observed head data obtained at monitoring points. In the calibration process, estimates of the values of recharge for each recharge zone and the representative hydraulic conductivity of each layer were calibrated. Results of simulated heads calibration are shown in Table 3.

Table 3 Comparison of observed and simulated heads

Location ID	observed head(m)	computed head(m)	Residual (m)
P1a	61.44	62.48	-1.04
P1b	62.65	63.65	-1
P2	63.27	63.12	0.15
P3	60.29	64.9	-0.11
P4	60.41	60.86	-0.45
P5	57	57.14	-0.14
P6	58.33	59.09	-0.76
P7	56.39	59.05	-2.66
P8D	53.96	54.68	-0.72
P9D	56.57	56.69	-0.12
P10	57.63	58.03	-0.4
P11	57.16	57.87	-0.71
P12	56.44	56.91	-0.47
P13	56.89	56.9	-0.01
P14	57.06	57.55	-0.49
P16	56.27	57.16	-0.89
P17	56.45	57.29	-0.84
P18	57.28	58.27	-0.99

P19	57.35	58.23	-0.88
P20	51.01	51.69	-0.68

The flow model is calibrated first to adjust the model input parameters that control the flow process, such as hydraulic conductivity and recharge. When the flow model is calibrated to a certain degree, calibration of the transport model is initialized to adjust those parameters specific to the transport model, such as porosity, dispersivity and concentration. Recalibration of the flow model is done to further adjust the initially calibrated flow parameters. After readjustment of the flow parameters, a new velocity distribution is calculated and used in the transport model where further adjustments in the transport parameters are made. This iterative procedure is repeated until both flow and transport related field data are matched by the simulation results. The results of the calibrated heads against the observed heads is shown in Table 3. These results show fairly good history matching of the hydraulic heads. The reactive transport model simulation time is 2 years from 2012 to 2014 as of the availability of data. The chemical processes of aqueous complexation, precipitation-dissolution, adsorption-desorption, ion-exchange, redox, acid-base reactions and the changes of porosity, hydraulic conductivity, and hydrodynamic dispersion of aquifer media may be a key hydrogeochemical mechanism in the transport process. Thus, the effect of kinetic and equilibrium reactions on both flow and reactive transport are also considered in the simulations.

Copper (Cu^{2+}), sulphate (SO_4^{2-}), Manganese (Mn^{2+}), Uranium (UO_2^{2+}) and Iron (Fe^{2+}) are the main species of study. The concentrations of each species at specific monitoring points is utilized as initial concentration which is assume to be involved in chemical reactions listed in Table 2. The assumed initial conditions for contaminant concentrations are specified in the simulation model based on the available data and extrapolation. The sources considered for this study are dysons waste rock damp, dyson open pit, main waste rock damp, intermediate waste rock damp, main pit and intermediate pit. The objective was to demonstrate the flexibility of the model to adapt a known reaction network to an abandon mine site setting and to demonstrate the capability of the code to simulate large complex problems. The results of the simulated concentrations are shown in the Figure 4, Figure 5 and Figure 6.

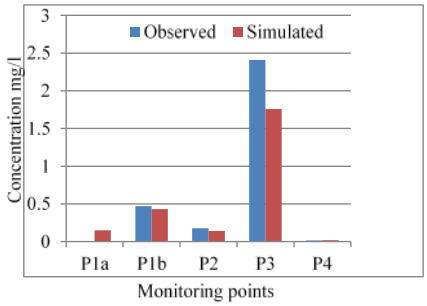

Fig. 4 Comparison of observed and simulated copper concentrations mg/ l at monitoring points

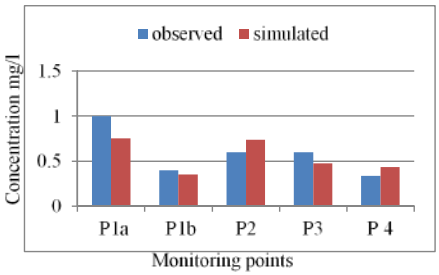

Fig. 5 Comparison of observed and simulated iron concentrations mg/ l at monitoring points

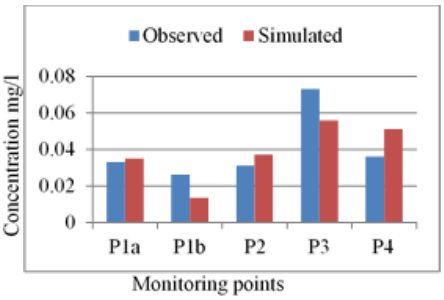

Fig. 7 Comparison of observed and simulated uranium concentrations mg/ l at monitoring points

5. CONCLUSION

The calibrated flow model, and the developed transport simulation model are capable of simulating the complex flow and multiple species reactive transport processes in the geologically complex multiple layer, heterogeneous aquifer comprising of a no longer in use uranium mine site in NT, Australia. The aquifer processes are considered as 3-dimentional, geologically layered and heterogeneous, with transient flow and transport. It was possible to calibrate the flow model by using sparse spatial-temporal hydraulic head measurements. The contamination simulation scenario resembled the field conditions to a certain extent. The concentrations predicted for the chosen species match with the concentration measurement values available. Therefore, the simulation model, based on HYDRGEOCHEM once calibrated for site conditions, is potentially a good candidate to

model the complex flow and transport processes in a geochemically and geologically complex mine site aquifer.

6. ACKNOWLEDGEMENTS

The authors acknowledge the financial support for this work provided by CRC-CARE, University of New Castle, Callaghan NSW 2308, Australia and James Cook University, Australia.

7. REFERENCES

[1] Yeh GT, Sun J, Jardine PM., Burgos WD, Fang Y, Li M., Seigel M.D, ("HYDROGEOCHEM 5.0:AThree-Dimensional Model of Coupled Fluid Flow, Thermal Transport, and Hydrogeochemical Transport through Variably Saturated Conditions - Version 5.0", 2004

[2] Kraatz M, "Rum Jungle Rehabilitation Site Scoping Study, Environmental Issues and Considerations for Future Management", Report to the Department of Industry, Tourism and Resources, M4K Environmental Consulting, February 2004.

[3] Esfahani HK, Datta B, "Linked Optimal Reactive Contaminant Source Characterization in Contaminated Mine Sites: Case Study", Journal of Water Resources Planning and Management, Vol 142, Dec. 2016

[4] Norther Territory Government. "Rum Jungle Conceptual Rehabilitation Plan", May 2013

[5] Ferguson PR, Wels C, Fawcett M, "Current water quality conditions at the historic Rum Jungle Mine Site, northern Australia". Proceedings of 9th International Conference on Acid Rock Drainage ICARD), May 20-26, 2012, Ottawa, Canada.

[6] Robertson GeoConsultants Inc., Phase 3 (Stage 2 Report) "Groundwater Flow Model for the Rum Jungle Mine", RGC Report submitted to NT DoR, May 2012.

Copyright © Int. J. of GEOMATE. All rights reserved, including the making of copies unless permission is obtained from the copyright proprietors.