Modelling Concrete Degradation by Coupled Non-linear Processes

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Received 14 June 2021, accepted 6 October 2021 doi:10.3151/jact.19.1075

Abstract

Concrete in a transuranic (TRU) waste repository is considered a suitable material to ensure safety, provide structural integrity and retard radionuclide migration after the waste containers fail. Modelling of concrete degradation often focuses solely on solid-water chemical reactions and related changes in porosity, however, cracking and damage from steel corrosion will likely exert significant control over the mass-transport properties of concrete. In the current study, coupling between chemical, mass-transport and mechanical, so-called non-linear processes that control concrete degradation and crack development were investigated by coupled numerical models. Application of such coupled numerical models allows identification of the dominant non-linear processes that will control long-term concrete degradation and crack development in a TRU waste repository.

1. Introduction

Deep geological disposal of transuranic (TRU) wastes in Japan will be in an engineered repository consisting of large diameter (> 10 m) disposal tunnels located at depths of at least 300 m underground. Construction and operation of the repository requires that the tunnel support, liner, invert, and backfill will use ordinary Portland cement (OPC) based concrete (Fig. 1) (JAEA and FEPC 2007). Concrete is considered a suitable material to ensure safety by providing structural integrity to the repository, impedes percolating groundwaters, slows down steel corrosion, and helps to retard radionuclide migration by sorption after the waste containers fail (NUMO 2019).

Current safety assessments for the disposal of TRU waste in Japan (JAEA and FEPC 2007; NUMO 2019) conceptualize the concrete in the disposal tunnels as being a homogeneous and continuous material with concrete degradation and crack development proceeding as a large, one-step failure event causing a rapid increase in groundwater flow and uninhibited radionuclide migration. This simple treatment of concrete in safety assessment is thought to be a conservative (i.e. worst case) scenario and provides an empirical means to evaluate other chemistry and mass transport processes relevant to radionuclide migration.

A more realistic conceptualization, however, is that coupled changes in chemical reaction, mass transport

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Fig. 1 Cross-section of a tunnel designed for geological disposal of TRU waste in Japan based on descriptions given in JAEA and FEPC (2007). Dimensions in meters. OPC based concrete is used for the backfill, invert, liner, and support.

*: Wastes in this design are used for the backfill, invert, liner, and support.
†: The tunnel support is reinforced by steel arched supports.
and mechanical (hereafter collectively referred to as non-linear) processes will control more gradual changes in concrete degradation and crack development, causing the concrete to become an anisotropic and discontinuous material that can either increase or decrease groundwater flow and promote or inhibit radionuclide migration. Despite this realism, coupled changes in non-linear processes quickly become complicated, because they can operate on different spatial and temporal scales, contain numerous positive and negative feedback loops and will continue to evolve over the lifetime of the repository. This combination of features makes it very difficult to identify which of the non-linear processes will ultimately control concrete degradation and crack development under a given set of service life conditions by expert judgement alone. For example, decalcification caused by the dissolution of the cement hydrate minerals and expansive reactions due to precipitation of secondary minerals and corrosion of steel components can all contribute to crack development in concrete (Okatsu et al. 2005; Ferraris et al. 1997; Hirano et al. 2016; Babaahmadi et al. 2015; JAEA 2018). Once established, groundwater flowing into the crack could either cause further decalcification thereby widening the crack and reducing the strength of the concrete (Höglund 2014) or else cause the precipitation of secondary minerals resulting in clogging and/or densification, which could lead to an increase in the strength of the concrete (Babaahmadi et al. 2015; Metcalfe et al. 2017). A more realistic conceptualization of concrete in the disposal tunnels therefore requires a new analytical approach based on numerical models that can quantify and so help identify the non-linear processes that contribute to concrete degradation and crack development.

In developing a new analytical approach, Benbow et al. (2020) demonstrated that coupling of non-linear processes could be achieved by coupling of the computer programs MACBECE (JAEA 2018; Ohno et al. 2009; Mihara et al. 2017), DEAFRAP (JAEA 2018; Shimizu et al. 2011) and GARFIELD-CHEM (Benbow et al. 2020) as shown schematically in Fig. 2. MACBECE implements a finite element method (FEM) model of the macro scale stress and deformation in the disposal tunnel caused by host rock creep and steel component corrosion and expansion. It uses a rotating crack model, which simulates the behaviour of the element containing the crack as a continuum. DEAFRAP uses the discrete element method (DEM) to model the micro scale emergence of cracks in regions where cracking is most likely to occur as identified by MACBECE (labelled “cracking area” in Fig 2[a]). DEAFRAP can represent the microscale structural features of materials directly, such as construction defects, pores and micro-cracks, thereby macroscopic behaviours such as crack generation, propagation, separation, and merging can all be analysed. Both MACBECE and DEAFRAP can be combined with the GARFIELD-CHEM reactive transport analysis tool to allow key inputs to be parameterized in terms of the evolving porewater and solid phase chemistry. GARFIELD-CHEM simulates the fully coupled porewater and solid phases chemistry on a finite element grid that can be automatically refined to produce a two dimensional (2D), multi-scale and fully coupled model corresponding to the extracted crack geometry and location as identified by DEAFRAP. GARFIELD-CHEM also uses the crack information in a separate hydraulic analysis to derive the advective velocity of groundwater flowing through the evolving crack network. Data-exchanges between these different computer programs in the context of a waste disposal tunnel are summarized in Fig. 2. Further technical details can be found in Benbow et al. (2020).

There are, however, a number of known issues relevant to coupling non-linear processes. Firstly, reactive transport simulations have problems in properly resolv-
ing concentration gradients in space and time, which cause changes in porosity (Marty et al. 2009). These gradients are influenced by transport processes and kinetic control of dissolution/precipitation processes. Discretization of the system therefore needs to resolve the concentration gradients in order to get the correct porosity change. For equilibrium reactions, “grid convergence” is not possible with continuum scale numerical methods (Hayek et al. 2011). For consideration of kinetic reactions, discretization on the mm scale has been identified as a necessary requirement for the numerical solution to agree with the analytical solution (Hayek et al. 2011), whereas discretization on the cm scale is generally used in long-term reactive-transport simulations to save computation time (Benbow et al. 2020; Kosakowski and Berner 2013; Trotignon et al. 2007; Mon et al. 2017). Previous experimental studies, for example, those by Sarott et al. (1992), Pfingsten (2001) and Nakarai et al. (2021), have found that thin surface layers of calcite can significantly reduce cement reactivity and mass transport properties due to pore-clogging at the outer cement surface. The significance of these surface layers on system evolution may be underestimated or could well be missed altogether if the cell size was on the scale of several centimetres. This discretization problem is particularly pertinent to the 2D reactive-transport simulations in GARFIELD-CHEM and by coupling, so too, to the entire analytical approach. Benbow et al. (2020) conservatively assumed that no mineral reactions took place in the cracks so that the cracks remained open and fully transmissive. Clogging of the cracks by the precipitation of secondary minerals is potentially very significant (Guppy 1998; Cao, et al. 2015; Brunet et al. 2016) and so it is important to demonstrate the consequences of this non-linear process with the use of a detailed numerical model.

In the current study, the computer programs MACBECE, DEAFRAP and GARFIELD-CHEM developed and coupled by Benbow et al. (2020) were extended to enable transient changes in hydraulic and transport properties due to effects of mineral precipitation and dissolution in the crack to be calculated. By so doing, the current study provides a demonstration of the possible coupling of non-linear processes that control concrete degradation and crack development in a TRU waste repository.

2. Numerical modelling

In the coupled non-linear process model analysis, DEAFRAP and GARFIELD-CHEM were “tightly” coupled (two-way data exchanges), and MACBECE was ‘loosely’ linked (one-way data flow) to DEAFRAP (Fig. 3). In the mechanical model built in DEAFRAP, the non-linear processes of “reduction of strength and stiffness”, “deformation/stress redistribution” and “cracking/crack propagation” were included. In the 2D reactive-transport model of GARFIELD-CHEM, coupling between “mineral dissolution”, “secondary mineral precipitation”, and subsequent “change in porewater chemistry”, “porosity increase”, “matrix diffusion”, “crack clogging” and “groundwater flow” were included. The non-linear processes of “expansion” by “steel corrosion” and resultant “deformation/stress redistribution” were modelled in MACBECE.

The MACBECE model used in this study was previously used for mechanical analysis of the TRU geological disposal tunnel (JAEA 2013) and was allowed to run for up to 100 000 years. OPC based concrete (the mass percentage of water to cement, w/c = 45%) was assumed to be used for the invert, liner, and support. Creep of the host rock and corrosive expansion of the 200 dm³ steel drums (0.0016 m thick), steel packages (0.1 m thick) and steel arched supports (0.009 m thick) in the concrete tunnel support were all considered. Bentonite buffer was assumed instead of a concrete backfill and applied with an elasto-viscoplastic constitutive model proposed by Ohno et al. (2006) as used previously (JAEA 2013). The bentonite buffer model was left unchanged and used directly in the MACBECE model.

Fig. 3 Non-linear processes and their coupled interactions related to concrete degradation and crack development. Black arrows are used to denote promotion and red arrows denote inhibition.

Note: Coupling between “change in porewater chemistry” and “steel corrosion” was not included in the current study.
in this study (further details are provided in the appendix). The finite element grid used in the MACBECE stress analysis model of the TRU disposal tunnel is shown in Fig. 4. To represent the evolving stresses and displacements in the tunnel system it was necessary to include the internal structures in the waste form and package and to include a large region of host rock surrounding the tunnel. A combination of quadrilateral and triangular elements was used in the finite element model, with a total number of 11 256 finite elements and 11 112 nodes.

After a simulation time of 40 years in MACBECE, corresponding to the concrete becoming saturated with groundwater, one analytical element (No. 4710, 0.09 × 0.10 m) (Fig. 5) was identified as a region in which a crack may develop. This analytical element was subsequently used as an analytical element of the DEM sub-grid model in DEAFRAP. Intact cementitious materials were modelled in DEAFAP as densely packed small rigid circular particles. Neighbouring particles were bonded together at their contact points by three types of springs (normal, tangential and rotational springs). The normal and shear stress acting on the cross-section of the bond were calculated by the forces exerted by respective springs and the area of the bond cross-section. The details of the formulations of mechanical behaviour in DEAFRAP can be found in references (JAEA 2018; Shimizu et al. 2011, 2018). Mechanical properties used in the MACBECE and DEAFRAP models are shown in the appendix. OPC based concrete (w/c = 45%) was used for the invert, liner and support. Steel components were defined as corrosion expansion elements.

\[
\sigma_c = \sigma_{ch} \cdot R_{cp}
\]

\[
R_{cp} = \exp \left[ \frac{100 \cdot \theta_p - b}{100 \cdot a} \cdot \ln \left( \frac{\sigma_{c(mm)}}{\sigma_{ch}} \right) \right]
\]

\[
a = 1.038 \cdot \exp \left[ -0.0135 \cdot \frac{W}{C} \right]
\]

\[
b = 36.41 \cdot \ln \left( \frac{W}{C} \right) - 114.72
\]

Fig. 4 Finite element grid in MACBECE. [a] Tunnel and host-rock. [b] Tunnel, [c] Liner and support, and [d] Waste. OPC based concrete (w/c = 45%) was used for the invert, liner and support. Steel components were defined as corrosion expansion elements.
\[
\theta_p = \frac{f(\theta_{chem}) - \varepsilon_c}{1 - V_c - \varepsilon_c} \tag{5}
\]

\[f(\theta_{chem}) = 1.3277 \cdot \theta_{chem} - 0.094167, \text{if } \theta_{chem} \geq 0.13 \tag{6}\]

\[f(\theta_{chem}) = 0.6018 \cdot \theta_{chem}, \text{if } \theta_{chem} < 0.13 \tag{7}\]

\[\sigma_i = \sigma_{i0} \cdot R_{\sigma} \tag{8}\]

\[R_{\sigma} = \exp \left[ 100 \cdot \frac{\theta_p - b}{100 \cdot a} \ln \left( \frac{\sigma_{(\text{ult})}}{\sigma_{i0}} \right) \right] \tag{9}\]

\[E = 21000 \cdot \left( \frac{\gamma}{2.3} \right) \cdot \left( \frac{\sigma_c}{20} \right)^{0.5} \tag{10}\]

where \(\sigma_c\) is the compressive strength (MPa), \(\sigma_{i0}\) is the

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**Fig. 5**

[a] Continuum scale FEM model (analytical element No. 4710 dimensions = 0.09 x 0.10 m), [b] The corresponding finite element grid in MACBECE, [c] Sub-grid scale DEM model concrete in DEAFRAP, and [d] Enlarged view of sub-grid scale DEM model concrete singled out for coupled analysis combined with GARFIELD-CHEM. Dashed lines shown in the liner and support in [b] delimit the data output range for DEM analysis. Red arrows shown in [c] denote the direction of load to reproduce a same stress condition calculated by MACBECE.

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**Fig. 6** Data exchanges. \(D_{\text{ref}}(x,t)\) is the effective diffusion coefficient \((m^2/s)\) at location \(x\) (m) and time \(t\) (s) as calculated from the in-built diffusion model, \(\theta_{chem}(x,t)\) is the chemically altered porosity (-), \(C_i(x,t)/C_{i0}\) is the leaching ratio (-) where \(C_i(x,t)\) is the concentration of the \(i\)th mineral in the simulation (mol/kg) and \(C_{i0}\) is the initial concentration of the \(i\)th mineral (mol/kg). Note that a user-defined volumetric flux for the cells containing crack on the analytical element of GARFIELD-CHEM was needed (see text).
Table 1: Model concrete components and their thermodynamic and kinetic properties as used in GARFIELD-CHEM.

| Component | Composition | Conc. (mol/m³) * | Vol. (%) | Log K † | Mol. Vol. (cm³/mol) | k (mol/m²/s) | $A_0$ (m²/g) |
|-----------|-------------|------------------|----------|---------|---------------------|-------------|-------------|
| Portlandite | Ca(OH)₂ | 4189.35 | 13.85 | 22.790 | 33.056 | 1 × 10⁻⁶ | 0.22 |
| Jennite (gel) | Ca₁₆Si₉O₂₅(OH)₆(H₂O)₆ | 1326.53 | 10.40 | 29.042 | 78.4 | 1 × 10⁻⁷.⁵ | 0.22 |
| Tobermorite-14A | Ca₅Si₆O₁₆(OH)₄(H₂O)₈ | 187.39 | 6.98 | 63.845 | 372.555 | 1 × 10⁻⁷.⁵ | 0.22 |
| Calcite ‡ | CaCO₃ | 0 | 0 | 1.849 | 36.934 | 3.3 × 10⁻³ | 0.22 |
| Aggregate § | - | 24577.04 | 55.76 | - | 22.688 | - | - |
| Porosity | - | - | - | - | - | - | - |

*Based on a model concrete (JAEA and FEPC, 2007) assuming 100% hydration of OPC at w/c = 45%.
1Log K values correspond to mass and charge balanced dissolution reactions of components at 25°C, written in terms of H₂O, H⁺, Ca²⁺, H₄SiO₄(aq), and/or HCO₃⁻ as necessary.
‡Calcite was a secondary mineral.
§Aggregate was assumed to be chemically inert (see text), and was assigned the same molar volume of quartz = 22.688 cm³/mol as a substitute and concentration to provide the same initial porosity = 13.01% as that modelled in JAEA and FEPC (2007).

Table 2: Groundwater composition used in GARFIELD-CHEM.

| Component * | pH | Ca | C | Si | Cl † |
|-------------|----|----|---|----|------|
| Value       | 8.5 | 1 × 10⁻⁶ | 3.54 × 10⁻⁴ | 3.39 × 10⁻⁴ | 1 × 10⁻⁶ |

*Element concentrations given in units of mol/kg. †Cl used as a tracer element.

Initial compressive strength (MPa), $\sigma_{c(min)}$ is the minimum compressive strength (MPa), $W/C$ is the mass percentage of water to cement (%), $\theta_p$ is the porosity of cement paste (-) (i.e. volume contributions from aggregates are not included), $\theta_{chem}$ is the chemically altered porosity of the concrete (-), $V_c$ is the volume ratio of aggregates to concrete (-), $c_v$ is the volumetric strain (-), $\sigma_c$ is the tensile strength (MPa), $\sigma_{io}$ is the initial tensile strength (MPa), $\sigma_{e(min)}$ is the minimum tensile strength (MPa), $E$ is the Young’s modulus (MPa), and $\gamma$ is the unit volume weight in the air-dried state (Mg/m³).

Subsequently, the tensile stress or the shear stress acting on the cross-section of the bond between particles could be calculated. When the tensile stress or the shear stress exceeded the tensile strength or the shear strength, the bond was defined to break and individual bond breakage between bonded particles was defined as a micro-crack. A crack pathway was defined when neighbouring micro-cracks connected with each other to form a larger crack. This was not a one-way process, however, as particles could be reconnected when the value of porosity reduction reached 0.1 (= 10%), on the premise that the crack could be repaired by secondary mineral precipitation. DEAFRAP provided details of timing and locations of the crack pathway that appeared in the analytical element and of crack growth to GARFIELD-CHEM every second $t = 125$-year step, after which the media grid was adaptively refined in GARFIELD-CHEM to resolve the specific locations of the cracks at greater resolution. Technical details of the methods of extraction of continuous micro-cracks from the bond breakage data, and conversion of the continuous porosity distribution of GARFIELD-CHEM into the parameters of the DEM in DEAFRAP can be found in Benbow et al. (2020).

Initial concrete porewater compositions can be calculated by assuming equilibrium between groundwater and cement hydrate minerals. Using all of the possible cement hydrate minerals and groundwater composition elements was found to be computationally expensive and so a simplified set of model components belonging to the CaO-SiO₂-CO₂-HCl system were selected (Tables 1 and 2). Aggregate was assumed to be chemically inert as non-reactive aggregate would be selected to minimize the likelihood of alkali silica reaction. This simplified selection was based on a model concrete assuming that hydration of OPC was 100% complete (JAEA and FEPC 2007), which was used with a representative model groundwater (Yui et al. 1999; Sasamoto et al. 1999) at w/c = 45%. Tobermorite was included in the initial composition to check that the GARFIED-CHEM program would recognize this as a metastable phase and consequently react the tobermorite with portlandite to form more jennite. The initial concentration of Ca = 1 × 10⁻⁶ mol/kg in the concrete porewater was purposefully set to promote dissolution.

Dissolution/precipitation reactions of all minerals, also including calcite as a secondary mineral, were simulated kinetically with Eq. (11) (Steefel and Lichtner 1998) using the parameters shown in Table 1:

$$r = A \cdot k \cdot \left(1 - \frac{IAP}{K}\right)$$

(11)

where $A$ is the reactive surface area per unit volume (m²/m³), $k$ is the rate constant (mol/m²/s), $IAP$ is the ion activity product (-), and $K$ is the equilibrium con-
stant for the mineral of interest (-). Inclusion of a pH dependent term in Eq. (11) was not used in the current study owing to a lack of reliable data. Values of $k$ in Eq. (11) were compiled from the literature (Palandri and Kharaka 2004; Brantley et al. 2008) (Table 1). Activities of aqueous species to calculate $IAP$ in Eq. (11) were determined for charged aqueous species using activity coefficients calculated from the B-Dot extended Debye-Hückel equation (Helgeson 1969) and activity coefficients of neutral species and water were assumed to be unity. Log $K$ values at 25°C for all chemical reactions and the molar volumes for minerals were taken from the Japan Atomic Energy Agency (JAEA) thermodynamic database (TDB) for geochemical reactions, JAEA-TDB (v1.07) (JAEA 2017). Values of $A$ in Eq. (11) were assumed to vary as a function of porosity in a porous medium according to (Steeef and Lichtner 1998):

$$A = A_0 \left( \frac{\theta_{\text{chem}}}{\theta_{\text{chem0}}} \right)^{2/3}$$  \hspace{1cm} (12)

where $A_0$ is the initial reactive surface area ($m^2/m^3$) (Table 1), $\theta_{\text{chem}}$ is the chemically altered porosity of the concrete (-), $\theta_{\text{chem0}} = 0.1301$ is the initial porosity of the concrete (-), and the exponent of $2/3$ is used to describe minerals that have either spherical or cubic morphologies.

No flux (Neumann) boundary conditions were applied to the analytical element in GARFIELD-CHEM (Fig. 7). The same dimensions of $0.09 \times 0.10$ m as used for the analytical element in the continuum scale FEM model (Fig. 5) was divided into $10 \times 10$ geometrically equivalent cells at the starting time $t = 0$. Solute transport was assumed to be controlled by in-plane 2D diffusion over the entire analytical element and perpendicular to plane 1D advective flow in the “crack cells” intersected by the crack pathway (Fig. 7) as identified by DEA-FRAP. Diffusive transport was described by Fick’s law and the effective diffusion coefficient, $D_{\text{eff}}(x,t)$ ($m^2/s$) was assumed to vary as a function of porosity, $\theta_{\text{chem}}$, and tortuosity, $(\theta_{\text{chem}})^{\alpha}$ (Archie 1942):

$$D_{\text{eff}}(x,t) = D_{\text{eff}}(x,0) \left( \frac{\theta_{\text{chem}}}{\theta_{\text{chem0}}} \right)^{w}$$  \hspace{1cm} (13)

where $w$ (-) is a cementation factor. Both $w$ and $D_{\text{eff}}(x,0)$ were here assumed to be $1$ and $1 \times 10^{-10}$ ($m^2/s$), respectively, which resulted in a higher diffusivity.

The perpendicular to plane 1D advective flow velocity, $q$ ($m/s$) was calculated on the assumption that the volumetric flux through the crack was equal to the flux over the region of the crack cell:

$$q = \frac{w_{\text{cell}}}{w_{\text{crack}}} \cdot i \cdot k_{\text{eff}}(\theta_{\text{chem}})$$  \hspace{1cm} (14)

where $w_{\text{cell}}$ is the cell size ($m$), $w_{\text{crack}}$ is the crack width ($m$), $i$ is the hydraulic gradient (m/m), and $k_{\text{eff}}(\theta_{\text{chem}})$ is the hydraulic conductivity ($m/s$) of the crack cell, as defined by:

$$k_{\text{eff}}(\theta_{\text{chem}}) = k_{\text{eff,d}} \left( \frac{1-\theta_{\text{chem}}}{\theta_{\text{h}}} \right)^{3/2} \frac{\theta_{\text{chem}}^{3}}{(1-\theta_{\text{chem}})^{3}}$$  \hspace{1cm} (15)

where $k_{\text{eff,d}}$ is the initial hydraulic coefficient of the crack, hypothetically taken to be $1.3 \times 10^{-8}$ ($m/s$), $\theta_{h}$ is the initial porosity (-) of the crack cell and the last term is adopted from the Kozeny-Carman equation (Kozeny 1927; Carman 1956) to account for changes in porosity during the course of the simulation.

3. Results

GARFIELD-CHEM correctly predicted the reaction of tobermorite with portlandite to form more jennite. This change occurred within the first 0.06 years of the simulation and caused an increase in porosity from 13.1 to 14.2%. Although slight, it is important to ensure that cement/concrete degradation models use mineral assemblages that are initially at equilibrium.

Distributions of crack widths calculated by DEA-FRAP are shown in Figs. 8[a] to 8[c], and distributions of porosities given to DEM particles based on outputs from GARFIELD-CHEM are shown in Figs. 8[d] to 8[f]. After 3500 years (Figs. 8[a] and 8[d]), it can be seen that fine cracks with widths of several microns
have started to appear in the upper left area of the analytical element (Fig. 8[a]). GARFIELD-CHEM adaptively refined the grid at 3500 years for the crack information provided from DEAFRAP. GARFIELD-CHEM then simulated advective transport in the direction perpendicular to the grid up to 3625 years. In the analysis, only the cracks with widths $\geq 10$ microns were identified and the minimum cell size after refinement was set to 0.002 m (i.e. 200 times larger than the crack width). DEAFRAP proceeded to perform the analysis from 3500 to 3650 years reading the porosity distribution data during this period provided from GARFIELD-CHEM.

At 3750 years (Figs. 8[b] and 8[e]), cracks have grown on the upper left and lower right areas of the analytical element and associated porosities have changed in and adjacent to the cracks. A decrease in porosity in the crack cells arises due to the precipitation of the secondary mineral calcite (Fig. 9[e]). Calcite precipitation was caused by the reaction of calcium released from the concrete matrix by the dissolution of portlandite with
the carbonate contained in the groundwater flowing into the crack. Consequently, the potential for the porosity to increase further by the continued dissolution and loss of portlandite (Fig. 9[b]) and jennite was compensated by calcite precipitation. At the same time, an increase in porosity was observed by the localized dissolution of portlandite around the cracks (Fig. 9[b]).

The location of the micro cracks caused by either tension or shearing between DEM particles, and the location of re-connected cracks, superimposed on the distribution of the porosity given to the DEM particles after 3750 years, are shown in Fig. 10. It can be seen from this figure that newly formed cracks have occurred in regions of increased porosity, which surround regions of lower porosity where cracks had been re-connected. Cracks continued to develop in a similar manner up to 4000 years (Figs. 8[c], 8[f], 9[c] and 9[f]).

4. Discussion

The coupled model analysis applied to the 0.09 × 0.10 m analytical element in the concrete liner of TRU waste disposal tunnel showed the complex nature of crack development relating changes in tensile and shearing stress, porosity and dissolution/precipitation reactions. Cracks once generated in the concrete started to close by secondary mineral precipitation and adjacent subsurface demineralization caused an increase in the porosity,
which marked the location of newly formed cracks by tensile and shearing stresses. The porosity was predicted to increase by up to 20% by the demineralization of portlandite at the subsurface adjacent to the crack (Figs. 8[c] and 8[f]). Porosity increase by localized decalcification have been observed by experimental studies such as the one by Kurumisawa et al. (2021). Using the empirical relations between the strength of OPC and porosity as in Eqs. (1) to (10) (Okutsu et al. 2005; JAEA 2018), the tensile strength decreased to 10% of the initial value when the concrete porosity increased to 20%. Such increases in the porosity are indicated to have been therefore weak points, which were in mechanically unstable states that lead to failure and crack development. When a crack develops, continued mineral dissolution would increase porosity thereby lowering mechanical strength and promoting further crack development. This emphasizes the necessity of modelling coupled nonlinear processes in considering not only changes in porosity, but also their subsequent effect on the mechanical properties of OPC.

The relationship between the stiffness and the strength with the porosity was based on experimental data obtained under the prevailing conditions of porosities increasing due to mineral dissolution (Okutsu et al. 2005). However, this scenario is not consistent with the simulated results where porosities decreased due to the precipitation of calcite. This means that if the relations between the stiffness and strength with the porosity are extrapolated then they may not be accurate. As previously noted, an increase in compressive strength can be observed if the porosity becomes filled by the precipitation of secondary minerals (Babaahmadi et al. 2015). However, it is not well known whether the decrease in strength caused by the coupled demineralization of primary cement hydrate minerals and cracking can ever fully recover by the precipitation of secondary minerals, raising doubts over the self-healing ability of cracked concrete. Moreover, to what extent this is affected by the type of secondary minerals is not well known. In the simple CaO-SiO$_2$-CO$_2$-HCl system used in the current study the secondary mineral was calcite, but in other more complicated systems calcium (aluminate) silicate hydrate (C(-A)-S-H) gel, ettringite, thaumasite, Kuzel’s salt or Friedel’s salt may appear as secondary minerals depending on the composition of the inflowing groundwater. Such uncertainties in the strength effects of cracks being self-healed by different secondary minerals needs to be addressed in the future.

Feedback of the porosity data from GARFIELD-CHEM to MACBEC and subsequent feedback of the deformation data from MACBEC to DEAFRAP was not included in the current study. Feedback from “change in porewater chemistry” in GARFIELD-CHEM to “steel corrosion” and subsequently to “expansion” due to “steel corrosion” in MACBEC (Fig. 3) was also not included in this study (i.e. “expansion” due to “steel corrosion” was taken to be constant), which would affect the crack generation/propagation in concrete. It is expected that future advances in computer hardware will make it possible to perform the calculations at higher specification, including these missing couplings, thereby allowing more realistic evaluation of the long-term evolution of the entire tunnel scale system to be made.

5. Conclusions

Concrete used in the construction of a repository for the geological disposal of TRU waste is conventionally treated as a homogenous and continuous material where cracking causes a significant, one-step increase in groundwater flow and change in radionuclide transport. A more realistic conceptualization is that coupled changes in chemical, mass-transport and mechanical processes will control gradual changes in concrete degradation and crack development, causing the concrete to become an anisotropic and discontinuous material that can either increase or decrease groundwater flow and promote or inhibit radionuclide migration. For instance, clogging of the crack by the precipitation of secondary minerals is potentially very significant in terms of self-healing (Guppy 1998; Cao, et al. 2015; Brunet et al. 2016). In the current study, the coupled numerical models recently developed by Benbow et al. (2020) were extended to include transient changes in hydraulic and transport properties due to effects of mineral precipitation and dissolution in the crack. An analytical element in the concrete liner of a TRU waste disposal tunnel was
simulated using the extended coupled models implementing the coupling processes of stress, crack formation, solute transport, solid-solute reaction, and porosity change at sub-grid scale. The simulation showed that cracks once generated in the concrete started to close by secondary mineral precipitation and adjacent subsurface demineralization caused an increase in the porosity, which marked the location of newly formed cracks by tensile and shearing stresses. This provides an example of a feedback loop between mechanical and reactive-transport processes, which can be predicted by the analytical approach demonstrated in the current study. Validating concrete degradation and crack development in a real OPC concrete repository for TRU waste is not straightforward. A large number of model parameters are required, which themselves contain a large number of uncertainties. Using a coupled numerical models approach will make it possible to simulate the behaviour of the concrete repository more realistically and provide new insights in understanding how these coupled nonlinear processes can control the evolution of an OPC concrete repository for TRU waste. It will also permit an assessment of the impact of different model parameters, determine which are important and if their uncertainties are significant. Furthermore, identification of dominant coupled non-linear processes, both known and currently unknown, allows research efforts to be streamlined and improve confidence in the reliability of the numerical model predictions. While this study demonstrates a "proof of concept" of such an approach, the goal now is to make it work on the larger scale and consider sub-grid model results everywhere in the whole scale continuum model.

Acknowledgements
This study was funded by the Ministry of Economy, Trade and Industry of Japan through "The project for validating assessment methodology in geological disposal system" in Financial Year 2016. The authors are grateful to Dr. Colin S. Walker (QJ Science) for his assistance in writing the paper and to the two anonymous reviewers whose insightful comments helped to improve the quality of this manuscript.

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Appendix
Mechanical properties of cement components (ordinary Portlandite cement, w/c 45%) and metal materials used in MACBECE (JAEA 2018; Ohno et al. 2009; Mihara et al. 2017) and DEAFRAP (JAEA 2018; Shimizu et al. 2011) are shown in Table A1. For bentonite buffer, MACBECE uses the elasto-viscoplastic constitutive model proposed by Ohno et al. (2006). The mechanical properties of the bentonite buffer (bentonite 70 wt%, silica sand 30 wt%, dry density 1.6 Mg/m$^3$) used in MACBECE are from the work of Hirano et al. (2016).

Table A1 Mechanical properties used in MACBECE and DEAFRAP.

| Properties for cement components | Inner / Invert / Backfill (concrete) | Waste (mortar) |
|----------------------------------|-------------------------------------|----------------|
| Initial Young’s modulus, $E_0$ (MPa) | $3.04 \times 10^4$ | $1.75 \times 10^4$ |
| Poisson ratio, $v$ (-) [before yield] | 0.20 | 0.20 |
| Poisson ratio, $v$ (-) [after yield] | 0.45 | 0.45 |
| Effective overburden pressure (MPa) | Automatically calculated by self-weight loading |  |
| Coefficient of earth pressure at rest $K_i$ (-) | Automatically calculated by self-weight loading |  |
| Initial elastic shear modulus, $G_0$ (MPa) | $G_0 = \frac{E_0}{2(1+v)}$ |  |
| Initial adhesive force (MPa) | 21.5 | 17.5 |
| Internal frictional angle (deg.) | 0 | 0 |
| Initial compressive strength, $\sigma_{c0}$ (MPa) | 43 | 35 |
| Minimum compressive strength, $\sigma_{c(min)}$ (MPa) | 0.43 | 0.35 |
| Air dried weight of unit volume, $\gamma$ (kN/m$^3$) | 22.8 | 20.9 |
| Initial tensile strength, $\sigma_{t0}$ (MPa) | 4.3 | 3.5 |
| Minimum tensile strength, $\sigma_{t(min)}$ (MPa) | 0.043 | 0.035 |
| Rigidity reduction ratio after cracking (-) | 1/100 | 1/100 |
| Kinematic viscosity of water (m$^2$/s) | $1.0 \times 10^6$ | $1.0 \times 10^6$ |
| Number of cracks per element | 1.0 | 1.0 |

Properties for metal materials

| Properties for metal materials | Waste (Steel package, drum can and steel arched support) |
|-------------------------------|--------------------------------------------------------|
| Corrosion expansion ratio (-) | 3.2 | - |
| Rigidly (sound region) (MPa)  | 211,400 | - |
| Rigidly (corroded region) (MPa) | 250 | - |
| Poisson ratio (-) | 0.333 | - |

Properties for host-rock

| Properties for host-rock | Rock | - |
|--------------------------|------|---|
| Unit weight (kN/m$^3$) | 22.0 | - |
| Uniaxial compressive strength (MPa) | 15.0 | - |
| Tensile strength (MPa) | 2.1 | - |
| Initial Young’s modulus (MPa) | $3.5 \times 10^3$ | - |
| Initial Poisson ratio (-) | 0.30 | - |
| Initial overburden pressure (MPa) | 11.0 | - |
| Lateral pressure coefficient (-) | 1.066 | - |