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Leaching resistance of hazardous waste cement solidification after accelerated carbonation

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Abstract: When cement-based materials are carbonated, some of their physicochemical properties are changed, which includes reductions of porosity by 20% and pH from 12-13 to 8-9. These changes can enhance the retention ability of cementitious solids containing hazard waste. This research studied the effect of carbonation on the leaching resistance of hazardous waste cement solidification. The finite element software COMSOL Multiphysics was used to simulate the process of accelerated carbonation and the effect of carbonation on leaching. Laboratory tests were conducted to validate the numerical models. Parametric studies from the numerical simulations revealed that carbonation could significantly improve leaching retention capabilities of cementitious solids containing hazardous wastes.

Keywords: Hazardous waste cement solidification, Accelerated carbonation, COMSOL Multiphysics simulation, Particle leaching test, Leaching rate and cumulative leaching rate.

1. Introduction

With the rapid development of human society, the amount of hazardous waste produced from our daily life has increased significantly. Since hazardous waste, among which nuclide waste represents a significant fraction, poses potential threats to both public health and environment, it must be safely disposed. Over the last few decades, more and more researchers have been working in this area, trying to find better solutions. Due to political, economic or military reasons, the number and scale of nuclear reactors have also increased rapidly worldwide over the years. In France or some other developed countries, nuclear power produces 50% or more of their total electricity, concurrently producing a large amount of nuclear waste that needs to be disposed safely [1]. Nuclear waste contains a large amount of hazardous substances, most of which are in a liquefied state. Before the final disposal, they must be solidified or immobilized. The cement solidification method has become a commonly-used method to deal with nuclear waste because the process is simple, technically proven and has good stability. However, since cement is porous, the retention capacity for the internal particles needs to be improved [2]. One of the practical approaches to improve this capacity is through a carbonation process to consolidate further the cement. This is because carbonation will change the physicochemical properties of cement solids, such as producing reductions of pH from alkaline to neutral and porosity by more than 20%. The process also increases the strength of the cement solids. All of the above will affect the curing properties of solidification

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[3].

There have been some applications and research on waste or sludge cement solidification treated by carbonization technology. Fernandezbertos et al. [4] published a review on accelerated carbonation for improving properties of cement-based materials. Guning et al.[5] demonstrated that accelerated carbonation could enhance the curing property of cement solidification and reduce the cost of disposal processes. Shen et al. [6] studied carbonation of cementitious materials in CO_2 geological storage conditions and published a study on particle exchange and porosity reduction during carbonation. Other researchers showed that accelerated carbonation could increase the impermeability and chemical consolidation of radionuclide solidification. This technology has been used in the production of cement-based materials in the United States [7].

Most of the above investigations were based on experimental studies. Since leaching tests are complex and time consuming, a real scenario test is normally not possible. Naturally, numerical simulations can be used as an alternative tool to assess the properties of cement solidification. A number of numerical models have been developed for heavy metal particles and radionuclides leaching from porous media. Suarez et al. [10] developed a one-dimensional model to predict leaching and pointed out that the effective diffusion coefficient was not constant during the process. Batchelor [11] developed a theoretical leaching model of solidified waste, but its actual application was limited by the lack of measured values for certain parameters. Birdsell et al. [12] developed a leaching model to support groundwater pathway analysis of low-level solidified radionuclides. Kamash et al. [13] developed a model of radionuclides leaching out from cement-based materials and verified the model through experiments. It was found that all these models were based on simplified theories that ignored many influential factors, and were almost exclusively of either a one- or two-dimensional spatial domain that inevitably had limitations and resulted in inaccurate solutions for a complex leaching process. Moreover, none of these models considered the effect of carbonation on the leaching process. Due to the above limitations, it is not possible to apply any of these models to simulate the leaching tests currently specified by industrial standards, such as GB7023-2011 [16].

This paper attempts to develop a three-dimensional leaching model, coupled with an existing carbonation model as proposed by Zha et al.[17], to accurately simulate cement solidification leaching tests. The simulations include the processes before and after accelerated carbonation, and are validated by carbonation experiments carried out also by the authors.

2. The theoretical basis of modeling

2.1 The leaching model

The leaching model is established according to the conservation of heat and mass. The flow of the hazardous particles is driven by concentration and temperature gradients. There are three main kinds of particle diffusion resistance: chemical fixation, mechanical seal and physical adsorption.

2.1.1 Coupled heat and mass transfer model

According to the laws of heat flow and the principle of energy conservation, and by ignoring the influence of particle movement on energy [18], the energy conservation equation of a cement material system can be written as:

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$$\rho c\frac{\partial \mathrm{T}}{\partial \mathrm{t}} = \nabla \cdot [\lambda \nabla \mathrm{T}] \tag{1}$$

where ρc is the heat capacity and λ is the coefficient of thermal conductivity.

In the same way, one can deduce the equation of mass conservation shown as below:

$$\nabla \cdot [\rho D_T \nabla T + \rho D \nabla C] - \lambda' C + F = \rho \frac{\partial C}{\partial t} \tag{2}$$

where ρ is the density of material; D_T is the thermal diffusion coefficient; D is the effective diffusion coefficient; λ' denotes a decay constant and $\lambda' = \ln 2/T_h$; T_h is the half-life period of the radionuclide; C is the nuclide mass per unit volume (g/cm^3), which is a function of position coordinates and time; $C = f(x, y, z, t)$; and F is the quality of adsorption.

Gawin [19] proposed the following equations, respectively, for effective heat capacity and effective thermal conductivity coefficients:

$$\rho c = (1-n)\rho_s c_s + nS_w \rho_w c_w + nS_g \rho_g c_g \tag{3}$$

$$\lambda = \lambda_0 [1 + A_\lambda (T - T_r)] \left(1 + 4\frac{nS_w \rho_w}{(1-n)\rho_s}\right) \tag{4}$$

where ρc is the effective heat capacity; λ is the effective thermal conductivity coefficient; ρ_w is the density of pore solution; ρ_g is the density of gas in the pores; ρ_s is the density of the solid skeleton; c_w is the heat capacity of pore solution; c_g is the heat capacity of gas in the pores; c_s is the heat capacity of the skeleton; S_w and S_g are the saturation of liquid and gas, respectively. In the simulation, S_g ranges from 0 to 1 for modeling carbonation and takes 1 for modelling leaching, where $S_g + S_w = 1$ is always satidied; λ_0 and A_λ are the fitting parameters taking 1.67 W/(m·s) and 0.0005 K^{-1}, respectively[19]. T_r is the room temperature and is taken as 298.15 K; and D_T takes a value of 4×10^{-11} cm^2/(K·s) from experiment [18].

2.1.2 Effective diffusion coefficient of nuclides or heavy metals

A tortuosity factor can be defined as the relationship between diffusion coefficient and void structure of a material [20]:

$$D_{(n)} = D_0 \frac{n}{\tau} \tag{5}$$

where $D_{(n)}$ is the effective diffusion coefficient; D_0 is the initial diffusion coefficient in water; n denote capillary porosity; and τ is the tortuosity factor that refers to the degree of hole twists and turns of the porous media. It is worthwhile to mention that both n and τ are normally determined by experimental tests, by which the effect of micro cracks or shrinkage on porosity should have been collectively included, though these were not individually evaluated.

The tortuosity factor, which depends on many properties such as water-to-cement ratio, proportion of filler in the cement, curing conditions, etc. is normally hard to determine, especially when leaching of cement hydrates and release of calcium are considered. Sanchez [21] found that there is an exponential relationship between tortuosity factor and porosity:

$$\tau = n^\eta \tag{6}$$

Hence the relationship between porosity and diffusion coefficient is reduced to：

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$$D_{(n)} = D_0 n^{1-\eta} = D_0 n^m \tag{7}$$

From Katz and Thompson [22], m is set to 2.5.

In order to introduce chemical fixation on the diffusion coefficient, we used the following equation proposed by Xue [23]:

$$D_{(r)} = D_0 \cdot r \tag{8}$$

where $D_{(r)}$ is the diffusion coefficient considering chemical fixation ability of particles; r is the ion coefficient representing the ratio of the number of free particles to the total number of particles. In this paper, r is 0.7 and 0.06, respectively, for Sr and Cs nuclides.

When considering the influence of temperature on the diffusion coefficient, we followed Amey`s theory [24]:

$$D_{(T)} = D_0 \frac{T}{T_0} e^{q\left(\frac{1}{T_0}-\frac{1}{T}\right)} \tag{9}$$

where $D_{(T)}$ is the particle diffusion coefficient at temperature T; D_0 is the initial particle diffusion coefficient at temperature T_0; T_0 normally takes 25℃; q is the constant of activity (related to activation energy), which is related to water-to-cement ratio.

When considering the influence of cement hydration effects on the diffusion coefficient, for short-term leaching, we used Kamash’s theory for diffusion coefficient [13]:

$$D(t) = D_0 \left(\frac{t_0}{t+t_0}\right)^{\omega} \tag{10}$$

where $D(t)$ is the diffusion coefficient at *t*; D_0 is the initial diffusion coefficient at t_0, t_0 is the curing time of the cement block often taking the value of 28 d; ω is the diffusion attenuation factor related to the properties of cement block and the leaching liquid, taking values of 1.1 and 10 for Sr and Cs, respectively.

Eqs.(7-10) are the respective variations of the diffusion coefficient with a change of porosity, ion coefficient, temperature or hydration time, representing a modification on the initial diffusion coefficient D_0. To include the effect of all the influences, the diffusion coefficient can be collectively expressed as:

$$D' = D_0 \cdot n^{2.5} \cdot r \cdot \frac{T}{T_0} \cdot e^{q(\frac{1}{T_0}-\frac{1}{T})} \cdot \left(\frac{t_0}{t_0+t}\right)^{\omega} \tag{11}$$

It can be seen from the above that chemical fixation ability and diffusion attenuation determine the diffusion properties of particles. In general, different particles (e.g., Sr and Cs) will have different diffusion coefficients.

2.1.3 The selection of an adsorption equation

According to the model proposed by Suarez [10], we adopted the linear isothermal adsorption equation for the particles in cement solidification:

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$$F_l = K_d C \tag{12}$$

where F_l is the linear isothermal adsorption concentration; K_d is the adsorption distribution coefficient of a material, taking values, respectively of 150 ml/g for Sr and 15 ml/g for Cs in this paper[25]; C is the concentration of nuclide in the pore solution.

However, when it came to the choice of adsorption equation of metal ions dissolved in pore solution, a Langmuir linear adsorption isotherm was employed. It was shown experimentally, that Langmuir's equation described the absorption process more accurately when the pH of cement is greater than 9:

$$F_e = \frac{S_m K_L C}{1 + K_L C} \tag{13}$$

where F_e is the amount of sorbate adsorbed at equilibrium; S_m is the maximum monolayer adsorption constant (243 $\mu\mathrm{eq/g}$ for Sr and 332 $\mu\mathrm{eq/g}$ for Cs, respectively in this paper); K_L is Langmuir adsorption distribution coefficient, taking values of 8.3 ml/g for Sr and 1.9 ml/g for Cs, respectively[26]. In general, temperature has an impact on absorption. However, one the basis of the authors' best knowledge, there are no published results on the relationship between absorption and temperature that can be used in the simulations. We used the adsorption constant that was obtained from experimental tests carried out under the room temperature [13] . In principal absorption will be reduced with increase of temperature there exists a limiting absorption [27] even as temperature is increased further.

When cement solids are not carbonated, their pH is greater than 9 and a Langmuir isothermal adsorption equation should be chosen. Otherwise, for the fully carbonated zone, the pH is less than 9 and the linear isothermal one should be used instead.

2.1.4 Index of leaching behavior

This study mainly focused on hazard waste cement solids containing nuclides. For nuclides, the Chinese standard test method for leachability of low and intermediate level solidified radioactive waste forms (GB/T 7023-2011) was followed. According to GB/T 7023-2011 [16], the measurements of leaching resistance of cement solidification are leaching rate R_n and accumulative leaching rate P_t. The leaching rate measures the speed and trend of leaching and the accumulative leaching rate is directly related to the degree of leaching. The R_n and P_t are calculated, respectively, as below:

$$R_n = \frac{a_n/A_0}{(S/V)(\Delta t)_n} \tag{14a}$$

$$P_t = \frac{\sum a_n/A_0}{S/V} \tag{14b}$$

where a_n is the leaching quality in the $\mathrm{n_{th}}$ leaching cycle; A_0 is the initial leaching quality of the components; S is the contact surface area between the test block and leaching liquid; V is the volume of the test block; $(\Delta t)_n$ is the number of days in the $\mathrm{n_{th}}$ leaching cycle.

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2.2 The theoretical basis of the carbonation model

2.2.1 Governing equation

With a pressure greater than 7.29 MPa and a temperature higher than 31.26 ℃, the process of carbonation will be accelerated and the state of carbon dioxide is called supercritical, at which the carbon dioxide has the viscosity and diffusion coefficient close to a gas while the density is close to that of a liquid. Under these conditions, it will be easier for carbon dioxide to penetrate into a porous media like cement and take part in carbonation reactions.

In this paper, we use a carbonation model that is different from the commonly-used one established by Saetta [28] for natural carbonation. We use solubility instead of concentration of CO_2 in the pore water, since supercritical carbonation takes place in an environment where there is always sufficient CO_2 in the pores of cement solids to keep the pore water constantly in a saturated state. Then, we utilize the following PDEs consisting of Darcy's law (Eq.15a), the conservation equation of energy (Eq. 15b), conservation of mass (Eq.15c), the reaction rate equation (Eq.15d) and the concentration equation of carbon dioxide (Eq.15e) [17]:

$$\overrightarrow{u_\alpha} = -\frac{kk_{r\alpha}}{\mu_\alpha}(\Delta P_\alpha - \rho_\alpha \vec{g}) \tag{15a}$$

$$\rho c \frac{\partial T}{\partial t} = \nabla \cdot (\lambda \nabla T) - \left(C_g \rho_g \overrightarrow{u_g} + C_w \rho_w \overrightarrow{u_w}\right)\nabla T \tag{15b}$$

$$\frac{\partial (n S_\alpha \rho_\alpha)}{\partial t} + \nabla \cdot (\rho_\alpha \overrightarrow{u_\alpha}) = q_\alpha \tag{15c}$$

$$\frac{\partial R_c}{\partial t} = \alpha_1 f_1(h) f_2(g_v) f_3(R_c) f_4(T) \tag{15d}$$

$$\frac{\partial m_{co_2}}{\partial t} = \frac{\partial g}{\partial t} \tag{15e}$$

where $\overrightarrow{u_\alpha}$ is the Darcy flow velocity of component α; k is the intrinsic permeability of the material, which is related to its porosity; $k_{r\alpha}$ is the relative permeability of component α; μ_α is the dynamic viscosity of component α; P_α is the pressure of component α; ρ_α is the density of component α; $\vec{g}$ is the acceleration of gravity, which is ignored in our model; q_α is the change of the source term caused by chemical reaction or precipitation of component α; λ is the equivalent thermal conductivity coefficient of the material; ρc is the equivalent heat capacity of the material; g is the concentration of carbon dioxide in the pore solution; and m_{co_2} is the solubility of carbon dioxide in the pore liquid, which is related to pressure and temperature [29].

The five equations (15a-15e) are coupled PDEs with five variables, including degree of carbonation R_c, pressure of gas P_g, pressure of water P_w, concentration of carbon dioxide in pore solution g and temperature T, that are the governing equations of supercritical carbonation.

2.2.2 Initial values and boundary conditions

In order to solve the above PDEs, one needs to measure the initial values and boundary conditions. The initial values of the five independent variables are imposed as shown below:

$$R_C = R_{c0} = 0, P_g = P_{g0},\ P_w = P_{w0},\ g = g_0 = 0 \text{ and } T = T_0$$

The boundary conditions on the insulated boundaries, including the pressure of carbon dioxide P_g, the pressure of the liquid P_w and the temperature T, satisfy the following equations:

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$$\vec{n} \cdot \nabla P_g = 0,\ \vec{n} \cdot \nabla P_w = 0 \text{ and } \vec{n} \cdot \nabla T = 0$$

On the inflow boundary, the pressure of carbon dioxide P_g, the pressure of the liquid in the pore P_w and the temperature T on the boundary are shown as below:

$$P_g = P_{g,sur},\ P_w = P_{w,sur} \text{ and } T = T_{sur}$$

2.3 Relation between degree of carbonation and porosity

Through the introduction of the theories of accelerated carbonation and leaching processes, we know that in the process of leaching, porosity directly affects the diffusion coefficient of the particles. In order to evaluate leaching, how carbonation changes porosity is essential. In this study, we used the relationship proposed by Kwan and Song [30] as shown below:

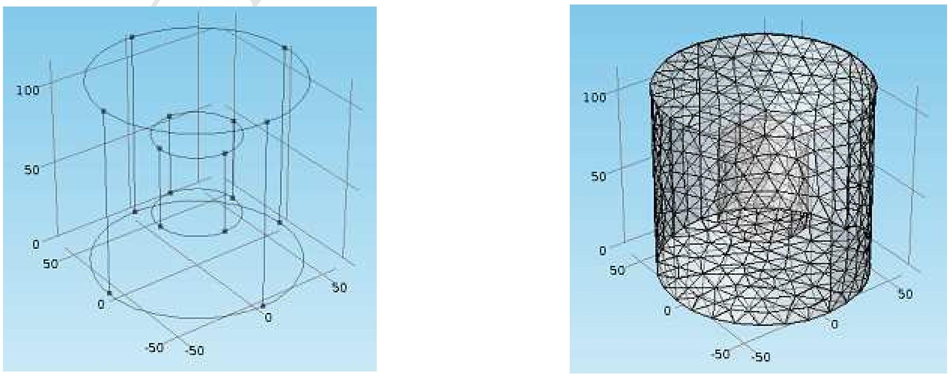
$$n = f(R_c) = \begin{cases} n_0(1 - 0.5R_c) & 0 \le R_c \le 0.4 \\ 0.8n_0 & 0.4 \le R_c \le 1 \end{cases} \tag{16}$$

where n_0 is the initial value of capillary porosity before accelerated carbonation, taken as 0.13[31] in this paper. R_c is the degree of carbonation defined as $R_c=c/c_{max}$; c and c_{max} are the current and the maximum amount of calcium carbonates, respectively.

3. Coupled model of carbonation and leaching

FEM simulations were conducted using COMSOL, as virtual tests, for cement blocks in the containers filled with deionized water during the test time. Following the guidelines in GB/T 7023-2011, we replaced the leach liquid at the 1st, 3rd, 7th, 10th, 14th, 21st, 28th, 35th and 42nd days. The replaced water was tested for leaching and cumulative leaching rates. The model is shown in Fig.1(a). According to GB/T 7023-2011, the chosen size of the cement block is φ50 mm×50 mm, and the size of the leaching container is φ120 mm×150 mm. The height of the liquid inside the container is 106.2 mm and the total volume of the leaching liquid is 1.2 L.

Boundary layer meshes were used at the boundary of the cement block and automatic tetrahedron meshing in other parts. The total number of elements is 12158. The FE-meshes are shown in Fig.1(b). In order to detect the concentration of waste particles in the leaching liquid, we added 40 numerical probes uniformly distributed in the solution to record the liquid concentration.



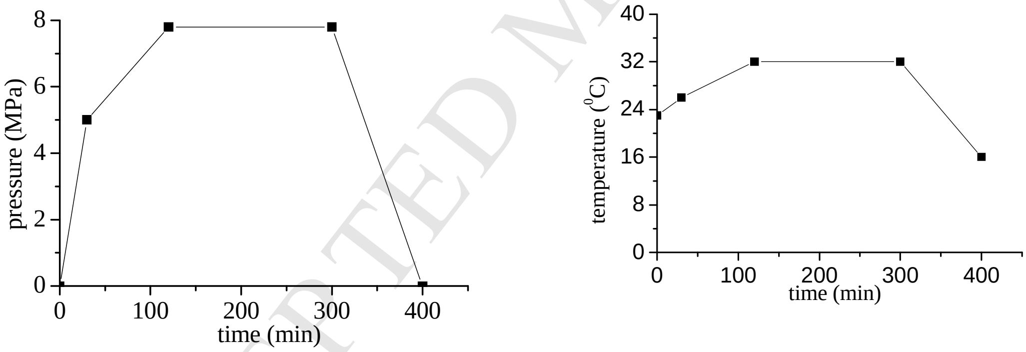
(a) Model geometry (b) Model meshing

Fig. 1 Test unit diagram

However, in order to calculate the required measurements continuously throughout the carbonation and then the leaching processes using the same model, the simulations consisted of the following two steps to decouple the processes of carbonation and leaching. The first step was to simulate supercritical carbonation. We set the diffusion coefficient of internal particles as infinitesimal to achieve the effect of carbonation without any leaching. The second step was for leaching assuming that the carbonation process had completed. We then used the desired diffusion coefficient of the particles and set the velocity of carbonation to zero to start leaching without carbonation.

Following the guidelines in GB/T 7023-2011, the simulation included two processes within each of the leaching phases. The first process was the normal leaching process for evaluating leaching resistance, during which the flux at the boundary of the container was set to zero to prevent any particles from leaching out of the container. The second process was for the moment of replacing the leaching solution prior to the next leaching phase. To do this, both the flux of the container and the diffusion coefficient of the particles in the leaching liquid were set to a large number to allow fast deionization and achieve zero ion concentration of the leaching liquid. During the leaching process, the cement block and leaching liquid were modelled as the same material with different diffusion coefficients that are much greater in the leaching liquid than in the cement block, in order to simulate particles leaching from the waste solids into the leaching liquid.

The simulation of the leaching process of the particles, Sr, was carried out for two different conditions imposed on the cement blocks, i.e., the carbonation condition shown in Fig.2 before leaching and the leaching process without applying supercritical carbonation.



(a) Pressure curve (b) Temperature curve

Fig.2 Supercritical carbonation condition

The predicted degree of carbonation R_c from the simulation is shown in Fig.3 under the supercritical conditions shown in Fig. 2, where a scale of 1 indicates that full carbonation has occurred. A close examination of the results shows that the carbonation depth is about 1 mm and the transition zone is about 13 mm. The 3D carbonation results for the same cement block under natural conditions (0.1 MPa and 25 ℃) for 4 h are shown in Fig.4, in which there is virtually no sign of carbonation.

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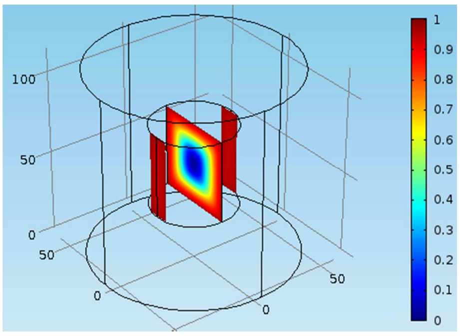

Fig.3 Degree of carbonation (supercritical)

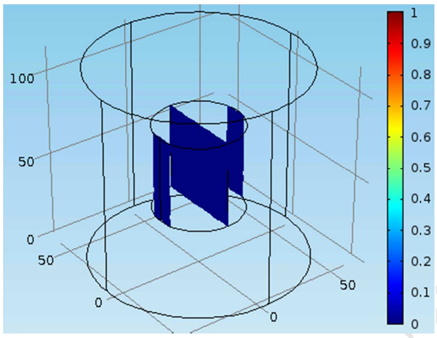

Fig.4 Degree of carbonation (natural)

The simulated concentrations of Sr in the leaching solution under the above two conditions taken before replacing the leaching liquid are shown in Fig.5, from which the leaching rates are calculated and presented in Fig. 6.

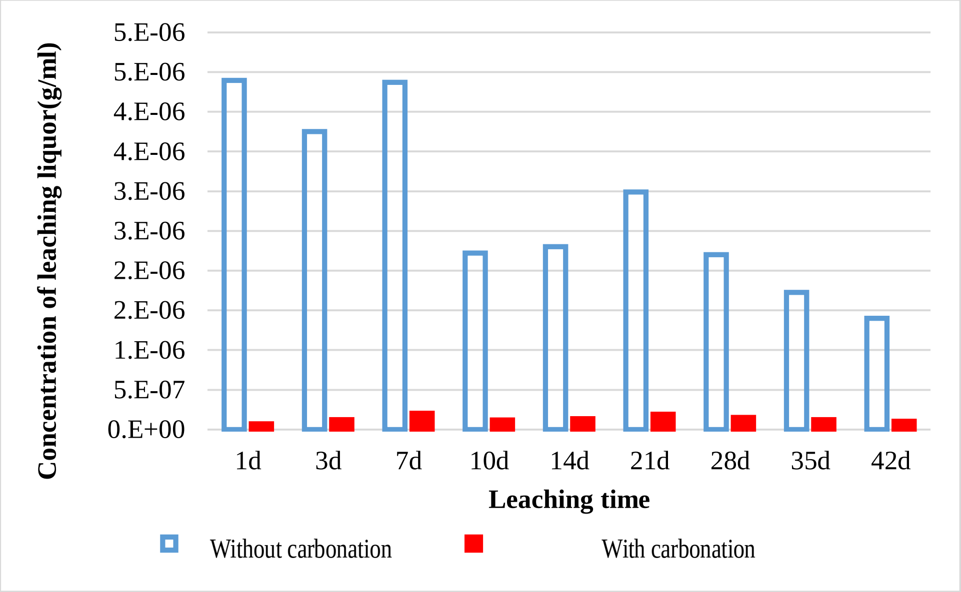

Fig.5 Concentration of leaching solution

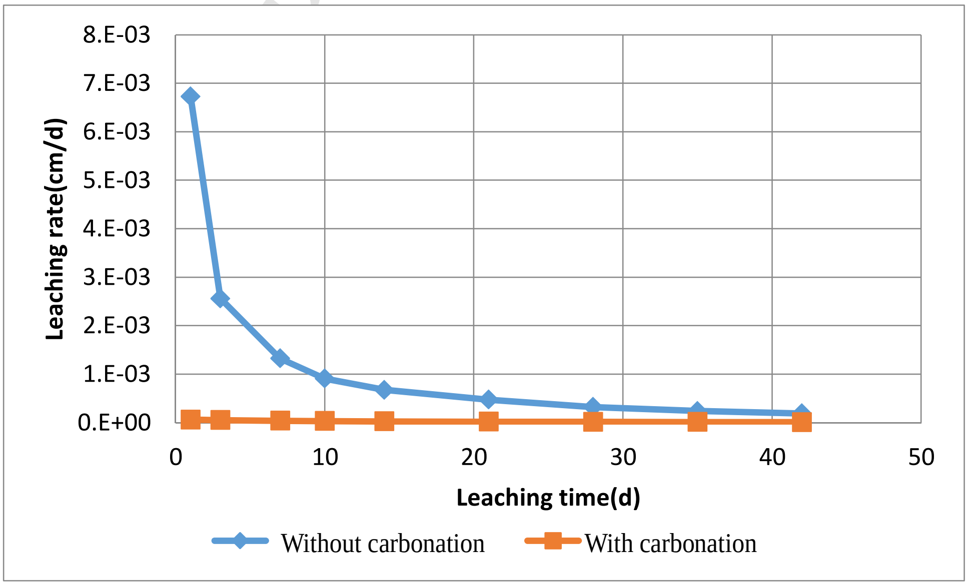

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Fig.6. Leaching rate curve

It is worthwhile to mention that the above model did not include the leaching effects of cement hydrates on the overall leaching process, though to some extent some of the effects might have been partially included due to the use of various material constants obtained from the leaching tests of other researchers. Detailed analyses of the release of the hydrates will be the subject of future studies.

4. Experiment

To verify the numerical model, 12 test blocks, which contain either Cs or Sr (the mass ratio of these elements is about 0.58) with a water-to-cement ratio of 0.37 by mass, were made for laboratory tests. All the blocks had the same dimensions as those used in the simulation. They were equally divided into four groups, two of which had added Sr and the others had added Cs. Within the groups with the same added chemicals, one group was carbonated while the other was not. Average leaching rates were taken from each of the groups and are compared in this Section. The specifications of the group of blocks are shown in Table 1.

Table 1. Leaching test samples

Chemicals added	Sample number	Leaching temperature (℃)	Carbonation
Strontium nitrate	Sr-7, Sr-10, Sr-12	25	No
	Sr-1, Sr-2, Sr-6	25	Yes
Cesium chloride	Cs-5, Cs-6, Cs-14	25	No
	Cs-2, Cs-4, Cs-10	25	Yes

4.1 Carbonation experiment

The procedure of the carbonation experiment is shown in Fig.7. Gaseous carbon dioxide flowing out from the gas cylinders turns into a supercritical state after flowing through the high pressure air pump and the temperature adjusting machine. The supercritical carbon dioxide then flows into the reaction vessel and takes part in carbonation with the test blocks. Finally, the remaining carbon dioxide flows through a switching equipment back to the gas cylinders. This experiment is conducted in a closed loop system, so that we can recycle the remaining carbon dioxide.

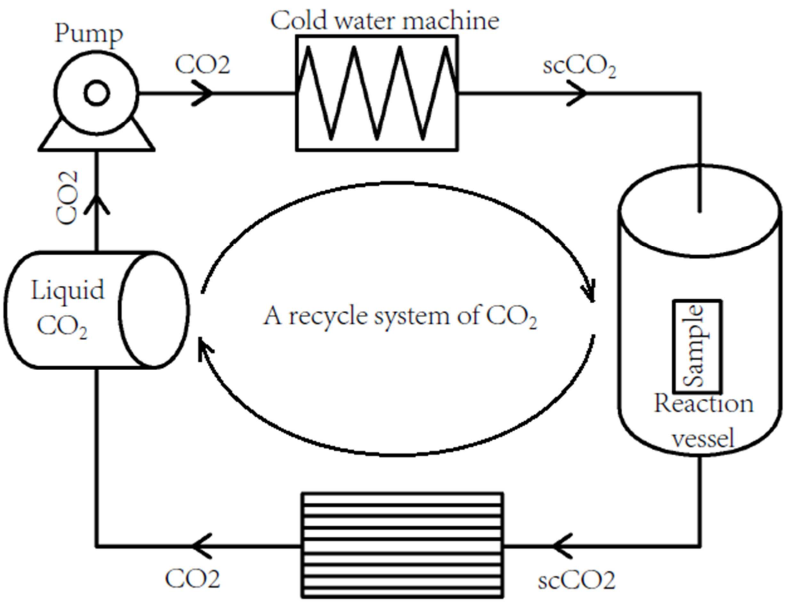

Fig.7. Design schedule and procedure of accelerated carbonation

The carbonation condition is the same as shown in Fig.2. After carbonation, a block was cut to show the internal section. The carbonation depth was measured using phenolphthalein, by which the fully carbonated area did not exhibit a red color. The measured carbonation depth is shown in Fig.8, where the carbonated depths in eight random locations were measured. The measured average was 1 mm and was consistent with the simulation results from the previous Section.

Fig.8 Carbonation depth

In order to estimate the amount of carbon dioxide absorbed by the test block during the process of carbonation, the mass of the blocks was measured before and after carbonation, so that the percentage of mass increase were calculated as shown in Table 2.

Table 2 Mass increase percentage

Sample number	Mass before carbonation (g)	Mass after carbonation (g)	Mass increase percentage (%)	Average mass increase percentage (%)
Cs-2	193.86	199.57	2.914	2.946
Cs-4	193.07	198.34	2.694	

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Cs-10	193.86	199.57	2.914	
Cs-5	192.06	197.97	3.036	
Cs-6	194.20	200.27	3.096	
Cs-14	194.07	200.12	3.087	
Sr-1	193.95	199.50	2.866	3.333
Sr-2	193.90	198.18	2.207	
Sr-6	194.31	198.91	2.367	
Sr-11	194.19	202.86	4.465	
Sr-14	193.42	201.65	4.255	
Sr-15	195.44	202.85	3.791	

From Table 2, it can be seen that the blocks with added chemicals have an approximate mass increase ratio of about 3 %.

4.2 Leaching test

To evaluate the effect of carbonation on leaching, the pairs of groups (Table 1) containing the same added chemicals were tested and compared for leaching, giving us the results for blocks with and without carbonation. The test blocks were produced from ordinary Portland cement and the water was deionized by an ultra-pure water machine.

The leaching temperature was controlled at 25 ℃ in the oven. The leaching solution at ages of 1, 3, 7, 10, 14, 21, 28, 35 and 42 d were replaced and measured for ion concentrations that were used to calculate the leaching rates of the cement blocks. The measured average leaching rates of Sr and Cs from the blocks with and without carbonation are shown in Fig.9, respectively. Similar tests were also carried out at 70 ℃ and the respective results are shown also in Fig.9.

Fig.9 shows all the measured leaching rates of the blocks listed in Table 1 against leaching time and two different temperatures. Fig.9 (a) and (b) are, respectively, for leaching of Sr and Cs. From Fig.9 (a), we can see that the leaching rates of Sr are all reduced significantly due to carbonation. This mainly attributes to the fact that carbonation improves the compactness of cement solidification resulting in a positive effect on retention of the internal particles. However, from Fig.9(b), an increase of leaching rate of Cs after carbonation occurs between day 5 and day 25 at 70 ℃. The reasons for this are not clear from the tests. More tests and long term leaching tests lasting several years are needed for further study.

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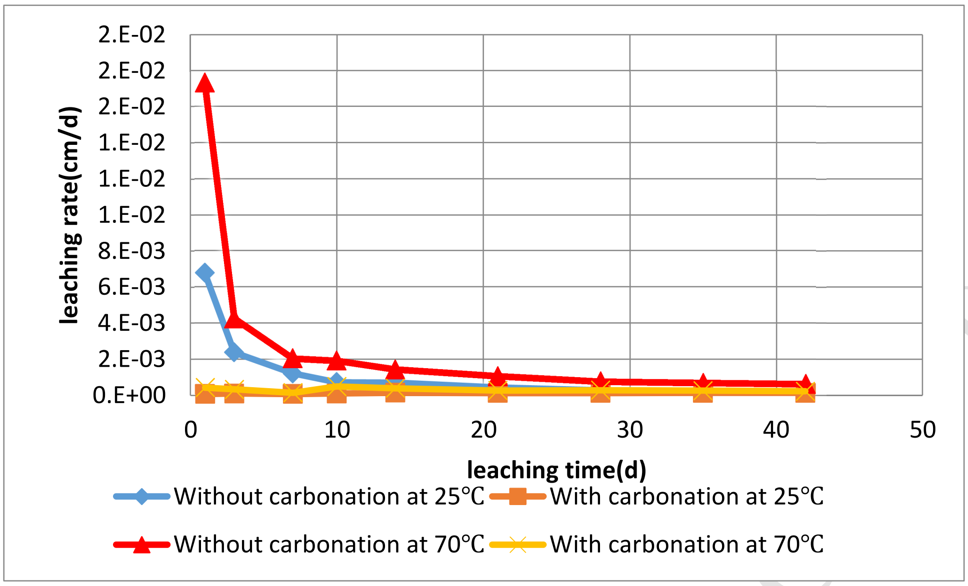
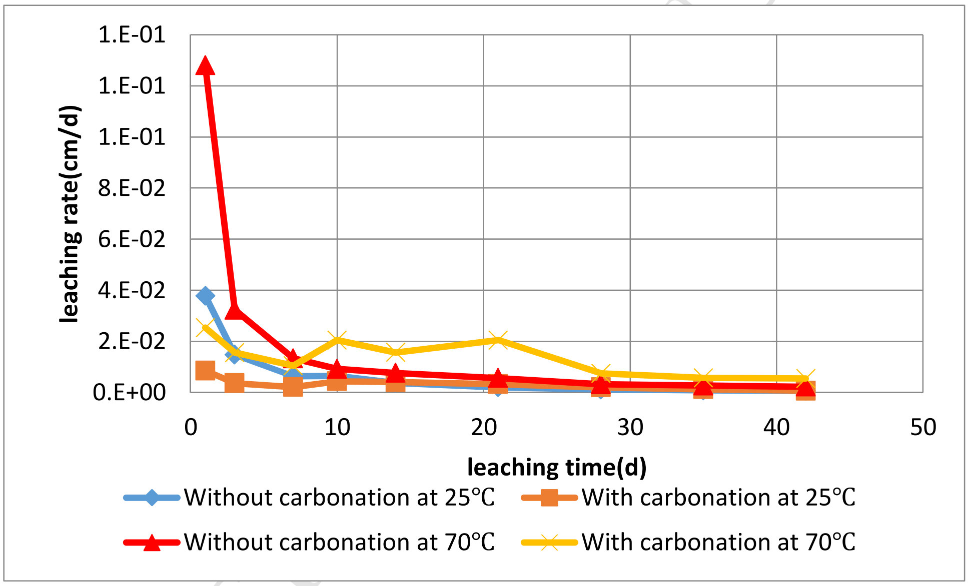


a) Leaching rate of Sr



b) Leaching rate of Cs

Fig.9. Leaching rate from experiments

In order to validate the numerical models, Fig.10 presents comparisons between the experimental and the simulation results of Sr and Cs leaching at 25 ℃. The comparisons show that the numerical models developed in this paper can accurately predict the process of leaching.

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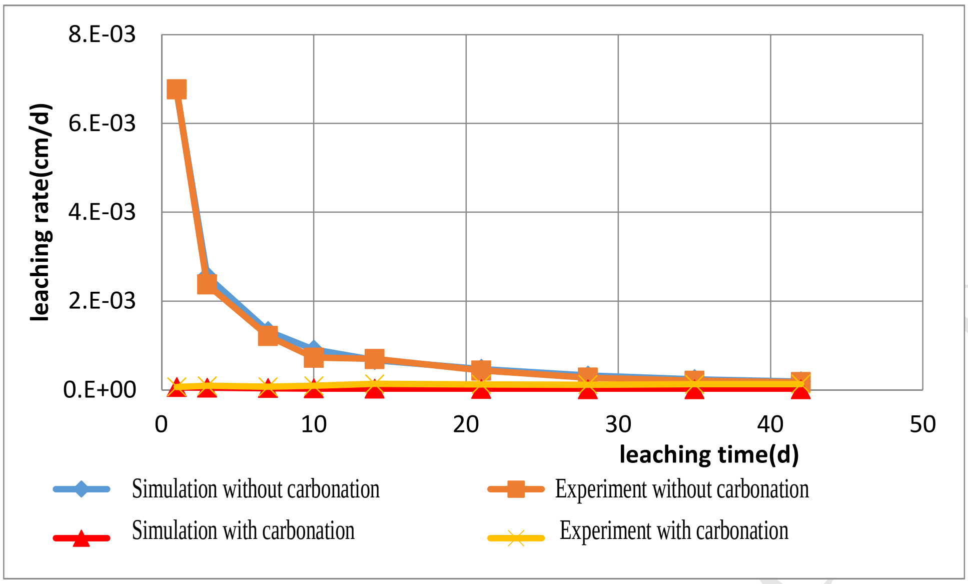
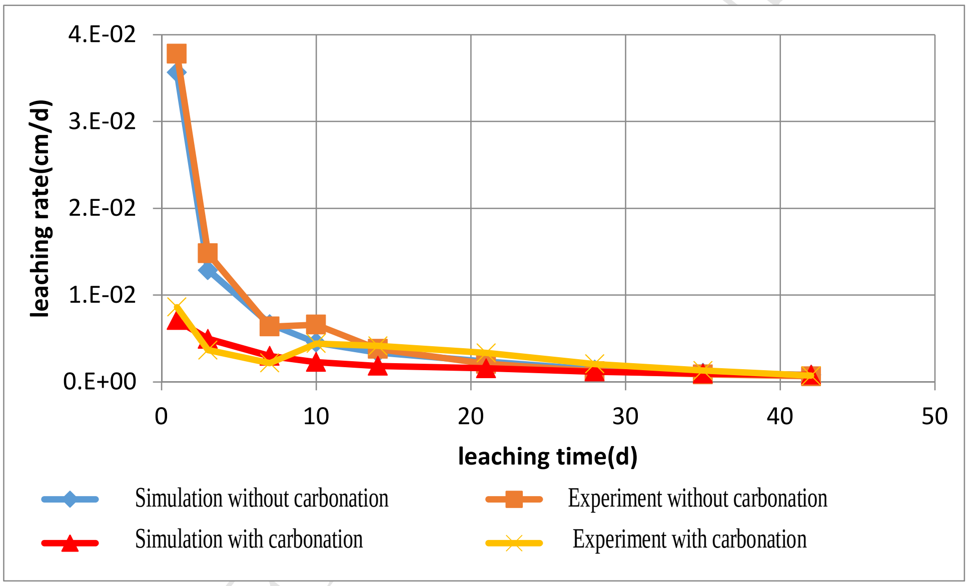


(a) Leaching rate of Sr



b) Leaching rate of Cs

Fig.10 Comparisons of experimental and simulation results

5. Conclusion

A three-dimensional model of particles leaching from cement solids has been developed in this paper. The effects of temperature, time, chemical consolidation, properties of cement can all be taken into account by this model.

Both carbonation and leaching tests were conducted in this research, which have shown that accelerated or supercritical carbonation can reduce the leaching rate and effectively improve the impermeability of cement solidification.

The experimental tests were also used to validate the developed three-dimensional leaching

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model. The satisfactory comparisons suggested that the combined carbonation and leaching model developed here can accurately simulate the processes of leaching tests with or without supercritical carbonation.

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Highlights

- We developed a multi- physics and –phase model for supercritical carbonation and leaching of cement based materials.
- We conducted experiments to validate the model.
- We investigated the impact of carbonation on the retention capacity of cement.
- We studied the effect of various physical properties on the leaching process of Sr and Cs ions.