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Stochastic leaching analysis on cementitious materials considering the influence of material uncertainty

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HIGHLIGHTS

- A stochastic numerical framework for modelling leaching is developed.
- Efficient stochastic analyses are achieved by employing a novel sampling algorithm.
- A modified root-time relationship for leaching is proposed and verified.
- The influence of the physical uncertainty on long-term leaching is overwhelming.
- The impact of chemical uncertainty is more evident in terms of short-term leaching.

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ABSTRACT

Uncertainties significantly influence the durability-related experiments and field studies, which can hardly be addressed by deterministic approaches. This work aims at developing a stochastic numerical framework to disentangle the influence of material uncertainty on the case of leaching. To ensure the robustness of the numerical framework, a realistic stochastic reactive-transportation model is developed, which consists of a novel sampling algorithm and a comprehensive deterministic model. By using the proposed sampling algorithm, a more effective and efficient sampling process can be achieved without compromising the randomness of the uncertain properties. Besides, realistic mechanisms of leaching are considered by the deterministic model, including the simultaneous processes of ionic transportation, chemical reactions and material degradation. By performing the stochastic leaching analyses, numerical results suggest the overwhelming influence of the physical uncertainty on long-term leaching, while the impact of chemical uncertainty is more evident in terms of short-term leaching. It is also revealed that the root-time relation as determined from short-term experiments is inappropriate for long-term predictions. Thus, a modified relation is developed based on the stochastic leaching analysis, which generates accurate predictions for both the short-term and long-term leaching.

1. Introduction

Performance-based analyses and designs of concrete structures were normally focused on the structural behaviours under catastrophic actions [1–4]. In engineering practice, cementitious materials are also widely adopted in a variety of waste disposal systems and geological facilities, where the chronical degradations of materials become dominant. For most types of cementitious materials, except for the novel low-alkalinity cements, the hydrated cement system usually creates a porous structure with highly alkaline pore solutions. The alkaline environment is generally desirable in the waste disposal system, as it ensures the low solubility of many radionuclides and favours the immobilization of hazardous wastes [5]. The durability of the cementitious material to maintain the alkaline environment for the containment of wastes is, thus, of great importance in general.

However, as the waste disposal systems are buried underground and constantly in contact with ground water, leaching of the cementitious material poses a serious durability concern [5]. Groundwater normally contains low mineral contents and is of neutral *p*H value. As a result, many cement hydrates will dissolve and be leached out of the material, including polandite (CH), calcium-silicate-hydrates (C-S-H), monosulfates (AFm), ettringite (AFt) etc. [6]. For most types of hydrated cements, the presence of CH buffers the *p*H of the pore solution, and the dissolution of

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CH leads to the significant decrease of pH value, which would cause the loss of durability in terms of immobilizing the hazardous wastes. Furthermore, leaching can also cause degradations of mechanical properties, alteration of microstructures, and loss of chloride binding capacity [7–10], which contribute significantly to the corrosion issue [10–16]. The decalcification of C-S-H and the dissolution of AFm during leaching would lower the chloride binding capacity, and hence potentially accelerate the corrosion initiation. Moreover, the decrease of pH value due to the dissolution of CH also contributes to the depassivation of reinforcements. Samson and Marchand [15] have shown the significant impact of leaching on the total chloride content in a 20-year-old parking structure.

In terms of evaluating the degree of leaching in the cementitious material, leached depth is the most straightforward indicator. For the past decades, experimental and numerical techniques have been greatly advanced, and the leaching mechanisms have been investigated by various deterministic approaches [7,17–25]. It is widely accepted that the development of leached depth follows a root-time relationship. Recently, a comprehensive deterministic method for modelling leaching and external sulfate attack was developed by the authors [19], where the obtained simulations also showed the general agreements with the root-time relation within the short-term durations of leaching.

However, the determination of the detailed parameters that govern the root-time relation has yet been systematically addressed. To the best of the authors' knowledge, only a few reported studies published the root-time relation with the constant parameters determined from specific short-term experiments, ranging from 3 months to 1 year [26]. Considering the random nature of the material and the changing pore structures under leaching, the validity of the reported root-time relations is limited by nature of the short-term deterministic studies. The accuracy and applicability of the root-time relation in modelling a long-term leaching case considering the influence of material uncertainty have yet been reported.

In this paper, a stochastic numerical framework is developed to investigate the influence of material uncertainty on the leaching of cementitious materials. The material uncertainty considered in the present research is classified into two categories, i.e. physical and chemical uncertainties. In the numerical framework, a Delayed Rejection Metropolis Hastings (DRMH) algorithm is introduced to achieve a more effective and efficient process for conducting the Monte Carlo simulations (MCS). The comprehensive deterministic model as developed by the authors [7,19,27] is coupled with the DRMH algorithm and a stochastic reactive-transportation model is proposed. The deterministic model is validated against the reported experiments, and the influence of material uncertainty on leaching is investigated by performing MCS. A modified root-time relation is then developed based on the stochastic leaching analyses. By implementing the proposed relation, the reliability of cementitious materials under a long-term laboratory leaching condition is studied. Furthermore, the sensitivity analyses on the material uncertainties are also conducted.

2. Material uncertainties in the stochastic leaching analysis

2.1. Limit state of leaching

Leaching is normally evaluated by measuring the leached depth [7], which is defined as the length measured from the depletion front of CH perpendicularly to the exposure surface [23,26]. As a drop of pH value is accompanied with the depletion of CH, pH value can be adopted as the indicator to determine the leached depth, such as using phenolphthalein in the test. In the present numerical simulation, the pH values along the leaching path are evaluated using Eq. (1) [11]. The leached depth is determined as the length in the leaching path where the pH value stays below a certain threshold. Considering the sharp front of CH dissolution, a critical pH value of 9 is assumed herein, where CH can be considered depleted below this value.

$$pH = \begin{cases} 14 + \log(2 \times 10^{-3} C_{CH}), & C_{CH} \geqslant 1 \times 10^{-3} \\ 8.3, & \text{otherwise} \end{cases} \tag{1}$$

where C_{CH} is the molar concentration (M) of the remaining CH content in the solid skeleton of the cementitious material.

In terms of assessing the durability of cementitious materials under leaching, two types of limit states are commonly referred to, i.e. (1) the probability of the leached depth to exceed the threshold value within an intended service period, and (2) the probability of the actual service life being shorter than as planned according to a given depth, often referred to the thickness of the designed concrete cover. Both criteria are relevant to engineering practices. While the first criterion mainly targets at the optimization designing of the material and structure, the second criterion is usually implemented to assess the reliability of the in-situ specimens. The limit state functions for both criteria are given in Eq. (2a) and (2b) respectively.

$$g_1(\boldsymbol{\sigma}^R, L_d) = L_d^{th} - L_d(\boldsymbol{\sigma}^R, t^i) \tag{2a}$$

$$g_2(\boldsymbol{\sigma}^R, t^s) = t^s(\boldsymbol{\sigma}^R, L_d^{th}) - t^{th} \tag{2b}$$

where $\boldsymbol{\sigma}^R$ is the vector of random variables, L_d^{th} is the threshold of leached depth, t^{th} is the planned service life, $L_d(\boldsymbol{\sigma}^R, t^i)$ is the leached depth after continuously leaching for the intended service period t^i, and $t^s(\boldsymbol{\sigma}^R, L_d^{th})$ is the actual service life when the threshold of leached depth is reached.

According to Eq. (2), it is obvious that $g_1(\boldsymbol{\sigma}^R, L_d) > 0$ or $g_2(\boldsymbol{\sigma}^R, t^s) > 0$ represents the desired state, and $g_1(\boldsymbol{\sigma}^R, L_d) \leqslant 0$ or $g_2(\boldsymbol{\sigma}^R, t^s) \leqslant 0$ is considered as the failure state of cementitious materials under the leaching condition. The probability of failure P_f following either criterion is evaluated using Eq. (3). Eq. (3a) generates the probability of exceeding the threshold of leached depth within the intended service period, and Eq. (3b) gives the probability for the actual service life of the material being shorter than as planned. It is noted that the probability of failure obtained by using the two different limit states, i.e. $P_f(L_d)$ and $P_f(t^s)$, would be identical, when the threshold L_d^{th}, the intended service period t^i and the planned service life t^s are set as the same.

$$P_f(L_d) = \int_{g_1(\boldsymbol{\sigma}^R, L_d) \leqslant 0} f(\boldsymbol{\sigma}^R, L_d) d\sigma \tag{3a}$$

$$P_f(t^s) = \int_{g_2(\boldsymbol{\sigma}^R, t^s) \leqslant 0} f(\boldsymbol{\sigma}^R, t^s) d\sigma \tag{3b}$$

where $f(\boldsymbol{\sigma}^R)$ is the probability distribution function (PDF) of random vector $\boldsymbol{\sigma}^R$.

2.2. Material uncertainties

As shown in Eqs. (2) and (3), the vector of random variables $\boldsymbol{\sigma}^R$ plays an important role in determining the PDF and the corresponding P_f. In terms of evaluating the probabilistic durability of cementitious materials, many uncertain sources can contribute to the randomness of the system, including data uncertainty, model uncertainty, environmental uncertainty, and material uncertainty. The data uncertainty is brought by the lack of knowledge on the input data. The model uncertainty comes from the lack of