

## Effect of interface equilibrium in a semi-dynamic leaching test of geomaterials

H. Sakanakura<sup>1</sup>, Y. Imoto<sup>2</sup>, S. Ogawa<sup>3</sup>, and S. Inoba<sup>4</sup>

<sup>1</sup>Section Head, National Institution for Environmental Studies, Tsukuba, Japan,

email: [sakanakura@nies.go.jp](mailto:sakanakura@nies.go.jp)

<sup>2</sup>Senior Researcher, National Institute of Advanced Industrial Science and Technology, Tsukuba, Japan,

email: [y-imoto@aist.go.jp](mailto:y-imoto@aist.go.jp)

<sup>3</sup>Senior Researcher, Central Research Institute of Electric Power Industry, Abiko, Japan,

email: [ogshohei@criepi.denken.or.jp](mailto:ogshohei@criepi.denken.or.jp)

<sup>4</sup>Senior Researcher, Central Research Institute of Electric Power Industry, Abiko, Japan,

email: [innova@criepi.denken.or.jp](mailto:innova@criepi.denken.or.jp)

### ABSTRACT

A good understanding of the leaching mechanisms of potentially toxic substances released from recycled geomaterials is crucial for estimating their environmental impact. Semi-Dynamic Leaching Tests (SDLTs) such as ANS 16.1, NEN 7375, and prEN 16637-2 are serial batch-type leaching tests with repeated replacements of the entire liquid phase according to a programmed schedule. This study discusses several issues of leaching behavior in SDLTs. In addition to the monolith specimen, granules and clays that have low diffusion coefficients can indicate diffusion-controlled release. However, the leaching mechanisms observed in SDLTs are not necessarily diffusion-controlled. To explain the unidentified leaching mechanisms, we used a model that linked solid-liquid interface equilibrium and intra-solid diffusion. The model could reproduce various leaching behaviors, suggesting the need for further attention to the interface equilibrium in the field of leaching research.

*Keywords: leaching mechanism, intra-solid diffusion, adsorption-desorption equilibrium, diffusion coefficient, distribution coefficient*

### 1 INTRODUCTION

The recycling of various wastes and byproducts in construction work is important for reasons such as reducing the destruction of nature, depletion of resources, generation of carbon dioxide associated with the extraction of new materials from the ground, and generation of disposable waste. For example, iron slag, steel slag, copper slag, coal ash, waste concrete, construction sludge, and excavated soil are used as roadbed, embankment, and landfill materials after solidification, crushing, and particle size adjustment, as necessary. They can also be used as aggregates in concrete-asphalt mixtures. However, as these materials may contain potentially toxic substances (PTSs), evaluating the environmental impact of leaching is necessary using leaching tests. Because leaching tests differ from the actual environment, it is crucial to precisely understand the leaching mechanisms of PTSs from the geomaterials to estimate their actual environmental impact.

The Semi-Dynamic Leaching Test (SDLT) intermittently replaces the entire liquid phase, measures the concentration of PTSs in each liquid fraction, and examines the change in concentration over the fractions to estimate the leaching mechanism of the PTSs (Wang et al., 2021). Unlike dynamic leaching tests that use a continuous liquid transfer device, SDLTs can be performed manually without any special equipment. To date, several test methods have been standardized (NEN 7375, 2004; prEN 16637-2, 2021; ANS-16.1, 2019; and ASTM C1308, 2021). According to prEN 16637-2 (2021), SDLTs can identify the leaching mechanism of PTSs. The primary mechanisms addressed in this standard are surface wash-off and intra-solid diffusion. Intra-solid diffusion is a phenomenon in which a substance diffuses

inside a solid such as a particle or monolith. Once intra-solid diffusion is recognized as the rate-limiting mechanism and the diffusion coefficient is obtained successfully, the long-term change in the release flux,  $J$  (mg/m<sup>2</sup>/s), of the PTSs can be estimated. However, there are some cases in which the mechanism cannot be identified, such as the continuation of an almost constant concentration through the fractions.

The prEN 16637 series modifies the leaching test according to the characteristics of the solid sample, depending on whether water passes through the interior of the solid (prEN 16637-1, 2021). SDLT is applied to monoliths with a minimum size of 40 mm per dimension or to a low-permeability material, such as clay, filled in a one-side-open column (prEN 16637-2, 2021). For highly permeable materials, such as sandy soil and steel slag, a column percolation test is applied because water can percolate the interparticle pores in both the actual environment and the column percolation test (prEN 16637-3, 2021). The advection-dispersion model is used as a numerical model for both permeable ground and column percolation tests (Grathwohl and Susset, 2009). The leaching mechanism of the advection-dispersion model is the solid-liquid interface equilibrium, and the distribution coefficient is used as a leaching parameter. The interface equilibrium has not been considered in the release mechanism of the SDLT standards listed above.

Some highly permeable materials, such as sandy soil and slag, were shape-stable throughout the leaching tests, similar to monolithic materials. Diffusion could be observed in the leaching test of such “permeable” materials when applying a test similar to SDLTs (Sakanakura et al., 2003; Sakanakura, 2005; Sakanakura et al., 2009). Although the size of a single particle (2 mm at most) was smaller than that of the monolith, the diffusion coefficient is considered to be small, resulting in a shallow elution depth. Furthermore, when the release flux was small, the specific surface area of the granule was much larger than that of the monolith. Consequently, the leachate concentration, or the total amount released, was sufficient for quantification.

Leaching mechanisms should be explained without contradictions between monoliths, granules, and clays, regardless of the size of the solids. In other words, if the relationship between intra-solid diffusion and solid-liquid interface equilibria is explained consistently, the leaching of PTSs from ground materials in SDLTs, column percolation tests, and actual environments will be better understood. Therefore, a model was developed that links intra-solid diffusion and interface equilibrium (Sakanakura et al., 2022).

This study describes and discusses the leaching mechanisms and theory of SDLTs. The relationships between the diffusion coefficient, elapsed time, and elution depth were elucidated. A model connecting intra-solid diffusion and solid-liquid interface equilibrium, termed the  $D_F$ - $K_d$  model, was introduced. We applied the  $D_F$ - $K_d$  model to explain the SDLT results that could not be explained by the mechanisms listed in the existing SDLT standards.

## 2 LEACHING MECHANISMS

### 2.1 Surface wash-off and dissolution-precipitation equilibrium

Highly soluble substances that adhere to solid surfaces dissolve rapidly (surface wash-off). SDLTs can detect surface wash-off by intermittently repeating the replacement of the leaching liquid. If the solid is a highly soluble substance, such as sodium chloride, dissolution will occur, and the solid will shrink. Dissolution stops when the concentration in the liquid phase reaches solubility. The solubility-control equilibrium indicates that the dissolution and precipitation rates are equal. If the primary mineral of the focusing material, such as a silicate mineral, is less soluble, the solid will retain its original shape. Such a primary mineral is called a “matrix” (Chandler et al., 1997).

### 2.2 Intra-solid diffusion and depletion

According to Fick’s law, if there is a difference in the concentration in a certain space, the substances diffuse to eliminate the difference. Flux,  $J$  (mg/m<sup>2</sup>/s), the mass transferred per unit area and per unit time, is expressed by Equation (1).

$$J = -D \frac{dC}{dx} \quad (1)$$

where  $D$ ,  $C$ , and  $x$  represent the diffusion coefficient ( $\text{m}^2/\text{s}$ ), concentration ( $\text{mg}/\text{m}^3$ ), and distance ( $\text{m}$ ), respectively. Diffusion occurs not only in the gas and liquid phases but also in the solid phase. A model is proposed in the field of leaching research where a substance in a solid matrix diffuses through small water-filled pores (Finkel et al., 1998). In addition, there are cases where substances diffuse through the solid phase where water is absent (Borg and Dienes, 1988); whether it is water-borne or not, intra-solid diffusion occurs and can be described by a single diffusion coefficient.

When a substance dissolves into the liquid phase, the concentration of the solid phase near the surface decreases. As a result, the substance inside the solid diffuses toward the solid surface. If this phenomenon continues for an extended amount of time, the substances inside the solid will be depleted.

### 2.3 Solid-liquid interface equilibrium

The solid-liquid interface equilibrium, a state in which the rates of adsorption and desorption are equal, is a phenomenon that also works in leaching tests. When an eluted substance exists in the liquid phase, a part of the substance is adsorbed on the solid phase surface. For example, the Henry-type adsorption isotherm is represented by Equation (2).

$$q = K_d C_L \quad (2)$$

where  $q$ ,  $K_d$ , and  $C$  are the mass of the substance adsorbed per unit mass of solid ( $\text{mg}/\text{kg}$ ), distribution coefficient ( $\text{L}/\text{kg}$ ), and concentration in the liquid ( $\text{mg}/\text{L}$ ), respectively. Equation (2) represents that the adsorbed mass increases as the liquid-phase concentration increases. In this study, the words “sorption” and “absorption” are not used to distinguish between the mechanisms of surface adsorption and diffusion into the solid phase (Manceau et al., 2002).

In the field of leaching research, the interface equilibrium has received little attention, as it is not accounted for as one of the leaching mechanisms in prEN 16637-2 (2021). However, consider that the PTSs were initially on the solid surface; when in contact with water, a part of the substance will dissolve into the water, and an interface equilibrium will be achieved. When the liquid phase is replaced, as in an SDLT, a new interface equilibrium is established instantaneously. If the  $K_d$  value is sufficiently small, the interface equilibrium can be omitted.

The distribution coefficient is incorporated into the advection-dispersion model. Using this model, Grathwohl and Susset (2009) analyzed the leaching behavior of PTSs in a column percolation test. Sakanakura et al. (2021) developed a procedure to determine the  $K_d$  value of contaminant-releasing materials by using batch-type leaching tests with different liquid-solid ratios.

### 2.4 Exposure environment, shape loss, and alteration

Leaching is a phenomenon in which substances in the solid phase migrate into the liquid phase. Substances in the liquid and gaseous phases can promote or inhibit the leaching of solids. In addition, the pH, Eh, and other indices of the surrounding media may have a significant effect on leaching. They may dissolve the matrix to accelerate leaching. Leaching may be suppressed if secondary minerals generate on the surface of the solid. The magnitude of these changes varies with temperature. In addition, when the solid cracks and the specific surface area increases, leaching will be enhanced. Particularly, in laboratory tests with vigorous stirring, fine particles are generated, and the test results may deviate from the actual environment (Ogawa et al., 2022). Although these indices and changes affect the leaching properties, they are not regarded as the leaching mechanisms of the solid itself.

In this study, the focus of the release mechanisms was limited to surface wash-off, intra-solid diffusion (including depletion), and the solid-liquid interface equilibrium.

## 3 ESTABLISHED STANDARDS OF SEMI-DYNAMIC LEACHING TESTS AND THEORY OF INTRA-SOLID DIFFUSION

SDLT is also called a “diffusion test” because the tests are primarily aimed at capturing leaching due to diffusion from the solid phase. Therefore, the following points are important for the application of solid samples in tests: (1) The solid phase should be homogeneous. However, a high concentration of

solubles at the surface can be accepted as surface wash-off in the initial fractions and can be excluded from the diffusion analysis. (2) The shape should be stable. To achieve this, the matrix must remain insoluble.

Table 1 lists the specifications of the SDLT standards. Solid specimens for SDLTs are monoliths (such as molded mortar), with a side length of minimum 40 mm, and low-permeability unconsolidated materials (such as clay) filled with a one-sided open vessel. The specific surface area of monoliths and filled materials can easily be estimated and then used to calculate the diffusion coefficient. The ratio of the liquid volume and surface area of the specimen are regulated as 8-10 m<sup>3</sup>/m<sup>2</sup>. The liquid replacement schedules differ among the standards. The basic solvent for the test is pure water (such as ion-exchanged water) in all the standards, and some reagents, such as nitric acid with a pH of 4, can be chosen according to the test purpose.

**Table 1. Standards of semi-dynamic leaching test**

Standard	Ratio of liquid volume to surface area for monolith (m <sup>3</sup> /m <sup>2</sup> )	Liquid replacement time schedule from the start	Published (Revised)
<b>ANS 16.1</b>	10	2 h, 7 h, 1 d, 2 d, 3 d, 4 d, 5 d, 19 d, 47 d, 90 d	1986 (2019)
<b>ASTM C1308-21</b>	10	2 h, 7 h, 1 d, 2 d, 3 d, 4 d, 5 d, 6 d, 7 d, 8 d, 9 d, 10 d, 11 d	1995 (2021)
<b>NEN 7375</b>	10	6 h, 1 d, 2.25 d, 4 d, 9 d, 16 d, 36 d, 64 d	1995 (2004)
<b>prEN 16637-2</b>	8	6 h, 1 d, 2.25 d, 4 d, 9 d, 16 d, 36 d, 64 d	2021
<b>Method 1315</b>	9	2 h, 1 d, 2 d, 7 d, 14 d, 28 d, 42 d, 49 d, 63 d	2013 (2017)

Apart from shaped specimens, powder and granular samples can also be evaluated using some of the SDLT standards, such as ASTM C1308-21. In fact, some substances in powders and granules can show a diffusion-controlled trend, whereas monoliths do not always exhibit diffusion-controlled behavior (Sakanakura, 2005).

Assuming that the concentration in the liquid,  $C_L$  (mg/L), remains low and at a constant value, and that at a certain depth the initial concentration,  $C_{F0}$  (mg/kg), remains unchanged, changes in concentration in the solid phase,  $C_F$  (mg/kg), changes in release flux at the solid surface,  $J$  (mg/m<sup>2</sup>/s), and the cumulative amount released from the solid surface during time  $t_{i-1}$  and  $t_i$  (s),  $M_i$  (mg/kg), are expressed by Equations (3), (4), and (5), respectively (Chandler et al., 1997).

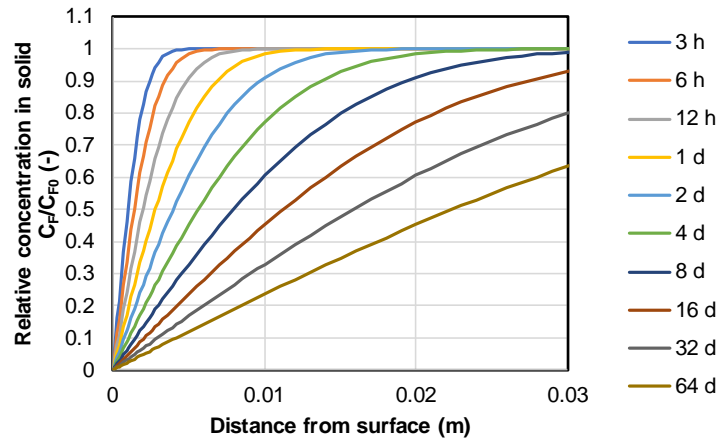
$$C_F = C_{F0} \operatorname{erf} \left( \frac{x}{\sqrt{4D_F t}} \right) \quad (3)$$

$$J = C_{F0} \sqrt{\frac{D_F}{\pi t}} \quad (4)$$

$$M_i = 2C_{F0} \sqrt{\frac{D_F}{\pi}} (\sqrt{t_i} - \sqrt{t_{i-1}}) \quad (5)$$

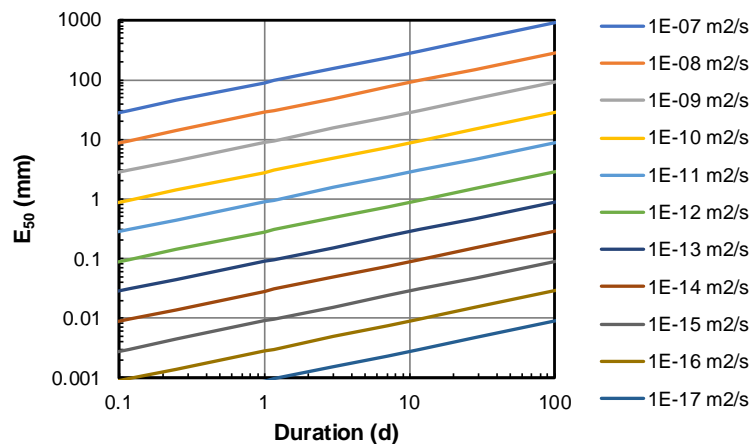
where  $x$  is the distance from the solid surface (m), and  $D_F$  is the intra-solid diffusion coefficient (m<sup>2</sup>/s). The parameter  $C_{F0}$  is the concentration value that contributes to diffusion, and its determination using an availability test was proposed earlier (NEN 7371, 2004).

Using Equation (3), the change in concentration distribution in the solid phase under  $D_F = 1 \times 10^{-10}$  m<sup>2</sup>/s is depicted as Figure 1. Equations (3)-(5) are derived based on the assumption that the depth of the solid is infinite. The SDLT standards specify the side length of the specimen as 40 mm (20 mm in terms of sample depth) to prevent depletion, such that the release flux follows Equation (3). Figure 1 shows that, the solid-phase concentration decreases to 50% (as an example of significant decrease from initial condition) after 32 days at a depth of 16 mm. Hence, if the sample depth is less than 16 mm in this case, depletion behavior will be observed, and the test result will deviate from Equation (3). This is just the case of  $D_F = 1 \times 10^{-10}$  m<sup>2</sup>/s. Depending on  $D_F$  of the solid, the design of SDLT should be optimized in terms of sample size and test duration to avoid depletion.



**Figure 1.** Changes in concentration distribution in solid with time ( $D_F = 1 \times 10^{-10} \text{ m}^2/\text{s}$ )

The elution depth,  $E_{50}$ , is defined as the depth at which the concentration in the solid phase reaches 50% of the initial concentration of  $C_{F0}$  (mg/kg). Figure 2 shows the changes in  $E_{50}$  over time. For a given test period, if the sample thickness is less than  $E_{50}$ , the release flux deviates from Equation (3), and depletion is observed. Depletion is a continuation of intra-solid diffusion.



**Figure 2.** Changes in elution depth ( $E_{50}$ ) with time for different diffusion coefficient values

Change in flux  $J$  of a substance decreases with time (Eq. (4)), depending on the initial concentration  $C_{F0}$  and the intra-solid diffusion coefficient  $D_F$ . SDLT can capture these parameters, and the environmental impact of the material can be calculated as long as the leaching mechanism is purely intra-solid diffusion, and the substance is not depleted during the observation period.

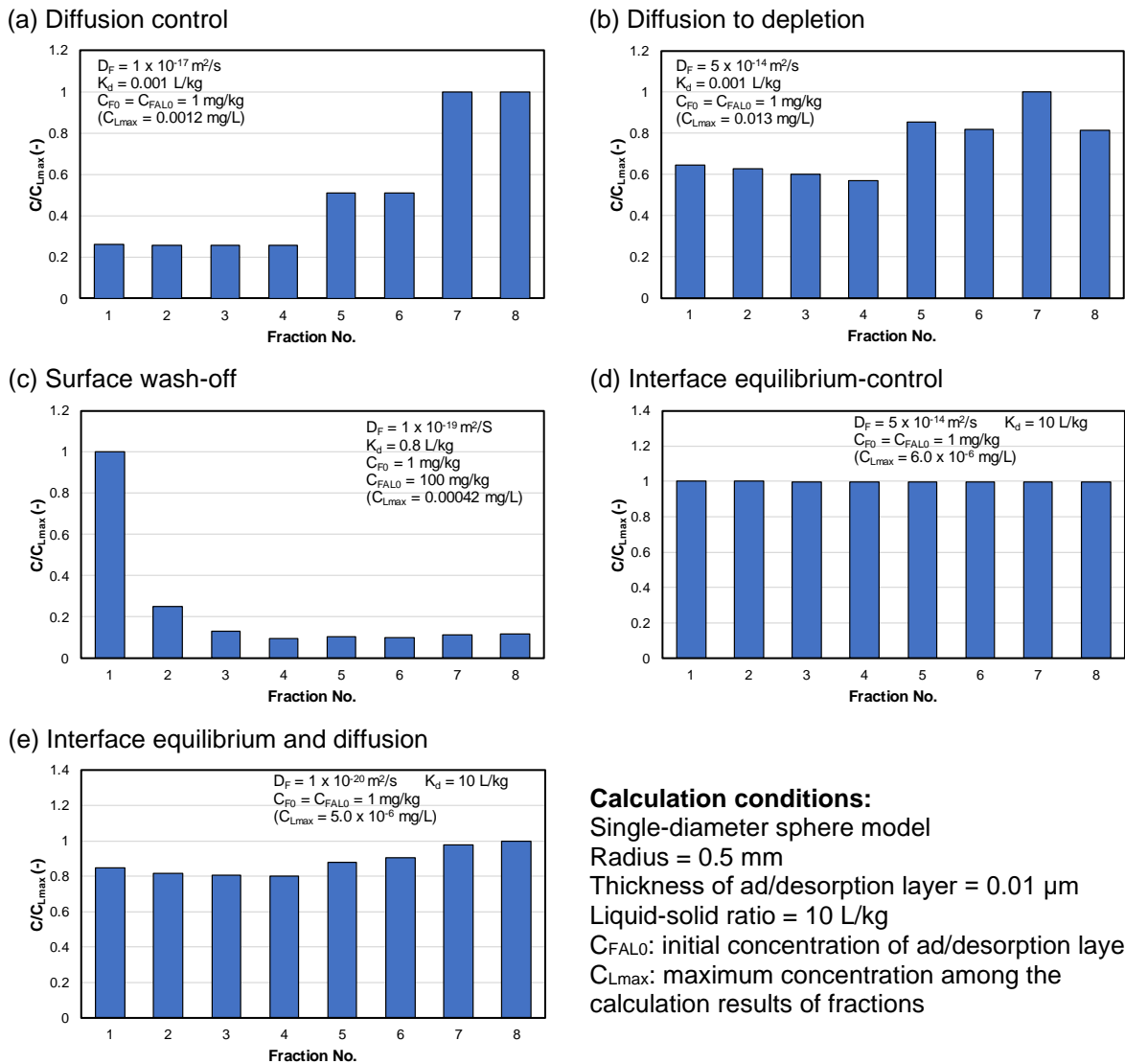
However, the actual SDLT results suggest that the leaching mechanism is rarely purely intra-solid diffusion (e.g., Sakanakura et al., 2009). Other mechanisms are surface wash off and dissolution-precipitation equilibrium (2.1), solid-liquid interface equilibrium (2.3), shape loss and alteration (2.4), etc. Among them, the effects of solid-liquid interface equilibrium on intra-solid diffusion will be specifically focused on in the next chapter.

## 4 A NEW MODEL LINKING INTRA-SOLID DIFFUSION AND INTERFACE EQUILIBRIUM

### 4.1 Interpretation and modelling of concentration change in SDLT

Figure 3 shows typical changes in liquid concentration by SDLT according to the replacement schedules of prEN 16637-2 and NEN 7375. Figure 3(a) shows very similar changes to the theoretical changes under diffusion control for these standard test methods. In addition, several cases are present where

the changes do not follow diffusion theory. Figure 3(b) shows that depletion progressed under diffusion control. In Figure 3(c), a constant concentration is obtained after surface wash-off that is difficult to explain by diffusion or solubility control; if the leaching mechanism is diffusion, the liquid-phase concentration should increase stepwise, as shown in Figure 3(a) (the stepwise increase can be observed slightly but not as clearly as in Figure 3(a)). Moreover, it is unlikely that the concentration is constant due to solubility limitations, as it occurs after a high concentration of surface wash-off. Here, it is assumed that the intra-solid diffusion and surface wash-off mechanisms are independent of the liquid-phase concentration. However, in the actual leaching tests, the substances released from the solid are present in the liquid, and simultaneously, an interface equilibrium between the solid and liquid phases is always established. Figure 3(d) shows a case in which the interface equilibrium mainly governs the solid-liquid system. The larger the distribution coefficient,  $K_d$ , the more substances are adsorbed on the solid surface. Consequently, the liquid concentration remained almost constant (decreasing slightly). Figure 3(e) shows the case of a large  $K_d$  value with a small  $D_F$  value, where both effects were identified along with the water replacement schedule. Therefore, in the interpretation of the leaching test results, the effect of the solid-liquid interface equilibrium needs to be examined in detail. We constructed a new model that links intra-solid diffusion and solid-liquid interface equilibrium, termed the  $D_F$ - $K_d$  model, and examined the change in liquid-phase concentration in a single batch test (Sakanakura et al., 2021). All figures in Figure 3 were calculated by modifying the  $D_F$ - $K_d$  model program.



**Figure 3.** Examples of liquid concentration changes over a semi-dynamic leaching test based on the liquid replacement schedules of prEN 16637-2 and NEN 7375.

The outline of the model is as follows: A unit consists of a flat, one-dimensional solid with a certain thickness (Figure 4(a)) or multiple spheres with a single grain size (Figure 4(b)), and a certain volume

of liquid. The solid phase is divided into cells of arbitrary thickness, and the cell in contact with water at the solid-liquid interface is defined as the adsorption-desorption (“ad/desorption”) layer, at which the ad/desorption reaction proceeds instantaneously. In the solid phase, the diffusion of Equations (6) and (7) dominates for the flat model (Figure 4(a)) and the sphere model (Figure 4(b)), respectively.

$$\frac{\partial C_F}{\partial t} = D_F \frac{\partial^2 C_F}{\partial x^2} \quad (6)$$

$$\frac{\partial C_F}{\partial t} = \frac{D_F}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial C_F}{\partial r} \right) \quad (7)$$

where  $r$  represents the distance from the center of the sphere (m). Because the interface equilibrium is present only between the ad/desorption layer and liquid phase, Equation (2) should be rewritten as Equation (8).

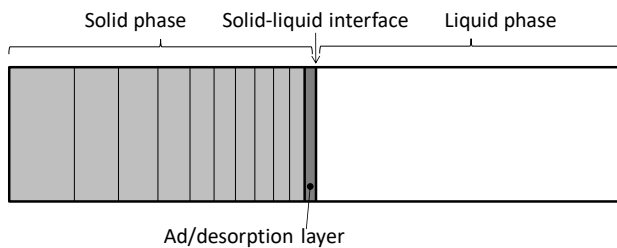
$$q_{AL} = K_{d(AL)} C_L \quad (8)$$

where  $q_{AL}$  is the adsorbed mass of a substance per mass of the ad/desorption layer (mg/kg), and  $K_{d(AL)}$  is the distribution coefficient between the ad/desorption layer and liquid phase.  $K_{d(AL)}$  should be calculated using Equation (9), as the substance involved in the interface equilibrium on the solid side exists only in the ad/desorption layer.

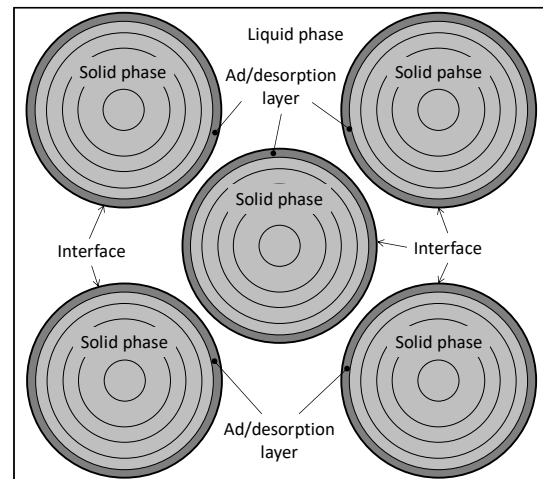
$$K_{d(AL)} = \frac{\rho_S V_S}{\rho_{AL} V_{AL}} K_d \quad (9)$$

where  $\rho_S$  and  $\rho_{AL}$  are the densities of the solid phase and the ad/desorption layer (kg/m<sup>3</sup>), respectively, and  $V_S$  and  $V_{AL}$  are the total volumes of the solid phase and the ad/desorption layer (m<sup>3</sup>), respectively. The liquid phase was assumed to be a completely mixed field. Based on the model, the mass transfer among cells in the solid phase, including the ad/desorption layer, is calculated by the finite volume method while maintaining the interface equilibrium between the ad/desorption layer and the liquid phase. This model was named the “ $D_F$ - $K_d$  model,” as the intra-solid diffusion and the interface equilibrium are linked (Sakanakura et al., 2022).

(a) Flat model



(b) Single diameter-sphere model



**Figure 4.** Schematic view of the  $D_F$ - $K_d$  model (Sakanakura et al., 2022)

#### 4.2 Reproduction of experimental data using the $D_F$ - $K_d$ model

The  $D_F$ - $K_d$  model was used to reproduce the SDLT results of a previous paper by Sakanakura (2005). In this study, the experiments were performed on both granules and monoliths. The liquid replacement schedules were 8 h, 1 d, 3 d, 7 d, 14 d, 28 d, and 56 d. In the calculation, the thickness of the ad/desorption layer was assumed to be 0.1  $\mu\text{m}$ . The initial concentration in the ad/desorption layer,  $C_{F,AL0}$ , was given separately from the initial concentration in the solid phase,  $C_{F0}$ , to represent surface wash-off.

The results in Figure 5 shows that the  $D_F$ - $K_d$  model can reproduce various leaching patterns by adjusting the parameters of the model. Figure 5(a) shows diffusion-controlled release of Mg with a low  $K_d$  value (0.001 L/kg). The solid sample was a glass-like granular material derived from melting of municipal solid waste, called "MSW slag", with a sample size of <2 mm. Figure 5(b) shows the Ca released from MSW slag. The approximated  $K_d$  value of 0.1 L/kg is higher than that of Mg. In Figure 5(b), a diffusion trend was observed, but the change in concentration was alleviated, as shown in Figure 5(a). Figure 5(c) shows surface wash-off followed by diffusion that can be reproduced by setting the concentration in the ad/desorption layer to 100 times higher than that of the solid phase. Figure 5(d) shows the case of a high  $D_F$  and low  $K_d$  value of molded fly ash solidified with cement. In this case, the depletion occurred from the first fraction. Additionally, in Figure 5(e) and 5(f), depletion is observed, and the higher the  $K_d$  value, the lower the change in the liquid-phase concentration.

Similar changes are observed depending on the combination of parameters. For example, a significantly low  $D_F$  and high  $K_d$  value can reproduce a monotonic decrease, as shown in Figure 5(d). Hence, it is necessary to obtain the  $D_F$  and  $K_d$  parameters together with other physical and chemical parameters, such as the sample size and initial concentration in the solid phase, using appropriate procedures. In addition, the thickness of the ad/desorption layer affects the calculation result, as the amount of substance retained inside the solid phase changes. Therefore, it is necessary to investigate the influence of the ad/desorption layer thickness on the  $D_F$ - $K_d$  model further, to develop an appropriate method for determining the related parameters.

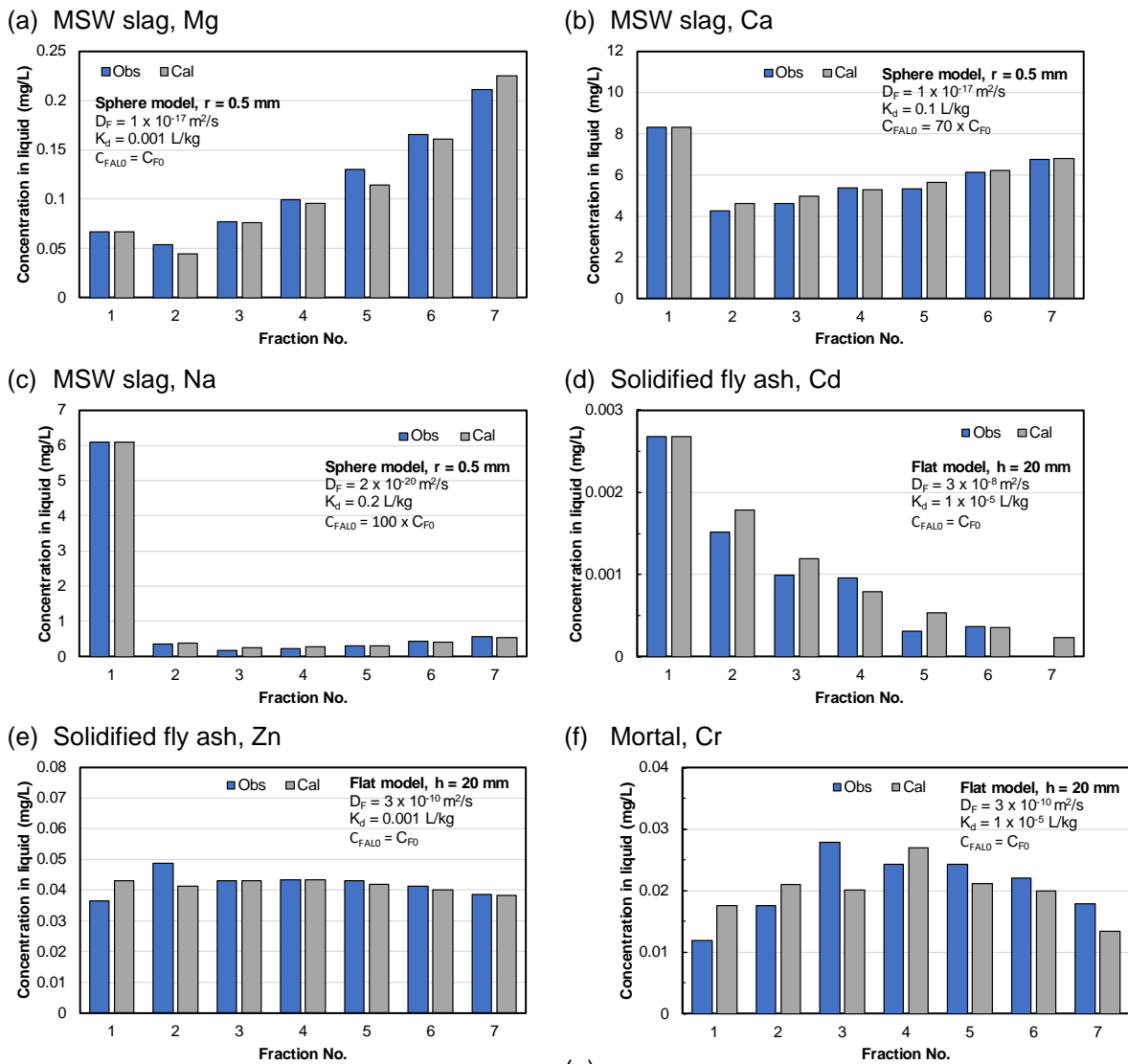


Figure 5. Reproduction of previous SDLT results (Sakanakura, 2005) by the  $D_F$ - $K_d$  model



## 5 CONCLUSION

SDLTs have been developed to determine leaching mechanisms based on changes in the liquid-phase concentration across fractions. The main leaching mechanisms dealt with in the SDLT standards are surface wash-off and intra-solid diffusion, although unknown mechanisms exist. This study establishes the relationship between the diffusion coefficient and elution depth. As the solid sample was finite, the leaching behavior approached depletion over time. Furthermore, we focused on the interface equilibrium that has not received sufficient attention in the field of leaching research. Using the  $D_F$ - $K_d$  model that links intra-solid diffusion and interface equilibrium, some of the previously unidentified leaching behaviors in the SDLT results could be reproduced. It is thus necessary to obtain the  $D_F$  and  $K_d$  parameter values, using suitable methods. The leaching behavior in an actual environment can then be estimated by combining the advection-dispersion and  $D_F$ - $K_d$  model.

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