

## Static Leaching Kinetics of $^{210}\text{Pb}$ from Uranium Tailings and Multi-factor Interaction Analysis

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### Abstract

Uranium tailings harbor the long-lived, highly biotoxic radionuclide  $^{210}\text{Pb}$  alongside associated heavy metals, posing persistent environmental risks. However, comprehensive systematic studies into the leaching mechanisms, kinetics, and interaction of key environmental factors on  $^{210}\text{Pb}$  release under static conditions remain critically lacking. To solve the above problems, in this study, the effects of pH, solid-to-liquid ratio,  $\text{H}_2\text{O}_2$  concentration, and NaCl concentration on  $^{210}\text{Pb}$  leaching were investigated using a custom-designed static leaching device, it was found that lower pH, reduced solid-to-liquid ratio, elevated  $\text{H}_2\text{O}_2$  concentration, and higher NaCl concentration are all conducive to enhancing  $^{210}\text{Pb}$  leaching rates. Mechanistic analysis reveals that lower pH elevates  $^{210}\text{Pb}$  leaching rates and solution-phase concentrations through mineral disintegration, increased specific surface area, and  $\text{H}^+$ -competitive displacement of  $\text{Pb}^{2+}$ . A lower solid-to-liquid ratio maintains a higher diffusion driving force, enhancing interfacial contact per unit mass of solid with leachant. High-concentration  $\text{H}_2\text{O}_2$  disrupts lead-bearing mineral structures via Strong oxidizing capacity, enhancing  $\text{Pb}^{2+}$  solubility and altering surface properties to promote desorption. NaCl accelerates  $^{210}\text{Pb}$  leaching via  $\text{Na}^+$ -competitive ion exchange and  $\text{Cl}^-$ -assisted chelation dissolution. The leaching kinetic model was established based on experimental data, then the effect of two-factor interaction on leaching rate was quantified using Box-Behnken design soft, identifying parameter combinations for maximal leaching rates. These findings provide a robust theoretical foundation for predicting the leaching and migration of hazardous elements in uranium tailings under static aquatic environment.

## Full Text

### Study on Static Leaching Kinetics of 210Pb from Uranium Tailings and Multi-factor Interaction Analysis

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## Abstract

Uranium tailings harbor the long-lived, highly biotoxic radionuclide 210Pb alongside associated heavy metals, posing persistent environmental risks. However, comprehensive systematic studies on the leaching mechanisms, kinetics, and interactive effects of key environmental factors on 210Pb release under static conditions remain critically lacking. To address these gaps, this study investigated the effects of pH, solid-to-liquid ratio, H<sub>2</sub>O<sub>2</sub> concentration, and NaCl concentration on 210Pb leaching using a custom-designed static leaching device. The results demonstrated that lower pH, reduced solid-to-liquid ratio, elevated H<sub>2</sub>O<sub>2</sub> concentration, and higher NaCl concentration all enhance 210Pb leaching rates. Mechanistic analysis reveals that low pH increases 210Pb leaching rates and solution-phase concentrations through mineral disintegration, increased specific surface area, and H<sup>+</sup>-competitive displacement of Pb<sup>2+</sup>. A lower solid-to-liquid ratio maintains a higher diffusion driving force, enhancing interfacial contact per unit mass of solid with the leachant. High-concentration H<sub>2</sub>O<sub>2</sub> disrupts lead-bearing mineral structures via strong oxidizing capacity, enhancing Pb<sup>2+</sup> solubility and altering surface properties to promote desorption. NaCl accelerates 210Pb leaching through Na<sup>+</sup>-competitive ion exchange and Cl<sup>-</sup>-assisted chelation dissolution. A leaching kinetic model was established based on experimental data, and the effects of two-factor interactions on leaching rate were quantified using Box-Behnken design software to identify parameter combinations for maximal leaching rates. These findings provide a robust theoretical foundation for predicting the leaching and migration of hazardous elements in uranium tailings under static aquatic environments.

**Keywords:** 210Pb Migration; Static Leaching; Leaching Kinetics; Interaction; Uranium Tailings

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## 1. Introduction

Uranium tailings, a byproduct of uranium mining and milling operations, pose persistent environmental risks due to radionuclides in the uranium decay series and co-occurring heavy metals (e.g., Cd, Pb, As) (Abdelouas, 2006). Stored in open hydrogeological systems, tailings undergo continuous water-rock interactions governed by mineral dissolution-precipitation equilibria, ion exchange, and colloid transport (Nordstrom, 2011), which mobilizes bound-phase heavy metals into freely dissolved species and significantly elevates contaminant migration rates (Martin et al., 2003). Field studies of decommissioned tailings ponds in South China demonstrate that precipitation-driven leachate release constitutes the primary pathway for radionuclide and heavy metal dispersion into surrounding aquifers and soils (Wang et al., 2018). Secondary pollution arising from this hydrogeochemical process now represents a critical challenge for environmental remediation at tailings sites (Chen et al., 2022).

Consequently, revealing the release kinetics and migration mechanisms of pollutants from uranium tailings under aqueous conditions is of significant theoretical importance for advancing pollution prevention and risk management in decommissioned tailings facilities. In recent years, research on pollutant release mechanisms from uranium tailings has predominantly focused on the hydrogeochemical behavior and dynamic migration patterns of major radionuclides such as  $^{238}\text{U}$  and  $^{226}\text{Ra}$  (Martin et al., 2003; Wang et al., 2018). Within the uranium decay chain, the key radionuclide  $^{210}\text{Pb}$  exhibits complex leaching mechanisms due to its long half-life (22.3 years), high biotoxicity, and intricate occurrence characteristics at the tailings-water interface.

Current studies on  $^{210}\text{Pb}$  leaching primarily investigate its behavior in soils, concentrating on the effects of leachate pH (Jing et al., 2004), organic chelating agents (citric acid (Ke et al., 2020), polyepoxysuccinic acid (Wu et al., 2025), oxalic acid (Ash et al., 2016)), inorganic stabilizing materials (phosphates (Fayiga and Saha, 2017), apatite (Guo et al., 2019), zeolite (Shi et al., 2013)), and soil properties (organic matter content (Chamorro and Sánchez-Andica, 2024), compaction (Xue et al., 2014)). Research specifically targeting  $^{210}\text{Pb}$  leaching from uranium tailings remains limited, having addressed only the influence of pH. Liu et al. (2020) demonstrated through column leaching experiments simulating rainwater percolation through uranium tailings at varying pH levels that  $^{210}\text{Pb}$  leaching is dissolution-controlled, with lower pH enhancing migration. Xie et al. (2023) conducted static leaching experiments on uranium tailings under different pH conditions and concluded that pH primarily influences leaching by altering the chemical speciation of  $^{210}\text{Pb}$  and the physicochemical properties of tailings particles. However, the leaching behavior and mechanisms of  $^{210}\text{Pb}$

under static immersion conditions, its leaching kinetics, and the interactive effects of critical environmental factors on its leaching rates still lack systematic and in-depth investigation.

To elucidate the static leaching behavior of  $^{210}\text{Pb}$  from uranium tailings under natural precipitation, this study systematically investigated the leaching characteristics and kinetics using fresh uranium tailings samples under various influencing factors. Single-factor influence experiments were conducted with a custom-designed static leaching device to investigate the leaching behavior and mechanisms of  $^{210}\text{Pb}$ . Subsequently, leaching kinetics analysis was performed based on the experimental results to establish a leaching kinetic model. Finally, response surface methodology was applied to study the interactive effects of key environmental factors on the  $^{210}\text{Pb}$  leaching rate and to identify the critical parameters for achieving maximum leaching rate. This research provides a theoretical basis for revealing the leaching and migration behavior of harmful metal elements in uranium tailings under static water immersion conditions.

## 2. Materials and Methods

### 2.1 Experimental Materials

Samples of tailings sand from the tailings pond and clay from the dam impermeable layer were selected. After air-drying, the tailings sand and clay samples were subjected to sample reduction via the quartering method. Mineral composition and trace element analysis were performed on representative specimens using X-ray diffraction (XRD), with results presented in Tables 1 and 2. As shown in Table 1, the tailings sand is predominantly composed of quartz (45–50%), while the clay consists mainly of gypsum (35–40%). Table 2 indicates that the uranium (U) content in the tailings sand is 1035 g/g and thorium (Th) content is 240 g/g, both values significantly exceeding the average soil concentrations of U (3.08 g/g) and Th (15.4 g/g). The samples were subjected to sequential extraction, and the Pb speciation data are presented in Table 3. The mobile fraction of  $^{210}\text{Pb}$  constitutes 22.2%.

**Table 1. Mineral composition of uranium tailings samples (%)**

Mineral component	Quartz	Plagioclase	K-feldspar	Illite	Kaolinite	Gypsum
Fine tailings sand	45~50	25~30	20~25	10~15	15~20	20~25
Clay	35~40	-	-	-	-	-

**Table 2. Elemental concentrations in uranium tailings samples (g/g)**

Element	Fine tailings sand
U	1035
Th	240

**Table 3. Speciation of Lead (Pb) in Uranium Tailings Residue Samples**

Water-soluble	Carbonate-exchangeable	Fe-Mn oxide-bound	Organic matter-bound	Residual
Percentage	1.5%	77.8%	-	-

## 2.2 Experimental Device

A custom-designed static leaching device was employed, with its structure illustrated in Figure 1 [Figure 1: see original paper]. The device measures 22 cm in length, 11.5 cm in width, and 14.5 cm in height. A partition is installed at 6.6 cm along the length of the device to prevent tailings particle entrainment during supernatant withdrawal. The upper portion of the partition features multiple small apertures to facilitate solution flow and diffusion on both sides. Additionally, two ventilation apertures at the top of the device allow for the creation of both aerobic and anaerobic leaching conditions.

## 2.3 Experimental Protocol

Four sets of single-factor experiments were conducted:

- (1) **Effect of pH:** Solutions with pH values of 3, 5, 7, 9, and 11 were accurately prepared by dropwise addition of HCl or NaOH solutions into the container. Subsequently, 100 g of tailings (particle size: 0.2 mm) was placed into a static leaching device, and 2000 mL of the prepared solution was added. After standing for a period, radionuclide concentrations in the supernatant were measured to determine the effects of different pH values and leaching durations.
- (2) **Effect of solid-to-liquid ratio:** Tailings with a particle size of 0.2 mm were selected. Portions of 200 g, 100 g, 66.7 g, and 50 g were placed into separate static leaching devices, and 2000 mL of deionized water was added to each. After standing for a period, radionuclide concentrations in the supernatant were measured to determine the effects of different leaching durations at solid-to-liquid ratios of 1:10, 1:20, 1:30, and 1:40.
- (3) **Effect of oxidizing conditions:** 100 g of ore tailings particles (0.2 mm) were placed into separate static leaching devices, and 2000 mL of H<sub>2</sub>O<sub>2</sub> solutions at concentrations of 0.05 mol/L, 0.01 mol/L, 0.5 mol/L, and 0.1

mol/L were added to each. The pH was adjusted to 7. After standing for a period, radionuclide concentrations in the supernatant were measured to determine the effects of different leaching durations under different oxidizing conditions.

- (4) **Effect of ionic strength:** 100 g of ore tailings particles (0.2 mm) were placed into separate static leaching devices, and 2000 mL of NaCl solutions at concentrations of 0.1 mol/L, 0.5 mol/L, and 1 mol/L were added to each. The pH was adjusted to 7. After standing for a period, radionuclide concentrations in the supernatant were measured to determine the effects of different leaching durations at different ionic strengths.

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### 3. Results and Discussion

#### 3.1 Leaching Behavior Under Different Conditions

##### Effect of pH

The variation of  $^{210}\text{Pb}$  activity concentration in the leachate with leaching time is illustrated in Fig. 2a [Figure 2: see original paper]. The curves indicate that under varying pH conditions, the  $^{210}\text{Pb}$  activity concentration in solution undergoes three distinct stages: rapid leaching, slow leaching, and leaching equilibrium. At identical leaching times, a higher pH value corresponds to a lower activity concentration of  $^{210}\text{Pb}$  in solution, with the activity concentration at pH 3 being significantly greater than that at pH 11. Fig. 2b [Figure 2: see original paper] demonstrates that an increase in pH value results in a decreased leaching rate of  $^{210}\text{Pb}$  at equilibrium. Comparative analysis reveals that lower solution pH yields higher  $^{210}\text{Pb}$  leaching concentration, a shorter duration to attain leaching equilibrium, and greater favorability for  $^{210}\text{Pb}$  leaching.

To further elucidate the experimental causation, the chemical speciation of Pb in the leachate and the characterization of the leached ore residue were analyzed. As depicted in Fig. 2c [Figure 2: see original paper], at  $\text{pH} < 3$ , Pb predominantly exists as  $\text{Pb}^{2+}$ ; between pH 3 and 7,  $\text{PbSO}_4$  precipitates and gradually becomes the dominant solid phase; between pH 7 and 10,  $\text{PbCO}_3$  emerges as the predominant precipitate phase; and between pH 10 and 11,  $\text{Pb}(\text{OH})_2$  serves as the primary precipitate phase, further inhibiting release. SEM images of post-leaching tailings (Fig. 3 [Figure 3: see original paper]) reveal that at  $\text{pH} = 3$ , the uranium tailings residue exhibits a fragmented morphology with well-developed microporosity, enhancing solid-liquid contact area and facilitating lead leaching. With increasing pH, tailings consolidation intensifies, accompanied by micropore closure, which impedes solution penetration and ion diffusion.

These concentration variations arise from the following mechanisms. At  $\text{pH} < 3$ , the strongly acidic environment promotes  $\text{H}^+$ -driven erosion of lead-bearing minerals, dissolving carbonate and oxide cementing agents and causing tailings disintegration into fragments. This increases the specific surface area of tailings

and opens diffusion channels. Concurrently,  $H^+$  competes with  $Pb^{2+}$  for adsorption sites, with high  $H^+$  concentrations occupying surface sites on tailings and inducing desorption of bound  $Pb^{2+}$ , thereby accelerating  $Pb^{2+}$  release (Owen et al., 2021). At  $3 < pH < 7$ , the acidic environment facilitates  $H^+$ -mediated sulfate dissolution in tailings, generating  $SO_4^{2-}$ , which combines with  $Pb^{2+}$  to form low-solubility  $PbSO_4$ , thereby reducing aqueous  $Pb^{2+}$  concentration. The leaching rate is thus governed by the combined kinetics of  $Pb^{2+}$  dissolution and  $PbSO_4$  precipitation (Cao et al., 2008). At  $7 < pH < 10$ , the alkaline environment drives  $OH^-$ -induced carbonate mineral dissolution, elevating  $CO_3^{2-}$  concentration and forming  $PbCO_3$ , which has lower solubility than  $PbSO_4$  (Xue et al., 2023). This precipitate further clogs tailings micropores and covers active sites on mineral surfaces, impeding solution contact with unreacted minerals. At  $10 < pH < 11$ , the conditional solubility product of  $Pb(OH)_2$  reaches its minimum, minimizing Pb solubility; high  $OH^-$  concentrations induce gel-like coagulation of  $Pb(OH)_2$ , exacerbating cementation (Wang et al., 2010).

### Effect of Solid-to-Liquid Ratio

Figure 4 [Figure 4: see original paper] shows the leaching behavior of 210Pb under varying solid-to-liquid ratios. As shown in Fig. 4a, with increasing leaching time, the 210Pb activity concentration in solution shows a rapid increase (0–120 h), a slow increase (120–600 h), and equilibrium (600–840 h). This is attributed to the dominance of readily accessible and soluble 210Pb on particle surfaces during 0–120 h, where dissolution rates are governed by surface chemical reaction kinetics, resulting in rapid concentration increase (Zhao et al., 2020). During 120–600 h, after depletion of surface-soluble components, leachate diffuses into the solid interior to react, and the resulting 210Pb ions diffuse from internal pores or lattices to particle surfaces, with dissolution rates controlled by solid-phase or pore diffusion mass transfer, significantly slowing the process (Owen et al., 2021). During 600–840 h, the leached 210Pb quantity approaches the maximum solubility achievable under the given solid-to-liquid ratio, reaching dissolution equilibrium.

Figure 4b shows that a lower solid-to-liquid ratio yields higher 210Pb leaching efficiency. Comparative analysis reveals that a higher solid-to-liquid ratio increases 210Pb concentration but decreases leaching efficiency. This occurs because a higher ratio indicates more solid particles per unit solution volume, which elevates the total 210Pb dissolved per unit time. However, high concentrations reduce the concentration gradient between particle surfaces and bulk solution, inhibiting 210Pb diffusion and causing incomplete dissolution. Concurrently, reduced leachate volume per unit solid mass promotes local saturation near particles, hindering further dissolution of internal 210Pb and lowering leaching efficiency (Fan et al., 2018). Conversely, a lower ratio corresponds to fewer solids per unit volume, reducing total 210Pb dissolution per unit time and maintaining a lower concentration gradient. This low-concentration environment sustains high diffusional driving force, enabling more complete reaction. With greater leachate volume per unit solid mass, a larger proportion of 210Pb

dissolves, increasing leaching efficiency. Given constant total reactant mass, the leachable  $^{210}\text{Pb}$  quantity is fixed, and sufficient leachate maximizes the concentration gradient, achieving theoretical maximum leaching efficiency (Owen et al., 2021).

### Effect of $\text{H}_2\text{O}_2$ Concentration

As shown in Fig. 5a [Figure 5: see original paper], comparison with the blank experiment (no  $\text{H}_2\text{O}_2$  added) demonstrates that oxidation of uranium tailings by  $\text{H}_2\text{O}_2$  increases the  $^{210}\text{Pb}$  activity concentration in solution by approximately 50% at equilibrium. The trend exhibits three sequential stages: rapid leaching, slow leaching, and leaching equilibrium. At identical leaching durations, higher  $\text{H}_2\text{O}_2$  concentrations result in higher  $^{210}\text{Pb}$  activity concentrations and leaching rates in solution, consistent with the leaching behavior of uranium in the tailings (Xie et al., 2024). The primary cause of this variation is the strong oxidizing action of  $\text{H}_2\text{O}_2$ , which (i) dissolves minerals, (ii) modifies mineral surface chemistry to promote  $^{210}\text{Pb}$  desorption, and (iii) elevates redox potential (Kurniawan et al., 2021), thereby enhancing  $\text{Pb}^{2+}$  solubility and the  $^{210}\text{Pb}$  activity concentration in solution.

Figure 5b [Figure 5: see original paper] shows that the increase in  $\text{H}_2\text{O}_2$  concentration is not proportional to increases in equilibrium  $^{210}\text{Pb}$  concentration and leaching rate. As  $\text{H}_2\text{O}_2$  concentration rises, the increases in equilibrium  $^{210}\text{Pb}$  concentration and leaching rate initially increase and subsequently decrease. This behavior is primarily attributed to the limited total amount of oxidizable  $^{210}\text{Pb}$ -bearing minerals present in the tailings. At low  $\text{H}_2\text{O}_2$  concentrations, the reaction is predominantly controlled by chemical reaction kinetics (Chen et al., 2024). With further increases in  $\text{H}_2\text{O}_2$  concentration, the reaction rate gradually diminishes, indicating a reduction in reaction order, as readily reactive mineral phases are progressively consumed, leaving only less reactive mineral phases. Consequently, even with additional increases in  $\text{H}_2\text{O}_2$  concentration, the dissolution rate becomes difficult to increase.

### Effect of $\text{NaCl}$ Concentration

The curve in Figure 6a [Figure 6: see original paper] shows that the  $^{210}\text{Pb}$  activity concentration increased sharply from 0 to 150 h, with the rate of increase slowing from 150 to 900 h as the system gradually approached equilibrium. Concurrently, the leaching process exhibited significant fluctuations throughout. This behavior is primarily attributed to the predominant adsorption of  $^{210}\text{Pb}$  onto clay mineral or iron-manganese oxide surfaces through electrostatic forces.  $\text{Na}^+$  acts as a competitive cation, displacing  $\text{Pb}^{2+}$  from adsorption sites and facilitating the rapid desorption of weakly bound  $\text{Pb}$  from tailings particle surfaces (Khalaji and Khanday, 2025; Hamid et al., 2021). During the initial experimental phase, higher  $\text{Na}^+$  concentrations were associated with greater  $\text{Pb}^{2+}$  release, as ion exchange predominated. In the later phase, strongly bound  $\text{Pb}$  or  $\text{Pb}$  encapsulated within the mineral lattice was slowly released. Subsequently,  $\text{Cl}^-$  formed soluble complexes with  $\text{Pb}^{2+}$  (e.g.,  $\text{PbCl}^+$ ,  $\text{PbCl}_2(\text{aq})$ ), promoting  $\text{Pb}$

dissolution from the solid phase. However, at high  $\text{Cl}^-$  concentrations, partial precipitation of  $\text{Pb}^{2+}$  as  $\text{PbCl}_2$  occurred. Specifically, when the  $\text{Cl}^-$  concentration exceeded 0.3 mol/L, lead-containing complex ions in solution partially precipitated as  $\text{PbCl}_2$ , inducing a decline in solution-phase  $^{210}\text{Pb}$  concentration and thereby generating significant fluctuations. At this stage, dissolution via complexation and the inhibition of precipitation collectively regulated the process (Cao et al., 2008).

To elucidate the effect of varying NaCl concentrations on  $^{210}\text{Pb}$ , the activity concentrations from Figure 6a were cumulatively summed, yielding the trend depicted in Figure 6b [Figure 6: see original paper]. This curve demonstrates that for identical time intervals, higher NaCl concentrations correlate with increased cumulative activity concentrations of  $^{210}\text{Pb}$  in solution, indicating elevated leaching rates and concentrations of  $^{210}\text{Pb}$ . Figure 6c [Figure 6: see original paper] corroborates this observation, showing that elevated NaCl concentrations result in greater leaching rates of  $^{210}\text{Pb}$ . These findings indicate that the stability of  $^{210}\text{Pb}$  in tailings is highly dependent on environmental ionic strength: under low ionic strength conditions (e.g., rainwater, freshwater),  $^{210}\text{Pb}$  is more firmly bound with lower release risk, whereas under high ionic strength conditions (e.g., seawater intrusion zones, saline-alkali soils, high-mineralization groundwater), desorption/dissolution is significantly enhanced, leading to a substantial increase in release risk (Attallah et al., 2020).

### 3.2 Leaching Kinetics Analysis

The leaching of radionuclides from uranium tailings primarily involves five sequential processes: diffusion of radionuclides to the tailings surface, adsorption onto the surface, surface reaction, desorption from the surface, and diffusion of products away from the interface. Consequently, diffusion, adsorption, chemical reaction, and desorption collectively influence the concentration of  $^{210}\text{Pb}$  in solution. To identify the dominant mechanism under varying leaching conditions, data were fitted using the pseudo-second-order kinetic model (Chiron et al., 2003; Baigenzhenov et al., 2020), shrinking core model (Hou et al., 2010; Chen et al., 2020), two-constant model (Lou et al., 2024; Jalali, 2006), and Avrami model (Yazawa et al., 2023). The fitting curves are presented in Figure 7 [Figure 7: see original paper], and the correlation coefficients are summarized in Tables 4 and 5.

**Table 4. Correlation coefficients  $R^2$  for fitting  $^{210}\text{Pb}$  leaching under different pH conditions using various kinetic models**

Model	Equation	$R^2$
Pseudo-second-order	$t/q = a + kt$	-
Shrinking core	$1-(1-\alpha)^{1/3} = kt$	-
Two-constant	$\ln(q) = a + k \cdot \ln(t)$	-
Avrami	$-\ln(1-\alpha) = kt^n$	-

Based on the data in Table 4 and the fitted curves, the pseudo-second-order kinetic model provides the best fit, indicating that under varying pH conditions, the rate of the 210Pb leaching process is governed by the chemical reaction step at the solid surface rather than the diffusion process. Consequently, the reaction rate is primarily determined by the chemical interaction of reactants at the solid-liquid interface.

**Table 5. R<sup>2</sup> values for the pseudo-second-order kinetic model fit of 210Pb leaching under different experimental conditions**

Condition	R <sup>2</sup>
Solid-to-liquid ratio	-
H <sub>2</sub> O <sub>2</sub> concentration	-
NaCl concentration	-

Table 5 shows the coefficient of determination for the pseudo-second-order kinetic model under different experimental conditions. The coefficients in Tables 4 and 5 indicate that the leaching process is controlled by chemical reactions on the solid surface. During the experiment, the rate constant  $k$  depends on factors including pH ( $\alpha$ ), solid-to-liquid ratio ( $\beta$ ), H<sub>2</sub>O<sub>2</sub> concentration ( $\omega$ ), and NaCl concentration ( $\rho$ ).

The overall reaction rate constant for the leaching of 210Pb can be expressed by equation (1) based on the Arrhenius equation (Li, 2021; Aghili et al., 2024):

$$k = C_0 \alpha^a \beta^b \omega^c \rho^d \quad (1)$$

where  $a$ ,  $b$ ,  $c$ , and  $d$  are the influence indices for pH ( $\alpha$ ), solid-to-liquid ratio ( $\beta$ ), H<sub>2</sub>O<sub>2</sub> concentration ( $\omega$ ), and NaCl concentration ( $\rho$ ), respectively. Taking the natural logarithm of both sides yields equation (2):

$$\ln k = \ln C_0 + a \ln \alpha + b \ln \beta + c \ln \omega + d \ln \rho \quad (2)$$

Plots of  $\ln k$  versus  $\ln \alpha$ ,  $\ln \beta$ ,  $\ln \omega$ , and  $\ln \rho$  were constructed, from which the following values were obtained:  $a = 0.1537$ ,  $b = 2.234$ ,  $c = -0.098$ ,  $d = -0.029$ . During the experiment, temperature  $T$  was held constant, leading to equation (3):

$$k = C_0 \alpha^{0.154} \beta^{2.234} \omega^{-0.098} \rho^{-0.029} \quad (3)$$

where  $C_0$  is a constant. Substituting equation (3) into the pseudo-second-order kinetic model gives equation (4):

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{t}{q_e} = a + C_0\alpha^{0.154}\beta^{2.234}\omega^{-0.098}\rho^{-0.029}t \quad (4)$$

All experimental data were substituted into Eq. (4) and fitted with  $\alpha^{0.154}\beta^{2.234}\omega^{-0.098}\rho^{-0.029}t$  on the x-axis and  $t/q$  on the y-axis, as shown in Fig. 8 [Figure 8: see original paper]. From the fitted data:  $C_0 = 48.008$ ,  $a = 4$ . Therefore, the leaching kinetic model is given by equation (5):

$$\frac{t}{q_t} = 4 + 48.008\alpha^{0.154}\beta^{2.234}\omega^{-0.098}\rho^{-0.029}t \quad (5)$$

### 3.3 Optimization Analysis of the Leaching Process

Box-Behnken Design (BBD) was employed to optimize the leaching process (Mazurek-Holys et al., 2025). The independent variables were pH, solid-to-liquid ratio,  $H_2O_2$  concentration, and NaCl concentration, with leaching rate as the response. The variable ranges were consistent with the single-factor experiments: pH 3-11, solid-to-liquid ratio 0.025-0.1,  $H_2O_2$  concentration 0-1 mol/L, and NaCl concentration 0-1 mol/L. Experiments were conducted sequentially according to the software-generated scheme, and results are presented in Table 6.

**Table 6. Box-Behnken Design Response Surface Model Experimental Design and Results**

Experiment	pH	Solid-to-liquid ratio	$H_2O_2$ Concentration (mol/L)	NaCl concentration (mol/L)	Leaching rate (%)
1-15	-	-	0.5	-	-
16-29	-	0.025-0.1	0.5	-	-

Analysis of variance and significance tests were performed on the established polynomial model for leaching rate, as shown in Table 7. The response surface model exhibited an F-value of 50.81 and  $P < 0.0001$ , indicating that the model of 210Pb leaching rate is statistically significant overall and effectively explains the variation in the response variable. Both the coefficient of determination ( $R^2 = 0.9807$ ) and the adjusted  $R^2$  (0.9614) were close to 1, demonstrating excellent model fit and the ability to capture predominant patterns in the data. The signal-to-noise ratio of 27.2907 indicates adequate precision. The difference between  $R^2$  and adjusted  $R^2$  was less than 0.2, confirming that the number of independent variables matches the sample size, thereby ensuring strong explanatory and predictive power of the model for the experimental data.

Analysis of the sum of squares revealed that the four factors influencing the 210Pb leaching rate, in descending order of influence, are solid-to-liquid ratio,

H<sub>2</sub>O<sub>2</sub> concentration, pH, and NaCl concentration. All two-way interactions among factors were not significant, while for the quadratic terms, only the H<sub>2</sub>O<sub>2</sub> concentration term (C<sup>2</sup>) was significant, indicating a nonlinear effect of H<sub>2</sub>O<sub>2</sub> concentration. The difference between experimental and predicted leaching rates ranged from -0.15 to +0.05, reflecting minimal prediction error.

**Table 7. Analysis of Variance Results for the Response Surface Model**

Source	Sum of Squares	df	Mean Square	F-value	p-value
Model	-	-	-	50.81	< 0.0001
A-pH	-	-	-	-	< 0.0001
B-Solid-to-Liquid Ratio	-	-	-	-	< 0.0001
C-H <sub>2</sub> O <sub>2</sub> Concentration	-	-	-	-	< 0.0001
D-NaCl Concentration	-	-	-	-	< 0.0001
Residual	-	-	-	-	-
Lack of Fit	-	-	-	-	-
Pure Error	-	-	-	-	-
Cor Total	-	-	-	-	-

Based on the leaching rate polynomial model, any two of the factors—pH, solid-to-liquid ratio, H<sub>2</sub>O<sub>2</sub> concentration, and NaCl concentration—were selected as independent variables, with the remaining two factors set to the midpoint values of the experimental design. The 3D response surface and corresponding contour plots for Pb leaching rate are shown in Fig. 9 [Figure 9: see original paper].

Figures 9a and 9b show the effects of pH and solid-to-liquid ratio on 210Pb leaching rate at 0.5 mol/L H<sub>2</sub>O<sub>2</sub> and 0.5 mol/L NaCl. In Fig. 9a, with solid-to-liquid ratio fixed at 0.025, pH ranged from 3 to 11 and leaching rate varied from 1.38 to 1.82. When pH was fixed at 11, solid-to-liquid ratio ranged from 0.025 to 0.1 and leaching rate varied from 0.72 to 1.38. The smaller variation range in leaching rate for pH versus solid-to-liquid ratio indicates that solid-to-liquid ratio exerts a stronger influence on leaching rate in the interaction experiment. In Fig. 9b, the smooth curvature of the contour lines suggests a weak interaction effect between pH and solid-to-liquid ratio on leaching rate.

Figures 9c and 9d show the effects of pH and H<sub>2</sub>O<sub>2</sub> concentration on Pb leaching rate at a solid-to-liquid ratio of 0.0625 and 0.5 mol/L NaCl. In Fig. 9c, with H<sub>2</sub>O<sub>2</sub> concentration fixed at 0 mol/L, pH ranged from 3 to 11 and leaching rate varied from 0.768 to 1.053. When pH was fixed at 11, H<sub>2</sub>O<sub>2</sub> concentration ranged from 0 to 1 mol/L and leaching rate exhibited a distinct trend of initial increase followed by decrease, varying from 0.768 to 1.124. The smaller variation range in leaching rate for pH versus H<sub>2</sub>O<sub>2</sub> concentration indicates that H<sub>2</sub>O<sub>2</sub> exerts a stronger influence on leaching rate in the interaction experiment. In Fig. 9d, the pronounced curvature of the contour lines suggests a strong interaction effect between pH and H<sub>2</sub>O<sub>2</sub> concentration on leaching rate.

Figures 9e and 9f show the effects of pH and NaCl concentration on Pb leaching rate at a solid-to-liquid ratio of 0.0625 and 0.5 mol/L  $\text{H}_2\text{O}_2$ . In Fig. 9e, with NaCl concentration fixed at 0 mol/L, pH ranged from 3 to 11 and leaching rate exhibited pronounced curvature, varying from 1.036 to 1.279. When pH was fixed at 11, NaCl concentration ranged from 0 to 1 mol/L and leaching rate varied from 1.036 to 1.180. The larger variation range in leaching rate for pH versus NaCl concentration indicates that pH exerts a stronger influence on leaching rate in the interaction experiment. In Fig. 9f, the smooth curvature of the contour lines suggests a weak interaction effect between pH and NaCl concentration on leaching rate.

Figures 9g and 9h show the effects of solid-to-liquid ratio and  $\text{H}_2\text{O}_2$  concentration on leaching rate at pH 7 and 0.5 mol/L NaCl. In Fig. 9g, with  $\text{H}_2\text{O}_2$  concentration fixed at 0 mol/L, solid-to-liquid ratio ranged from 0.025 to 0.1 and leaching rate varied from 0.560 to 1.277. When solid-to-liquid ratio was fixed at 0.1,  $\text{H}_2\text{O}_2$  concentration ranged from 0.025 to 0.1 mol/L and leaching rate varied from 0.560 to 0.919, exhibiting pronounced curvature. The larger variation range in leaching rate for solid-to-liquid ratio versus  $\text{H}_2\text{O}_2$  concentration indicates that solid-to-liquid ratio exerts a stronger influence on leaching rate in the interaction experiment, and the leaching rate demonstrates a nonlinear relationship with  $\text{H}_2\text{O}_2$  concentration. In Fig. 9h, the smooth curvature of the contour lines suggests a weak interaction effect between solid-to-liquid ratio and  $\text{H}_2\text{O}_2$  concentration on leaching rate.

Figures 9i and 9j show the effects of solid-to-liquid ratio and NaCl concentration on leaching rate at pH 7 and 0.5 mol/L  $\text{H}_2\text{O}_2$ . In Fig. 9i, with NaCl concentration fixed at 0 mol/L, solid-to-liquid ratio ranged from 0.025 to 0.1 and leaching rate varied from 0.785 to 1.468. When solid-to-liquid ratio was fixed at 0.1, NaCl concentration ranged from 0 to 1 mol/L and leaching rate varied from 0.785 to 0.980. The larger variation range in leaching rate for solid-to-liquid ratio versus NaCl concentration indicates that solid-to-liquid ratio exerts a stronger influence on leaching rate in the interaction experiment. In Fig. 9j, the straight-line pattern of the contour lines suggests no significant interaction effect between solid-to-liquid ratio and NaCl concentration on leaching rate.

Figures 9k and 9m show the effects of  $\text{H}_2\text{O}_2$  and NaCl concentration on  $^{210}\text{Pb}$  leaching rate at pH 7 and a solid-to-liquid ratio of 0.0625. In Fig. 9k, with NaCl concentration fixed at 0 mol/L,  $\text{H}_2\text{O}_2$  concentration ranged from 0 to 1 mol/L and leaching rate varied from 0.8680 to 1.3368, exhibiting pronounced curvature. When  $\text{H}_2\text{O}_2$  concentration was fixed at 1 mol/L, NaCl concentration ranged from 0 to 1 mol/L and leaching rate varied from 1.2782 to 1.4540. The larger variation range in leaching rate for  $\text{H}_2\text{O}_2$  versus NaCl concentration indicates that  $\text{H}_2\text{O}_2$  exerts a stronger influence on leaching rate in the interaction experiment. In Fig. 9m, pronounced curvature of the contour lines suggests a strong interaction effect between  $\text{H}_2\text{O}_2$  and NaCl concentration on leaching rate.

Comparison of contour line curvature across Fig. 9 reveals that the curvature

magnitude decreases in the order (m), (d), (h), (f), (b), (j), indicating a strong-to-weak interaction effect sequence of:  $\text{H}_2\text{O}_2$  and NaCl concentration, pH and  $\text{H}_2\text{O}_2$  concentration, solid-to-liquid ratio and  $\text{H}_2\text{O}_2$  concentration, pH and NaCl concentration, pH and solid-to-liquid ratio, solid-to-liquid ratio and NaCl concentration on leaching rate. Model optimization with leaching rate maximized at 100% yielded optimal experimental conditions: pH 3, solid-to-liquid ratio of 0.025,  $\text{H}_2\text{O}_2$  concentration at 0.935 mol/L, NaCl concentration at 1 mol/L, achieving a leaching rate of 2.064%.

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## 4. Conclusions

This study, based on a self-made static leaching experimental setup, systematically investigated the leaching behavior and mechanism, leaching kinetics, and interactive effects of multiple environmental factors on 210Pb in uranium tailings under static aqueous conditions. The main conclusions are as follows:

- (1) Environmental factors exert a decisive influence on the behavior and mechanism of 210Pb leaching. Under low-pH conditions (particularly  $\text{pH} < 3$ ), the leaching rate and final concentration of 210Pb are significantly enhanced through the promotion of mineral disintegration, an increase in the specific surface area for reaction, and competitive adsorption by  $\text{H}^+$  ions. A low solid-to-liquid ratio maintains a high concentration gradient diffusion driving force, facilitating greater contact between the unit mass of tailings and the leaching solution, thereby improving leaching efficiency. The strong oxidizing property of  $\text{H}_2\text{O}_2$  effectively promotes 210Pb release by disrupting the crystal structure of lead-bearing minerals and altering mineral surface properties; however, its enhancing effect exhibits a nonlinear trend with an initial increase followed by a decrease as concentration increases. NaCl primarily promotes 210Pb leaching synergistically through ion exchange by  $\text{Na}^+$  and complexation by  $\text{Cl}^-$ ; yet, at high concentrations ( $>0.3$  mol/L), fluctuations in leaching concentration arise due to  $\text{PbCl}_2$  precipitation.
- (2) Leaching kinetics analysis demonstrates that leaching behavior of 210Pb is best described by the pseudo-second-order kinetic model, with the rate primarily controlled by the chemical reaction step at the solid-liquid interface rather than diffusion. Building on this model, an empirical leaching kinetics model incorporating pH, solid-to-liquid ratio,  $\text{H}_2\text{O}_2$  concentration, and NaCl concentration as key parameters was established, which effectively predicts leaching behavior under diverse environmental conditions.
- (3) The relative magnitudes of the effects of various factors on leaching rate and their interaction effects were determined. The order of influence on 210Pb leaching rate, from greatest to least, was: solid-to-liquid ratio  $>$   $\text{H}_2\text{O}_2$  concentration  $>$  pH  $>$  NaCl concentration. Response surface analysis indicated strong interactive effects between  $\text{H}_2\text{O}_2$  concentration and

NaCl concentration, and between pH and H<sub>2</sub>O<sub>2</sub> concentration, while other factor interactions were relatively weak. Through model optimization, the optimal parameters to achieve the maximum leaching rate (2.064%) were determined as: pH = 3, solid-to-liquid ratio = 0.025, H<sub>2</sub>O<sub>2</sub> concentration = 0.935 mol/L, NaCl concentration = 1 mol/L.

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### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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