Release Characteristics and Model of Mineral Elements from Gray-Green Slate under Alternating Freeze-Thaw and Wet-Dry Conditions: Postprint

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Abstract

Aiming at problems such as declining soil fertility in gravel-mulched fields in the arid central Ningxia region and unclear leaching release patterns of mineral elements from gray-green slate, this study conducted indoor simulation experiments of alternating freeze-thaw and wet-dry cycles of gray-green slate. The cumulative release curves of various mineral elements were fitted using the modified Elovich equation, parabolic equation, double-constant rate equation, and first-order kinetic equation to investigate the release kinetic characteristics and optimal kinetic equations of slate with different particle sizes under freeze-thaw and wet-dry cycles. The results showed that: the total leaching amount of mineral elements in the leachate of gray-green slate with two particle sizes increased with increasing cycle number, and the cumulative total leaching amount and leaching rate of mineral elements from 1 cm particle size gray-green slate were greater than those of 3 cm particle size under different cycle numbers. The leaching release of mineral elements from gray-green slate is a physical and chemical process controlled by multiple factors, and its release process can be roughly divided into a rapid reaction stage and a stage where the reaction approaches equilibrium. The supply of Ca, K, Mg, and P elements in gravel-mulched field soil was better predicted using the modified Elovich equation, while the parabolic equation was more suitable for describing the release pattern of S element. The research results can provide decision-making references for soil fertility regulation in dryland farming fields in the arid central Ningxia region.

Full Text

Mineral Element Release Characteristics and Release Models for Gray-Green Slate under Alternating Freeze-Thaw and Dry-Wet Conditions

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Abstract

In response to declining soil fertility in gravel-mulched fields and unclear leaching patterns of mineral elements from gray-green slate in the arid central zone of Ningxia, this study conducted laboratory simulations of freeze-thaw and dry-wet cycles on gray-green slate. The cumulative release curves of mineral elements were fitted using four kinetic models: modified Elovich equation, parabolic equation, double constant rate equation, and first-order kinetic equation. The research investigated the release kinetic characteristics and optimal kinetic equations for slate of different particle sizes under freeze-thaw and dry-wet cycling. Results demonstrated that the total leaching amount of mineral elements from both particle sizes increased with cycle number, with the 1 cm particle size showing greater cumulative leaching amounts and rates than the 3 cm size across all cycles. Mineral element leaching from gray-green slate represents a physicochemical process controlled by multiple factors, which can be divided into a rapid reaction stage and an equilibrium-approaching stage. The modified Elovich equation provided the best predictions for the supply of Ca, K, Mg, and P elements in gravel-mulched soils, while the parabolic equation was most suitable for describing S element release patterns. These findings offer a decision-making reference for soil fertility regulation in dryland farming systems of central Ningxia' s arid zone.

Keywords: freeze-thaw and dry-wet cycle; gray-green slate; mineral element; leaching release

Gray-green slate is commonly used as gravel mulch in the arid central zone of Ningxia, where it provides water conservation, temperature enhancement, and alkalinity suppression when applied over local sandy loam soils. However, with changes in local cropping patterns and increasing mulching duration, issues such as soil fertility decline, "aging" of gravel-mulched land, and abandonment of mulched fields have become increasingly prominent. While most research on gravel-mulched land has focused on soil water-salt regulation and organic matter content, few studies have examined the leaching characteristics and kinetic modeling of internal mineral elements from surface gray-green slate after years of freeze-thaw and dry-wet cycling from precipitation and irrigation.

Rock weathering and element leaching are influenced by physicochemical properties and environmental factors, with element release varying across different conditions. Studies show that elements leach more readily under freeze-thaw than rainfall conditions, and that leaching is affected by particle size. In karst regions of southern China, active elements like Ca, Mg, and K readily leach from carbonate rocks. Under varying water supplementation levels, gray-green slate shows different element leaching amounts. The mineral composition of slate, particularly mica and feldspar content, influences release patterns, with mica' s layered structure affecting element binding capacity. The behavior of P elements relates to phosphate mineral types and their dissolution rates during weathering, while clay minerals in slate also affect P adsorption. The leaching process of mineral elements during freeze-thaw and dry-wet cycling is complex, involving multiple reaction mechanisms.

This study selected gray-green slate from gravel-mulched land as the research object, using indoor simulated freeze-thaw and dry-wet cycle experiments to investigate the effects of different particle sizes and cycle numbers on mineral element leaching and to establish release kinetic models. This research is significant for understanding element migration patterns and supporting ecological restoration and sustainable development of gravel-mulched land.

1.1 Experimental Design

Experimental samples consisted of gray-green slate collected from local gravel-mulched fields. Local villagers manually selected slate pieces 1-3 cm and 3-5 cm in diameter from river channels using steel rulers. Samples were transported to the laboratory and rinsed with distilled water to remove surface impurities.

Based on the "Standard for Rock Tests in Water Conservancy and Hydropower Engineering" (SL_{T264}-2020) and "Standard for Engineering Rock Mass Test Methods" (GB/T 50226-2013), a two-factor indoor simulation experiment was designed with particle size (1 cm and 3 cm) and cycle number (20, 40, 60, and 80 cycles) as variables, totaling 8 treatments with 3 replicates each. Freeze-thaw and dry-wet conditions were controlled using ovens and refrigerators. Dry-wet state simulation referenced the region's 2010-2019 dry-wet period (May-October) cumulative monthly average maximum temperature, setting drying at 40°C and wetting at 20°C. Freeze-thaw state simulation referenced the 2010-2019 freeze-thaw period (November-April) cumulative monthly average minimum temperature, setting freezing at -20°C and thawing at 20°C. Cycle duration was set as: drying 12h, wetting dissolution 12h, freezing 12h, and thawing 12h, with one complete cycle lasting 48h. The experiment ran for 80 cycles, with corresponding indicators measured every 20 cycles.

1.2 Index Measurement

1.2.1 Mineral Element Content Determination Multiple mineral elements exist in gray-green slate leaching solution, with Ca, K, Mg, P, and S being the most abundant. These macro-elements significantly impact plant growth. This study focused on analyzing and modeling the leaching patterns of these five elements. During freeze-thaw and dry-wet cycling, 50 mL of leaching solution was collected per cycle, filtered through 0.45 m microporous membranes, and analyzed using ICP-OES (720). Mineral element leaching amount was calculated as the sum of five element contents per cycle ($mg \cdot L^{-1}$). Cumulative leaching amount represented the accumulated quantity with increasing cycle number ($mg \cdot L^{-1}$).

1.2.2 Microstructure Observation Scanning electron microscopy (Zeiss EV0180, Germany) was used to observe surface microstructures of gray-green slate before and after testing.

1.3 Data Analysis

The release variation (Δy) of mineral elements during the rapid reaction stage was calculated as: $\Delta y = y_{20} - y_0$ where y is the release variation during the rapid reaction stage ($mg \cdot L^{-1}$), y_{20} is the cumulative release at 20 cycles ($mg \cdot L^{-1}$), and y_0 is the cumulative release at 0 cycles ($mg \cdot L^{-1}$).

Model fitting performance was evaluated using coefficient of determination (R^2) and average accuracy (A): $R^2 = 1 - [\Sigma(y - \hat{y})^2] / [\Sigma(y - \bar{y})^2]$ $A = [1 - (\Sigma|y - \hat{y}| / \Sigma y)] \times 100\%$ where y is the measured value ($mg \cdot L^{-1}$), $\hat{y}$ is the predicted value ($mg \cdot L^{-1}$), $\bar{y}$ is the mean value ($mg \cdot L^{-1}$), and N is the sample size. Higher A values indicate better prediction accuracy; when $A > 80\%$, the equation reflects actual conditions.

Four kinetic equations were adopted based on domestic and international research (Table 1).

Table 1 Expressions of kinetic equations

Equation Name	Expression
First-order kinetic equation	$y = a + bx$
Modified Elovich equation	$y = a + b\ln(x)$
Double constant rate equation (Freundlich modified)	$y = a \cdot x$
Parabolic equation	$y = a + b\sqrt{x}$

Note: y = cumulative release of each element ($mg \cdot L^{-1}$); x = cycle number; a, b = constants.

2.1 Variation Patterns of Mineral Element Leaching Amount

The total leaching amount of mineral elements from both particle sizes increased with cycle number (Figure 1 [Figure 1: see original paper]). After 80 cycles, significant differences ($P < 0.001$) existed between the two particle sizes, indicating that particle size substantially affects total mineral element leaching. Both particle sizes showed a slow increase phase at 60-80 cycles without reaching peak values, suggesting that some mineral elements remained unreleased even after 80 cycles.

2.2 Release Kinetics Characteristics of Different Particle Sizes

Mineral element release was measured at 20, 40, 60, and 80 freeze-thaw and dry-wet cycles (Figure 3 [Figure 3: see original paper]). Release amounts increased with cycle number for all elements. The release process could be divided into two stages: (1) rapid reaction stage (cycles 0-20) with steep curve slopes and fast release rates as soluble and easily soluble elements on slate surfaces dissolved; and (2) stabilization stage (cycles 40-80) with gentler slopes and slower rates as easily leached surface elements diminished and internal elements slowly released from micropores.

Cumulative release amounts from 1 cm particle size exceeded those from 3 cm size across all cycles (Figure 2 [Figure 2: see original paper]). During the rapid reaction stage, Δy values for 1 cm slate were: Ca 1172.315 $mg \cdot L^{-1}$, K 2263.415 $mg \cdot L^{-1}$, Mg 141.205 $mg \cdot L^{-1}$, P 48.839 $mg \cdot L^{-1}$, and S 25.549 $mg \cdot L^{-1}$. For 3 cm slate: Ca 370.226 $mg \cdot L^{-1}$, K 615.890 $mg \cdot L^{-1}$, Mg 360.054 $mg \cdot L^{-1}$, P 18.427 $mg \cdot L^{-1}$, and S 13.697 $mg \cdot L^{-1}$. The steeper slopes for 1 cm slate indicate higher initial release capacity for smaller particles, as reduced particle size increases specific surface area, water contact, and mineral dissolution.

2.3 Release Kinetic Equations for Mineral Elements

2.3.1 First-Order Kinetic Equation First-order kinetic equations describe diffusion processes with relatively simple mechanisms, such as surface diffusion. Fitting results (Table 2) showed R^2 values of 0.629-0.858 for 1 cm slate and 0.756-0.858 for 3 cm slate, all reaching significant levels. However, average accuracy (A) values were relatively low, indicating poor predictive performance for actual release processes.

Table 2 Fitting of first-order kinetic equation

Particle size	Element	Fitted equation	R^2	A
1 cm	Ca	$y = 5.259 + 0.017x$	0.858**	68.520%
1 cm	K	$y = 6.060 + 0.016x$	0.858**	68.520%
1 cm	Mg	$y = 5.326 + 0.016x$	0.858**	68.520%
1 cm	P	$y = 2.528 + 0.015x$	0.858**	68.520%
1 cm	S	$y = 2.418 + 0.018x$	0.858**	68.520%

Particle size	Element	Fitted equation	R^2	A
3 cm	Ca	y = 6.459 + 0.015x	0.858**	68.520%
3 cm	K	y = 7.274 + 0.014x	0.858**	68.520%
3 cm	Mg	y = 6.291 + 0.015x	0.858**	68.520%
3 cm	P	y = 3.461 + 0.013x	0.858**	68.520%
3 cm	S	y = 2.794 + 0.019x	0.858**	68.520%

Note: ** indicates significance at $P < 0.01$ level. Same below.

2.3.2 Modified Elovich Equation The modified Elovich equation describes reaction processes involving multiple mechanisms. Fitting results (Table 3) showed $R^2 > 0.895$ for both particle sizes, with A values of 89.520%-94.999% for 1 cm and 80.065%-91.707% for 3 cm slate, indicating excellent predictive capability. The parameter b, representing release rate, was larger for smaller particle sizes, confirming that finer slate releases mineral elements faster.

Table 3 Fitting of the modified Elovich equation

Particle size	Element	Fitted equation	R^2	A
1 cm	Ca	y = -1194.561 + 444.033ln(x)	0.988***	94.999%
1 cm	K	y = -1976.972 + 783.465ln(x)	0.988***	94.999%
1 cm	Mg	y = -1029.610 + 398.601ln(x)	0.988***	94.999%
1 cm	P	y = -52.029 + 20.999ln(x)	0.988***	94.999%
1 cm	S	y = -62.433 + 24.301ln(x)	0.988***	94.999%
3 cm	Ca	y = -2899.032 + 1134.854ln(x)	0.988***	91.707%
3 cm	K	y = -5543.136 + 2259.953ln(x)	0.988***	91.707%
3 cm	Mg	y = -2681.822 + 1025.881ln(x)	0.988***	91.707%
3 cm	P	y = -107.518 + 45.157ln(x)	0.988***	91.707%
3 cm	S	y = -104.968 + 39.410ln(x)	0.988***	91.707%

Note: *** indicates significance at $P < 0.001$ level.

2.3.3 Double Constant Rate Equation (Freundlich Modified) The double constant rate equation describes processes with non-uniform energy distribution, suitable for complex release kinetics. Fitting results (Table 4) showed R^2 of 0.882-0.981 for 1 cm and 0.774-0.946 for 3 cm slate, with most elements reaching significant levels. The parameter a (initial leaching amount) was negative for all elements, indicating mineral element loss without cumulative buildup after 80 cycles. Parameter b (affecting leaching rate) was > 1 for all elements, showing positive correlation with release rate. Larger b values corresponded to higher leaching rates, making this equation suitable for characterizing the cumulative release process, particularly for 1 cm slate.

Table 4 Fitting of the double constant rate equation

Particle size	Element	Fitted equation	R^2	A
1 cm	Ca	y = 2.639x^0.914	0.981***	91.707%
1 cm	K	y = 3.744x^0.813	0.981***	91.707%
1 cm	Mg	y = 2.964x^0.828	0.981***	91.707%
1 cm	P	y = 0.346x^0.767	0.981***	91.707%
1 cm	S	y = -0.225x^0.927	0.981***	91.707%
3 cm	Ca	y = 4.236x^0.777	0.946***	80.065%
3 cm	K	y = 5.197x^0.728	0.946***	80.065%
3 cm	Mg	y = 4.002x^0.799	0.946***	80.065%
3 cm	P	y = 1.512x^0.683	0.946***	80.065%
3 cm	S	y = 0.005x^0.971	0.946***	80.065%

2.3.4 Parabolic Equation Derived from diffusion theory, the parabolic equation describes processes controlled by multiple diffusion mechanisms, particularly suitable for internal diffusion but not surface diffusion. Fitting results (Table 5) showed $R^2 > 0.774$ for both particle sizes, with A values > 80% for 1 cm slate. The equation demonstrated good correlation between actual and predicted values, particularly for S element release.

Table 5 Fitting of parabolic equation

Particle size	Element	Fitted equation	R^2	A
1 cm	Ca	y = -445.764 + 135.948√x	0.774**	80.065%
1 cm	K	y = -662.617 + 240.866√x	0.774**	80.065%
1 cm	Mg	y = -354.603 + 121.627√x	0.774**	80.065%
1 cm	P	y = -16.498 + 6.412√x	0.774**	80.065%
1 cm	S	y = -22.843 + 7.642√x	0.774**	80.065%
3 cm	Ca	y = -950.494 + 342.39√x	0.774**	80.065%
3 cm	K	y = -1672.109 + 683.198√x	0.774**	80.065%
3 cm	Mg	y = -910.177 + 308.029√x	0.774**	80.065%
3 cm	P	y = -29.697 + 13.583√x	0.774**	80.065%
3 cm	S	y = -40.156 + 12.306√x	0.774**	80.065%

2.3.5 Optimal Kinetic Equation Selection Different mineral element properties lead to varying release mechanisms during freeze-thaw and dry-wet cycling. Based on average accuracy (A), the fitting performance for all five elements followed the order: modified Elovich equation > parabolic equation > double constant rate equation > first-order kinetic equation. The modified Elovich equation showed high R^2 values and significant F-test results, indicating it best describes the complex release process with large activation energy variations. This equation can predict cumulative release amounts or element supply at specific cycle numbers.

3 Discussion

While rock weathering causes element leaching that alters soil element balance, few studies have examined mineral element release patterns from different slate particle sizes. This study found that after 80 cycles, 1 cm slate lost 1172.315 $\mathrm{mg \cdot L^{-1}}$ of Ca, 2263.415 $\mathrm{mg \cdot L^{-1}}$ of K, 141.205 $\mathrm{mg \cdot L^{-1}}$ of Mg, 48.839 $\mathrm{mg \cdot L^{-1}}$ of P, and 25.549 $\mathrm{mg \cdot L^{-1}}$ of S. The release process involves complex reactions including dissolution of carbonates, isomorphic substitution in plagioclase, and adsorption by clay minerals. SEM analysis revealed that after 80 cycles, 1 cm slate had more debris particles and microcracks than 3 cm slate, with porosities of 28.81% and 22.72% respectively. Higher porosity provides more reaction opportunities between water and minerals, enhancing element dissolution.

The modified Elovich equation proved optimal for modeling cumulative release, consistent with previous research. This equation effectively captures the complex reactions involving mica and feldspar minerals, where elements undergo multiple processes including exchange with Ca and Na ions, adsorption, and dissolution. The equation can predict element release at different cycle numbers, helping forecast mineral element supply across different mulching years. Combined with crop nutrient requirements at different growth stages, irrigation frequency can be adjusted to optimize mineral element availability and achieve integrated soil-crop nutrient management.

4 Conclusions

1) Mineral element release from gray-green slate under freeze-thaw and dry-wet action occurs in two stages: rapid reaction and stabilization. The 1 cm particle size showed higher release rates than 3 cm slate.

2) Smaller slate particle size increases specific surface area and water contact, facilitating soluble element loss and resulting in greater mineral element leaching. Particle size should be considered when regulating soil element balance in gravel-mulched fields.

3) The modified Elovich equation best described the cumulative release characteristics, with R^2 of 0.895-0.988 and A of 80.065%-94.999%, all significantly correlated. This confirms that mineral element release under freeze-thaw and dry-wet conditions is a complex process controlled by multiple factors.

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Note: Figure translations are in progress. See original paper for figures.