

Mechanisms of salt rejection at the ice-liquid interface during the freezing of pore fluids in the seasonal frozen soil area

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ARTICLE INFO

Article history:

Received 23 July 2021

Received in revised form 13 September 2021

Accepted 18 September 2021

Available online 22 September 2021

Keywords:

Freezing area

Pore fluid

Ice-liquid interface

Salt rejection

Solute migration

Building

Qianghai-Tibet Plateau

Hydrogeological survey engineering

China

ABSTRACT

Seasonal frozen soil accounts for about 53.50% of the land area in China. Frozen soil is a complex multiphase system where ice, water, soil, and air coexist. The distribution and migration of salts in frozen soil during soil freezing are notably different from those in unfrozen soil areas. However, little knowledge is available about the process and mechanisms of salt migration in frozen soil. This study explores the mechanisms of salt migration at the ice-liquid interface during the freezing of pore fluids through batch experiments. The results are as follows. The solute concentrations of liquid and solid phases at the ice-liquid interface (C_L^* , C_S^*) gradually increased at the initial stage of freezing and remained approximately constant at the middle stage. As the ice-liquid interface advanced toward the system boundary, the diffusion of the liquid phase was blocked but the ice phase continued rejecting salts. As a result, C_L^* and C_S^* rapidly increased at the final stage of freezing. The distribution characteristics of solutes in ice and the liquid phases before C_L^* and C_S^* became steady were mainly affected by the freezing temperature, initial concentrations, and particle-size distribution of media (quartz sand and kaolin). In detail, the lower the freezing temperature and the better the particle-size distribution of media, the higher the solute proportion in the ice phase at the initial stage of freezing. Meanwhile, the increase in concentration first promoted but then inhibited the increase of solutes in the ice phase. These results have insights and scientific significance for the tackling of climate change, the environmental protection of groundwater and soil, and infrastructure protection such as roads, among other things.

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1. Introduction

Seasonal frozen soil that freezes in winter and melts in summer is widely distributed in the northeast, northwest, and the Qinghai-Tibet Plateau of China, accounting for about 53.50% of the land area in China (Xu XZ et al., 2010). A multiphase system where ice, water, soil, and gas coexist will be formed downward from the ground surface after the soil starts to freeze. The components in water will be rejected from the ice phase into the liquid phase during the freezing due to the decrease in their solubility and the squeeze effect of

ice dendrites (Konrad JM and Mccammon AW, 1990; Lorain O et al., 2001; Akyurt M et al. 2002), resulting in the accumulation and concentration of these components on the liquid side of the ice-liquid interface (Deng Y et al., 2012; Mtombeni T et al., 2013; Yang H et al., 2016). This will produce certain impacts on the quality of the soil and water in seasonal frozen soil areas (Zuo C, 2018).

The solute redistribution between ice and the liquid phases is a very complicated process since it is affected by freezing rate, salt rejection rate, diffusion rate, and the crystallization induced by phase transition (Terwilliger JP and Dizon SF, 1970; Baker GC and Osterkamp TE, 1989; Konrad JM and Mccammon AW, 1990; Yu T and Ma J, 2005; Zhang LN, 2013; Yang P et al., 2021; Panday S and Corapcioglu MY, 1991; Suh HS and Yun TS, 2018). Owing to the difficulty in the sampling of the unfrozen solution, most of the previous

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studies researched the completely frozen samples by dividing them into 2–3 stages and concluded that freezing concentration occurred during experiments and obtained the distribution patterns of solute in ice and the liquid phases under different experimental conditions (Liu L et al., 1999; Ostroumov VE et al., 2001; Xue S et al., 2014; Williams PM et al., 2013). However, the characteristics and mechanisms of solute migration in the microenvironment at the ice-liquid interface and the effects of different media on the salt rejection process during soil freezing are yet to be ascertained. This study obtains the concentrations of unfrozen solution at different freezing stages through batch experiments, further studies the characteristics and influencing factors of solute redistribution of pore fluids during soil freezing, and explores the characteristics and mechanisms of salt rejection in the microenvironment at the ice-liquid interface. It will provide theoretical bases for the analysis of salt migration and accumulation during the freezing of water-saturated soil such as wetlands in cold regions as well as the prevention and control of water and soil pollution caused by salt accumulation.

2. Materials and conditions of experiments

Nitrates were selected as the target components in this study. They are the most common contaminants in groundwater in many countries at present. The materials and the specifications of instruments and devices selected for experiments and the sampling method of the unfrozen solution are stated in Huang H (2019). Meanwhile, the experimental conditions are designed as shown in Table 1.

3. Analytical methods

As for the solute redistribution during crystallization, the solute redistribution during the solidification of binary alloys is divided into the initial transition stage, stable growth stage, and final transition stage in the metallurgical field. Assuming that only diffusion (no convection) occurs in the liquid phase, when the alloy with a concentration of C_0 solidifies from the left, the solid with a concentration of k_0C_0 is precipitated at the solid-liquid interface (Fig. 1a) and the remaining solutes are rejected into the liquid phase at the interface. As a result, a

solute-enriched boundary layer is formed, but the liquid in the outer layer still has a concentration of C_0 . As the freezing process continues, the solid-liquid interface advances rightward, the solutes are continuously enriched at the front of the ice-liquid interface, and both C_S^* and C_L^* increase accordingly (Fig. 1b). When the solutes diffusing from the solute-enriched layer are equal to the salts rejected during freezing, C_S^* and C_L^* remain unchanged (Fig. 1c). Thus the stable growth stage starts, and the previous stage is referred to as the initial transition stage. At the end of growth, the solute-enriched layer advances into the residual small-volume liquid at the right end and cannot diffuse outward. Consequently, the solutes are enriched again at the front of the interface, forming the final transition stage of the solidification (Fig. 1d).

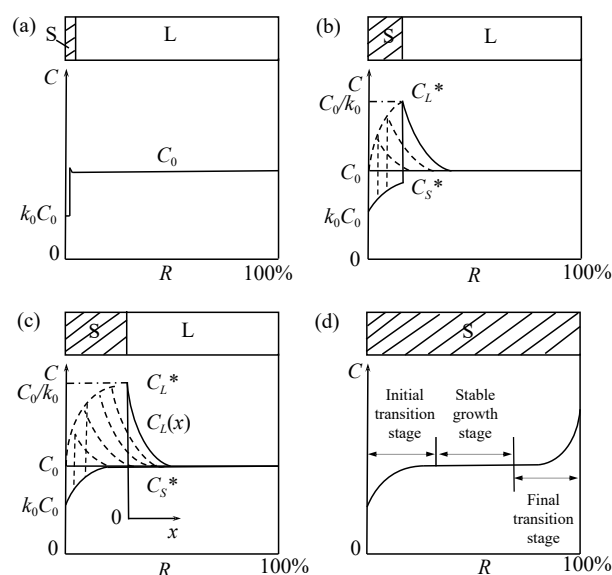


Fig. 1. Solute redistribution in the case that only diffusion occurs in the liquid phase during metal solidification. a–start of solidification; b–initial transition stage; c–stable growth stage; d–complete solidification; S–solid phase; L–liquid phase (after Ma YP et al., 2008).

Although there are differences between the liquid freezing process and the metal solidification process, the solute redistribution at the ice-liquid interface has similar characteristics to a certain extent. This study analyzed the solute redistribution during the freezing of pore fluids by referring to the characteristics of solute redistribution during metal solidification. To better analyze the solute redistribution patterns during freezing, some characteristic parameters are defined as follows.

(i) Volume of unfrozen solution (V_f , mL): The volume of unfrozen solution during the freezing of pore fluids in saturated media refers to the solution volume obtained after centrifugation, excluding the unfrozen water film that remains unfrozen under a low temperature between the soil particle surface and ice (Bittelli M et al., 2003; O'Neill K and Miller RD, 1985) and the unfrozen solution inside inclusions mentioned below.

(ii) Freezing rate (R , %): Referring to the ratio of the volume of frozen solution to the volume of initial solution (V_0 , mL). It can be calculated after measuring the volume of

Table 1. Experimental conditions of the freezing of saturated pore fluid.

Experiment No.	Particle size of quartz sand/mm	Proportion of kaolin /%	Initial concentration /(g/L)	Freezing temperature /°C	Solution volume /mL
S1	0.1–0.25	0	0.3	–7	68
S2	0.1–0.25	0	0.3	–10	68
S3	0.1–0.25	0	0.3	–13	68
S4	0.1–0.25	0	0.01	–10	68
S5	0.1–0.25	0	2.0	–10	68
S6	0.1–0.25	0	5.0	–10	68
S7	0.1–0.25	5	0.3	–7	67
S8	0.1–0.25	8	0.3	–7	64

unfrozen solution.

$$R = \left(1 - \frac{V_l}{V_0}\right) \times 100\% \quad (1)$$

(iii) Freezing rate (v , mL/min): Referring to the volume of solution that freezes per unit of time. In the case that the freezing rate (R) increases linearly with time (t), it can be calculated based on the slope of the freezing rate - time fitted curve (k) and the volume of initial solution (V_0).

$$v = k \times V_0 \quad (2)$$

(iv) Proportion of solutes in the ice phase (E , %): Referring to the ratio of solute mass in the ice phase (M_s) to the solute mass in initial solution (M_0).

$$E = \frac{M_s}{M_0} = \left(1 - \frac{C_l \times V_l}{C_0 \times V_0}\right) \times 100\% \quad (3)$$

(v) Volume fraction: Referring to the ratio of the volume of frozen solution to the total volume at a certain stage.

4. Results and discussion

4.1. Effects of freezing temperature on NO_3^- redistribution

The E - R relationship in experiment No. S1-3 is as shown in Fig. 2. If no NO_3^- redistribution occurred during freezing, the NO_3^- concentration in ice and the liquid phases should have always been equal to the initial NO_3^- concentration. Therefore, the E - R relationship would be present in the form of the dotted line (no redistribution, NR) in Fig. 2. However, the E - R curves during the freezing of pore fluids all fell below the NR line, indicating that the solutes did not evenly enter the ice phase but many of them stayed in the liquid phase during the freezing of pore fluids. That is, solute redistribution occurred at the ice-liquid interface. Furthermore, the E value

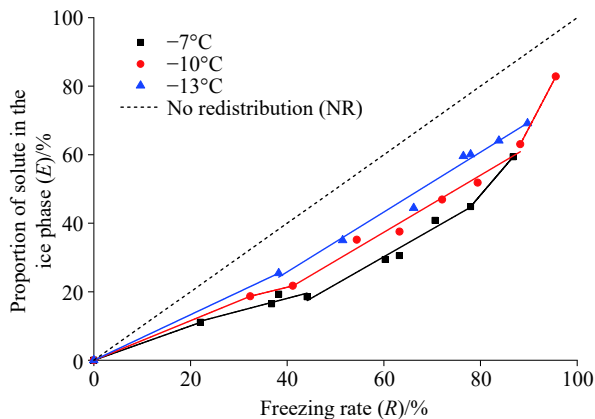


Fig. 2. E - R relationship of pore fluids at different freezing temperatures.

increased with an increase in the R value overall, but the change processes differed under different freezing temperatures. As indicated by the E - R relationship of pore fluids at different freezing temperatures (Fig. 2), the solute redistribution can be divided into different stages, namely the initial transition stage, steady-state stage, and final transition stage. According to the changes in the increasing rate of the E value, the E - R relationship shows four stages at freezing temperatures of -7°C and -10°C but three stages at a freezing temperature of -13°C , as shown in Table 2.

Fig. 3 shows the E - R relationship (by stage) at different freezing temperatures. Stages I and II can be classified as the “initial transition stage” of solute redistribution. At the beginning of freezing, suspended ice crystals would trap unfrozen pore fluid, thus promoting the increasing rate of the E value. As a result, the initial transition stage showed the characteristics that the increasing rate of the E value was higher at the early stage (stage I) than at the late stage (stage II). Meanwhile, at the initial transition stage, the lower the freezing temperature, the higher the solute rejection rate, the more liable the enrichment of solutes at front of the ice-liquid interface, and the higher the increasing rates of C_L^* and C_S^* at the ice-liquid interface. Therefore, the lower the freezing temperature, the higher the increasing rate of the E value and the higher the E value at the same freezing rate at the initial transition stage.

As the freezing temperature decreased, the advancement rate of the ice-liquid interface increased, the duration of solute diffusion at the front of the ice-liquid interface shortened, and the diffusion distance gradually reduced. Accordingly, the concentration gradient of the stable solute-enriched layer gradually increased. As a result, the stable solute-enriched layer showed decreasing thickness (l), increasing concentration gradients, and increasingly steep boundaries as the freezing temperature decreased, as shown in Fig. 4. Therefore, the lower the freezing temperature, the more liable the formation of the steady state of solute distribution. Therefore, the volume fraction of frozen solution gradually decreased at temperatures of -7°C , -10°C , and -13°C at this stage. Meanwhile, this stage was not notable at the temperature of -13°C . C_S^* increased with an increase in C_L^* at this stage. Furthermore, the faster the C_L^* increased, the faster the C_S^* increased, and accordingly, the higher the solute capture rate and the more the solute captured in the ice phase. Therefore, at the same freezing rate, the lower the freezing temperature of the solution, the more the solute trapped in the ice phase and the higher the E value.

Stage III was the steady-state stage of solute redistribution, at which the solute concentration in the ice and the liquid phases remained roughly unchanged at the ice-liquid interface. At this stage, solutes stably entered the ice

Table 2. Freezing stage division of pore fluids at different freezing temperatures.

Freezing temperature/ $^\circ\text{C}$	Stage I	Stage II	Stage III	Stage IV
-7	$0\% < R \leq 22\%$	$22\% < R \leq 44\%$	$44\% < R \leq 78\%$	$78\% < R < 100\%$
-10	$0\% < R \leq 32\%$	$32\% < R \leq 41\%$	$41\% < R \leq 88\%$	$88\% < R < 100\%$
-13	$0\% < R \leq 38\%$	–	$38\% < R \leq 90\%$	$90\% < R < 100\%$

phase, and the increasing rate of the E value was higher than that at Stage II. Meanwhile, the E - R relationship linearly changed at this stage. As shown by the results of the fitting equation, the solute concentration in the ice phase at this stage was close to the initial solute concentration (Table 3).

Stage IV was the final transition stage of solute redistribution, at which the solute-enriched layer at the front of the ice-liquid interface would have no space to migrate after reaching the boundary. As a result, the solute concentration in the liquid phase rapidly rose. Accordingly, the solute concentration in the ice phase also increased, and the increasing rate of the E value increased once again. The lower the freezing temperature, the narrower the solute-enriched layer formed at Stage II and the higher the freezing rate when the layer migrated to the boundary.

4.2. Effects of initial concentration on NO_3^- redistribution

The E - R relationship in experiments Nos. S2 and S4-6 is shown in Fig. 5. As indicated by the E - R relationship of the

pore fluids with an initial concentration of 0.01 g/L, 0.3 g/L, and 2 g/L, the solute redistribution can be divided into three stages. However, the E - R curve of the pore fluids with an initial concentration of 5 g/L only showed the initial transition stage and final transition stage. According to the changes in the increasing rate of the E value, the E - R relationship under different initial concentrations can be divided into four stages, as shown in Table 4.

As shown in Fig. 6, stages I and II were the initial transition stage of solute redistribution. According to Fig. 6 and Table 4, Stage I did not show significant differences under different initial concentration conditions, while Stage II exhibited different volume fractions of frozen solution and increasing rates of the E value under different initial concentrations (0.01 g/L, 0.3 g/L, 2 g/L, and 5 g/L). The freezing point of a water solution decreases with an increase in the solute concentration. Meanwhile, a low temperature contributes to an increase in the viscosity, a decrease in the kinetic energy of transition (returning to the most balanced and stable ground state), and a decrease in the diffusion

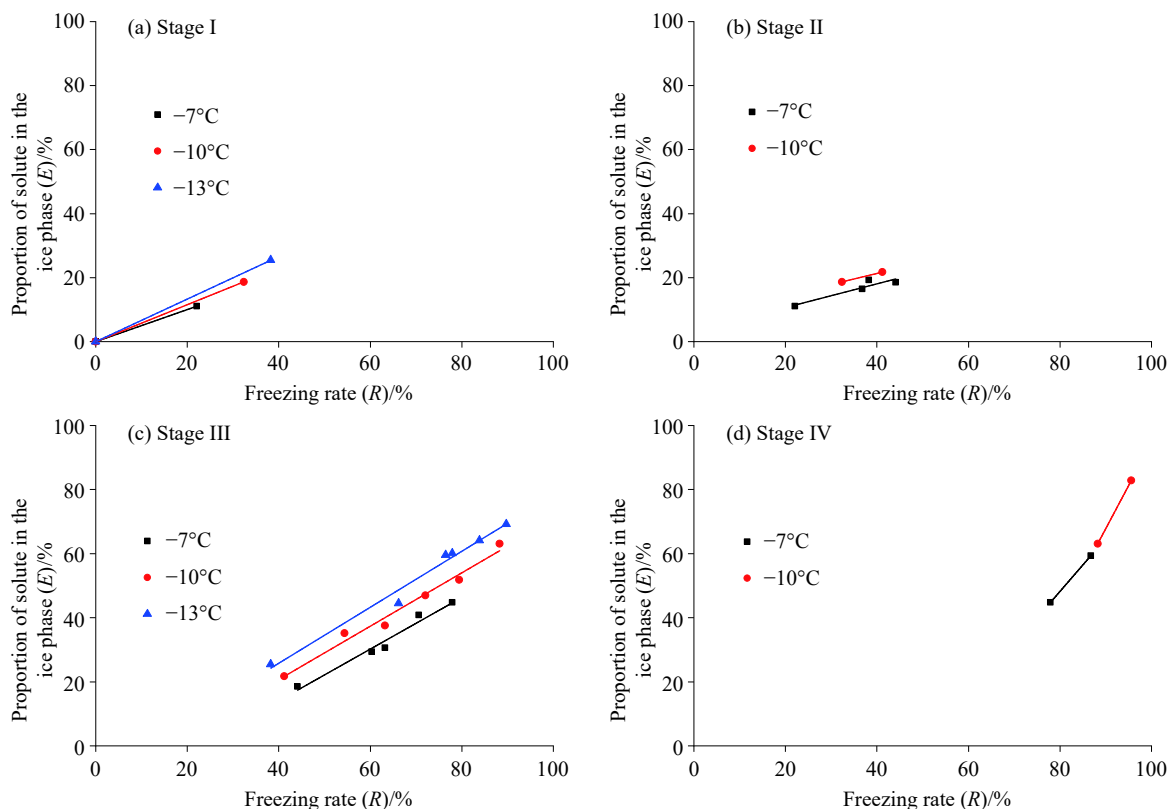


Fig. 3. E - R relationship (by stage) of the pore fluids at different freezing temperatures.

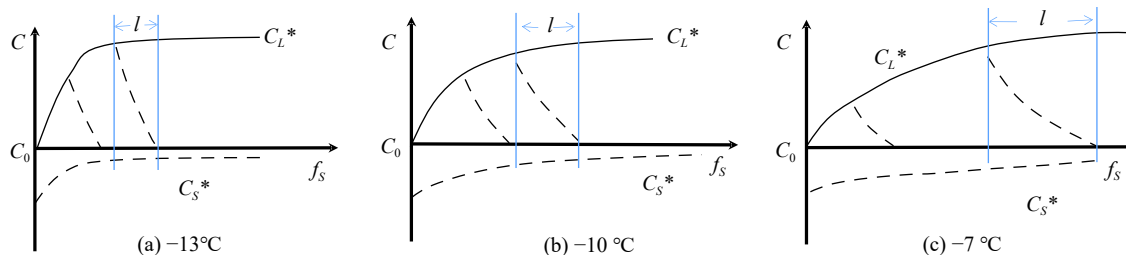


Fig. 4. Diagrams of solute enrichment characteristics at the freezing front of pore fluids under different freezing temperatures.

Table 3. NO_3^- concentration in the ice phase at the steady-state stage of solute distribution at different freezing temperatures.

Freezing temperature/ $^{\circ}\text{C}$	NO_3^- concentration in the ice phase C_S^* (g/L)	C_S^*/C_0
-7	0.24	0.81
-10	0.25	0.83
-13	0.26	0.88

coefficient of a water solution (Yu T and Ma J, 2005; Jie WQ, 2010; Lü HZ et al., 2015). The decrease in diffusivity will accelerate the solute enrichment at the front of the ice-liquid interface. In contrast, an increase in the initial concentration of pore fluids will reduce the freezing rate, which is unfavorable for the solute enrichment at the front of the interface. In this case, the solute redistribution is the competition result between the diffusion rate and freezing rate.

In the case that an initial concentration was less than 0.3 g/L, the freezing of every 1% (volume fraction) of pore fluids implied that the average solute content in the ice phase was about 0.31% and 0.37% of the total mass of solute, respectively for pore fluids with an initial concentration of 0.01 g/L and 0.3 g/L. That is, the average solute content in the ice phase increased with an increase in the initial concentration. In this case, the higher the initial concentration, the more liable the solute enrichment at the front of the ice-liquid interface. The difference in the freezing rate was 0.0008 mL/min between the pore fluids with an initial concentration of 0.01 g/L and 0.3 g/L, indicating that the effects of the freezing rate on solute rejection were smaller than those of the concentration on the diffusion. In this case, the higher the initial concentration, the smaller the volume fraction of the frozen solution at this stage.

In the case that an initial concentration was greater than 0.3 g/L, the freezing of every 1% (volume fraction) of pore fluids implied that the average solute content in the ice phase

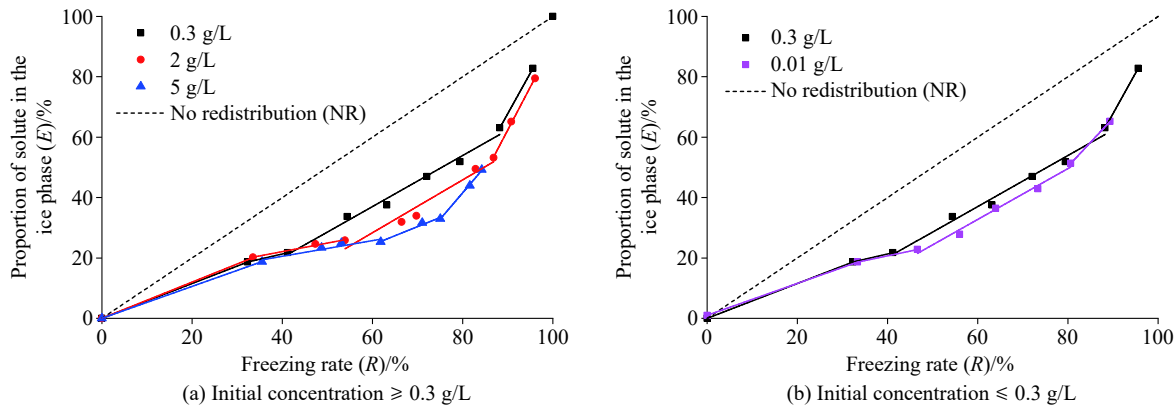
was about 0.37%, 0.28%, and 0.24% of the total mass of solute, respectively for pore fluids with an initial concentration of 0.3 g/L, 2 g/L, and 5 g/L. That is, the average solute content in the ice phase decreased with an increase in the initial concentration, which was contrary to the characteristics in the case that the initial concentration was less than 0.3 g/L. This indicates that the solute redistribution is significantly influenced by changes in the freezing rate. The difference in the freezing rate was 0.0311 mL/min between the pore fluids with an initial concentration of 0.3 g/L and 2g/L and was 0.0895 ml/min between the pore fluids with an initial concentration of 0.3 g/L and 5 g/L. Both of them are dozens of times the difference in the freezing rate between the pore fluids with an initial concentration of 0.01 g/L and of 0.3 g/L. Therefore, for the pore fluids with a high initial concentration, solutes were not liable to be enriched at the front of the interface due to the low freezing rate. Correspondingly, both C_L^* and C_S^* slowly increased, and it took a longer time for the solute redistribution to reach the steady-state stage.

At Stage III, the higher the initial concentration, the higher the solute concentration in the ice phase (Table 5). However, the C_S^*/C_0 ratio slightly changed under different initial concentrations.

Stage IV was the final transition stage of solute redistribution, at which the solute-enriched layer at the front of the ice-liquid interface reached the boundary. In this case, C_L^* and C_S^* quickly increased, so did the corresponding E value. The narrower the solute-enriched layer, the smaller the volume fraction of the frozen solution at this stage.

4.3. Effects of particle-size distribution of media on NO_3^- redistribution

The E - R relationship in experiments Nos. S1 and S7-8 is shown in Fig. 7. It can be seen that at the same freezing rate, the higher the kaolin content, the higher the E value. This

**Fig. 5.** E - R relationship during the freezing of pore fluids with different initial concentrations.**Table 4.** Freezing stage division of pore fluids with different initial concentrations

Initial concentration/(g/L)	Stage I	Stage II	Stage III	Stage IV
0.01	0%< R <33%	33%< R <46%	46%< R <81%	81%< R <100%
0.3	0%< R <32%	32%< R <42%	42%< R <88%	88%< R <100%
2	0%< R <32%	32%< R <52%	52%< R <86%	86%< R <100%
5	0%< R <35%	35%< R <61%	61%< R <75%	75%< R <100%

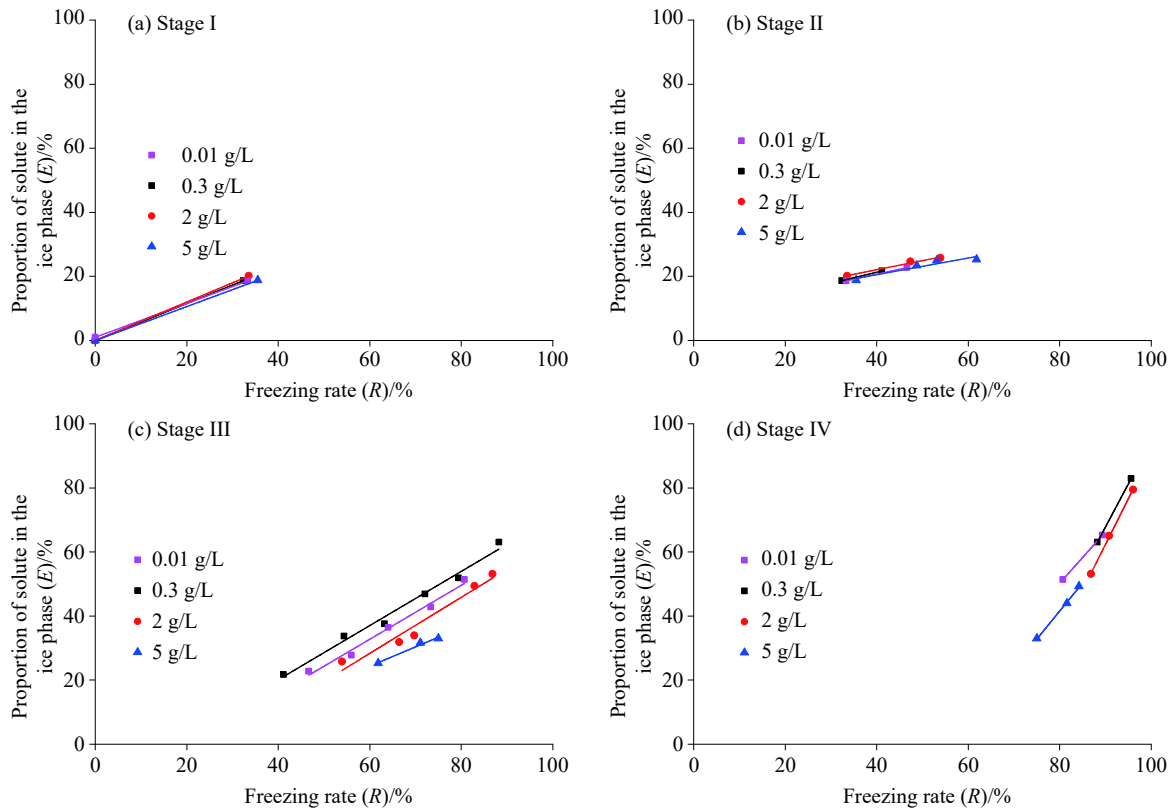


Fig. 6. E - R relationship (by stage) during the freezing of pore fluids with different initial concentrations.

Table 5. Solute concentrations in the ice phase at the steady-state stage of solute redistribution under different initial concentrations.

Initial concentration C_0 (g/L)	C_S^* (g/L)	C_S^*/C_0
0.01	0.008	0.84
0.3	0.25	0.83
2	1.74	0.87

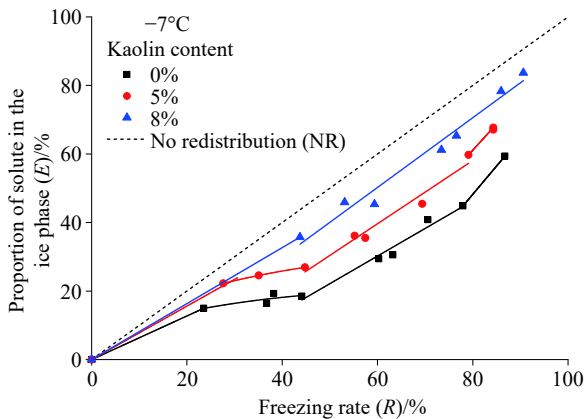


Fig. 7. E - R relationship of pore fluids under different particle-size distribution of media.

indicates that more solutes were trapped in the ice phase with an increase in kaolin content. The changes in the increasing rate of the E value differed under different particle-size distribution of media. Based on this, the E - R relationship can be divided into four stages under the kaolin content of 0% and

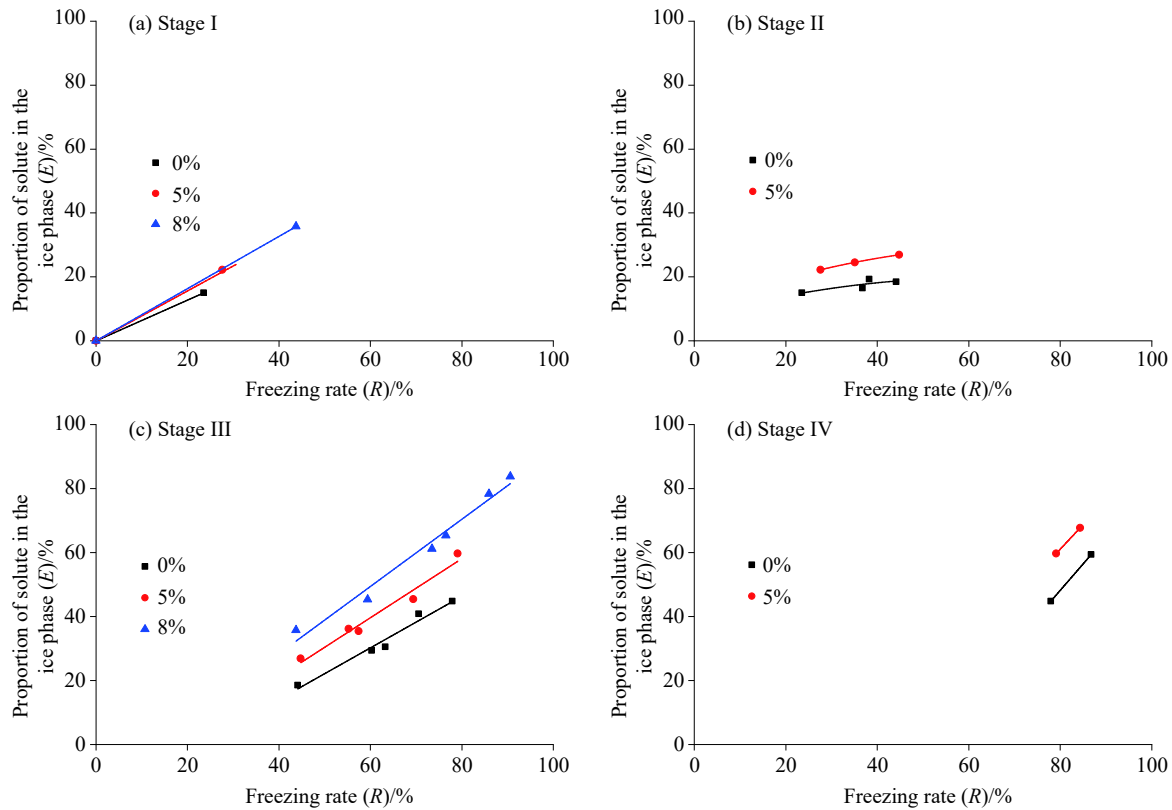
5% and three stages under the kaolin content of 8% (Table 6).

The E - R relationship was organized according to stages, as shown in Fig. 8. Stages I and II were the initial transition stage of solute redistribution. At Stage I, the E value continuously increased at a high rate affected by suspended ice crystals trapping unfrozen pore fluid. Afterward, the increasing rate of the E value decreased as the influence of suspended ice crystals weakened, and then Stage II commenced. At this stage, the solutes rejected during freezing were continuously enriched at the front of the ice-liquid interface. As a result, C_L^* and C_S^* continuously increased, and the solute redistribution showed different results under different particle-size distribution of media. In detail, the freezing of every 1% (volume fraction) of pore fluids implied that the average solute content in the ice phase was about 0.17% and 0.27% of the total mass of solutes, respectively under the kaolin content of 0% and 5%. Meanwhile, at the same freezing rate, the higher the kaolin proportion, the higher the E value. That is, the solute concentration in the ice phase increased with an increase in kaolin content. As analyzed above, the higher the freezing rate, the narrower the solute-enriched layer at the front of the ice-liquid interface and the more liable the formation of the steady state. Since the freezing rate increased with an increase in kaolin content, the duration of the initial transition stage decreased correspondingly. These characteristics were not notable in the case that the kaolin content was greater than 8% at this stage.

Since kaolin can be filled into the pores of quartz sand skeleton, an increase in the added kaolin led to a decrease in

Table 6. Freezing stage division of pore fluids under different particle-size distribution of media.

Kaolin content/%	Stage I	Stage II	Stage III	Stage IV
0	$0% < R \leq 22%$	$22% < R \leq 46%$	$46% < R \leq 78%$	$78% < R < 100%$
5	$0% < R \leq 27%$	$27% < R \leq 44%$	$44% < R \leq 79%$	$79% < R < 100%$
8	$0% < R \leq 45%$	–	$45% < R \leq 92%$	$92% < R < 100%$

**Fig. 8.** *E-R* relationship (by stage) of pore fluids under different particle-size distribution of media.

both the porosity of media and the pore throat size of the samples. As the ice crystals grew, they were more liable to freeze at pore throats and thus block the pore throats. Consequently, more high-concentration unfrozen-solution inclusions were formed. Meanwhile, samples with high kaolin content bore more ice cores and smaller pore throats, and thus the ice crystals in the samples were more liable to connect with each other and block the pore throats. Furthermore, since the thermal conductivity of ice is greater than that of water, the cold energy was transmitted forward at a higher rate and the freezing rate thus increased under a constant temperature in the test chamber. This further promoted the formation of inclusions, and the high-concentration solutes in the unfrozen solution in the inclusions would not diffuse into other unfrozen pore fluids. Therefore, at the same freezing rate, the higher the kaolin content in water-saturated media, the higher the proportion of solute trapped in the ice phase.

Solutes evenly entered the ice phase at Stage III, and thus the steady-state stage of solute redistribution began. Meanwhile, the *E-R* relationship linearly changed at this stage. As shown by the results of the fitting equation, the higher the kaolin content at this stage, the higher the solute concentration in the ice phase (Table 7).

Stage IV corresponded to the final transition stage of

solute redistribution. At this stage, the solute-enriched layer at the front of the ice-liquid interface diffused to the boundary, both C_L^* and C_S^* quickly increased, and the increasing rate of the *E* value further increased. According to the extended *E-R* curves under the kaolin content of 8%, the proportion of the solutes entered the ice phase was less than 100% at the freezing rate of 100%. This indicates that there existed a stage (freezing rate $< 92%$) at which the *E* value quickly rose in the late stage of the freezing. Therefore, the initial freezing rate at this stage tended to increase with an increase in kaolin content and the main reasons are as follows. With an increase in the kaolin content, the freezing rate increased and the solute-enriched layer formed at the front of the ice-liquid interface narrowed at Stage II. Accordingly, the freezing rate increased during the solute-enriched layer migrated to the

Table 7. Solute concentration in the ice phase at the steady-state stage of solute redistribution under different particle-size distribution of media.

Kaolin content/%	$C_S^*/(\text{g/L})$	C_S^*/C_0
0	0.24	0.81
5	0.26	0.86
8	0.29	0.97

boundary and the volume fraction of frozen solution at this stage correspondingly decreased.

Overall, the particle-size distribution of media grew better with an increase in kaolin content. This led to more solutes trapped in the ice phase in water-saturated medium and correspondingly poorer salt rejection effects under the same experimental condition.

4.4. Scientific significance of the results obtained in this study

The frozen soil is more than 1 m deep in the northeast, northwest, and the Qinghai-Tibet Plateau of China, with the freezing-thawing time longer than 200 days (Wang XW, 2010). The results of this study revealed the salt redistribution and its dynamic processes during the freezing of frozen soil, subject to which some salts in pore water will be displaced into unfrozen soil during the freezing of soil. In this way, the distribution of salts in frozen soil will be changed. The results of this study have important insights and scientific significance for the tackling of climate change, the environmental protection of groundwater and soil, and infrastructure protection such as roads, among other things. (1) In terms of the tackling of climate change, salt redistribution may affect the microbial activity and biocoenosis structure in seasonal frozen soil and further influence the temporal-spatial changes such as the emissions of greenhouse gases (N_2O and CO_2). The results of this study may have insights significance for the understanding and the tackling of climate change; (2) In terms of the environmental protection of groundwater and soil, many frozen soil areas in China suffer serious pollution, and a considerable area of farmland has been discarded due to pollution (Liu QS et al., 2004). The results of this study can help to better understand the causes and mechanisms of the pollution in frozen soil areas and then further provide technical support for the prevention and control of soil-groundwater pollution in these areas; (3) In terms of infrastructure protection, the infrastructures such as roads and bridges in frozen soil areas frequently suffer from expansion due to freezing-induced salt rejection. This will damage the infrastructures and thus cause huge safety hazards and economic losses. The results of this study further enrich the understanding of the processes and mechanisms of the salt rejection-induced swelling; (4) In other aspects, the frozen transportation and cold storage of foods also involve the freezing and salt rejection of porous media. The experiment results of this study may provide new ideas for the quality and freshness control of foods.

5. Conclusions

(i) The solute redistribution at the ice-liquid interface during the freezing of pore fluids in cold areas can be divided into three stages, namely the initial transition stage, steady-state stage, and final transition stage. (1) At the initial transition stage, the rate of freezing-induced solute rejection is higher than the rate at which solutes are taken away by the diffusion of the liquid phase, and thus C_L^* and C_S^* gradually

increase. The lower the freezing temperature, the larger the proportion of solutes entering the ice phase. Meanwhile, with an increase in the initial concentration, the proportion of solutes in the ice phase increases in the case that the initial concentration is lower than 0.3 g/L and decreases when the initial concentration is higher than 0.3 g/L. (2) At the steady-state stage, the rate of freezing-induced solute rejection is equal to the rate at which solute is taken away by diffusion of the liquid phase, and C_L^* and C_S^* remain unchanged. The solute redistribution at the ice-liquid interface is slightly affected by the difference in the freezing temperature and initial concentration at this stage. (3) At the final transition stage, the solute-enriched layer at the front of the ice-liquid interface has no space to migrate, thus leading to a rapid increase in the C_L^* and C_S^* at the ice-liquid interface.

(ii) The pore size of media has significant effects on the solute redistribution during the freezing of saturated pore fluid. Pores with small size are more liable to be blocked and form inclusions. The solutes in the unfrozen solution in the inclusions cannot diffuse into other unfrozen pore fluids, thus increasing the proportion of solute trapped in the ice phase.

(iii) Some solutes will be rejected into the unfrozen fluid during the freezing of water-saturated soil in seasonal frozen soil areas, thus leading to an increase in the solute concentration in unfrozen areas. The proportion of the solutes rejected depends on the initial solute concentration, freezing rate, and particle-size distribution of soil. In frozen soil, the solute concentration in the ice phase will be higher in areas with a higher freezing rate or better particle-size distribution of soil than that in other areas.

(iv) The results of this study have important insights and scientific significance for the tackling of climate change, the environmental protection of groundwater and soil, and infrastructure protection such as roads, among other things.

CRedit authorship contribution statement

Ming-zhu Liu, Hong-han Chen, Ding-ding Wu, and Huan Huang conceived and planned the experiments. Huan Huang, Chang-fu Chen, Xiao-jie Mo and Ding-ding Wu carried out the experiments. Huan Huang, Hong-han Chen, Ming-zhu Liu, and Chang-fu Chen contributed to the interpretation of the results. Huan Huang took the lead in writing the manuscript. All authors provided critical feedback and helped shape the research, analysis, and the manuscript.

Declaration of competing interest

The authors declare no conflicts of interest.

Acknowledgement

This research was financially supported by the National Natural Science Foundation of China (41572225), project of China Geological Survey (DD20189662, DD20211256). The authors would like to express sincere gratitude to the reviewers who have provided significant suggestions for this manuscript.

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