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Simulation study on clogging of suspended particles in in-situ leaching of uranium at different concentrations and flow velocity

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Abstract: Clogging problem has become one of the key problems restricting the mining efficiency of insitu leaching of uranium, and the related research on the law and mechanism of physical clogging has not been reported. In order to identify and understand the complicated law and mechanism, experimental device is established to simulate the physical clogging caused by suspended particles in the uranium process, the physical clogging law and mechanism under different concentrations and velocity of flow are studied. The experimental results show that with the concentration of suspended particles increasing from 100, 200, 300 to 400 mg/L, the permeability of porous media gradually decreases, and the clogging phenomenon becomes more and more obvious. When the size of suspended particles is small and the velocity is 15 mL/min, the porous medium will not appear clogging, while the velocity is 25mL/min, the whole porous medium will slowly appear internal deposition clogging. When the size of suspended solids is larger and the flow rate is 9, 12, 15mL/min, the higher the velocity, the faster the clogging will be, and backwash can alleviate the surface clogging but cannot change the final clogging result. According to the experiment and actual situation, the physical clogging in in-situ leaching of uranium is mainly surface clogging and filter clogging.

Key words: in-situ leaching of uranium, physical clogging, suspended particles, porous medium, permeability coefficient

1. The introduction

In-situ leaching uranium mining is a sustainable uranium mining method with the greatest potential, which is mainly suitable for loose sandstone ore-bearing layers. At present, the proportion of sandstone uranium mining is the highest in the world. With the continuous improvement of technology and process, the proportion of in-situ leaching uranium mining in the world will continue to increase year by year. In the process of In-situ leaching uranium, however, the interaction between groundwater and wall rock is very complex, there are significant problems with mine clogging as the cycles of liquid pumping and injection, including the decreasing in permeability and clogging of seam, the scaling and clogging of pumping and injection wells, resin poisoning and other phenomena (Cai et al., 2021), whether using the method of acid leaching, alkaline leaching or $CO_2 + O_2$ neutral leaching. For example, mineralogical evidence of pore clogging by sandstone had been found in Wuyier Uranium Mine in Yili Basin, Xinjiang, Northwest China; Because of the clogging of scale in Yining Uranium Mine, the pumping capacity was reduced by more than 20%, the well hole washing cycle was shortened to 20 days/time, the resin was clogging and poisoned, the treatment water was reduced by about 30%, and finally the production capacity was reduced by 20%-30%; The formation clogging phenomenon often occured in the process of in-situ uranium leaching in Tongliao Qianerkuai Uranium Mine in Inner Mongolia, which led to frequent well washing, and it took an average of 5-7 days to wash well (CAI et al., 2021). A large number of plugging phenomena in the field of uranium mining projects show that in the pumping and injection wells, it is mainly the accumulation of suspended particles, fine sand, precipitation and cement in the ore layer around the filter that leads to the clogging, and in the underground ore layer, it is mainly reflected in the decrease of permeability. It can be seen that the clogging problem in the process of in-situ leaching of uranium will lead to the reduction of the permeability in the filter and the surrounding ore layer, the reduction of pumping and injection volume, the decline in production capacity, and even lead to the abandonment of the borehole if not treated in time. Therefore, clogging is one of the key problems restricting the mining efficiency of in-situ leaching of uranium.

Clogging is closely related to a number of factors, generally, it can be divided into physical clogging, chemical clogging and biological clogging according to the source of blocking substances, among which physical clogging is the most common type of clogging and the most important and direct control factor for the formation of clogging (Wang et al., 2012), and suspended particles clogging is the most typical case of physical clogging. At present, there are many researches on the law, theory and mechanism of physical clogging caused by suspended particles in porous media, which have been carried out in many fields: Chromatography or substance in the micro fluid separation, biological filtration, water and wastewater treatment, the process of drilling and oil recovery process, the pollutant transport or storage in soil and sediment transportation, groundwater recharge, garbage landfill (Gerber et al., 2018), in which the study of the clogging in groundwater recharge is the most extensive and in-depth (Du et al., 2014; Fernández et al., 2015; Du et al., 2018; Ye et al., 2019; Xie et al., 2020; Xie et al., 2020).

Many scholars in these fields have conducted experiments to explore the influencing factors of migration clogging of suspended particles in porous media, and studied the clogging laws and mechanisms under different influencing factors. they classified the influencing factors of physical clogging caused by suspended particles in porous media into internal and external causes according to the source. Internal factors are mainly the characteristics of porous media, including porous media particle size, media type, media gradation, media porosity, etc. (Pan et al., 2013; Feia et al., 2015; Fetzer et al., 2017; Yousif et al., 2017; Wang et al., 2018; Du et al., 2018; Banihashem et al., 2020; Tang et al., 2020; Zhang et al., 2021). External factors mainly refer to the inlet water characteristics and external environment, such as particle size, particle gradation, particle type, particle concentration, velocity, temperature, etc. (Ahfir et al., 2009; Bedrikovetsky et al., 2012; Agbangla et al., 2012; Bai et al., 2016; Bennacer et al., 2017; Loizeau et al., 2017; Hou et al., 2020). Wang (Wang et al., 2012) found that the smaller the suspended particles are, the farther the movement distance is, and the larger the range of plugging in porous media is. Du (Du et al., 2018) proved that the diameter ratio of suspended particles to sand particles is the main factor affecting the physical plugging position through a series of sand column tests. Banihashem (Banihashem et al., 2020) studied the effect of the size of suspended particles in the fluid on the reduction of permeability and porosity in the soil filter media. The experimental results of Feia (Feia et al., 2015) show that the following basic parameters have a significant impact on particle transport and deposition in porous media: particle concentration in suspension, injection flow rate, size of injected particles, pore size of particle matrix and surface roughness of media particles. Zhang (Zhang et al., 2021) found that the particle size of porous medium also had significant influence on the migration of suspended particles, the permeability of fine sand decreased the fastest, while that of medium and coarse sand showed an oscillatory decline.

However, there are few studies on the clogging problem in the process of in-situ leaching uranium mining so far, and their search mainly focuses on chemical clogging (Zhao et al., 2018; Su et al., 2020;

Zeng et al., 2021), but the related research on the law and mechanism of physical clogging has not been reported. Moreover, the solution to the clogging problem is mainly to eliminate the blockage from the chemical, mechanical and physical aspects in the engineering practice (such as groundwater recharge, oil exploitation, etc.) (Ahfir et al., 2007; Zhang et al., 2018; Nikoloski et al., 2019; Odebiyi et al., 2021; Odebiyi et al., 2022), but it is not based on the mechanism of physical clogging to prevent and reduce the clogging from the source. Therefore, it is of great theoretical and practical significance to study the development law and main influencing factors, analyze the reason and mechanism of physical clogging of suspended particles in the process of in-situ leaching of uranium.

The purpose of this study is to establish a column experiment device to simulate the physical clogging process caused by suspended particles in in-situ leaching of uranium, to analyze the physical clogging law under the two influencing factors of suspended particles concentration and velocity, and to explain and discuss the clogging problem in in-situ leaching of uranium combined with the mechanism of physical clogging.

2. Experimental scheme

2.1. Experimental device

The plexiglass sand column was used to simulate the underground structure of uranium leaching to carry out the penetration experiment. as shown in Fig. 1. The Plexiglass column is a cylinder with a height of 180 mm and an inner diameter of 25 mm, and the ratio of height to inner diameter is 7.2, which can be regarded as a one-dimensional infiltration process (Zhang et al., 2018). The magnetic stirrer was used to stir the configured suspension solution to keep the concentration of suspension uniform and the suspension state of suspended particles. Stainless steel filters with a pore size of 0.1 mm were set at the inlet and outlet of the experimental column to prevent porous media from flowing out of the sample. Four pressure measuring holes were set on the experimental device along the flow direction of the suspension, one was set at the entrance of the experimental column, and the other three were set at the side of the column. The spacing of pressure measuring holes from bottom to top was 3 cm, 6 cm and 6 cm, respectively. During the experiment, the pressure sensors were used to measure the pressure values at different positions, and a data acquisition instrument was used to record all instantaneous pressure values. Lange precision peristaltic pump (LEAD15-44) was used to transport the suspended solution to the experimental sand column from bottom to top at a constant velocity, and the final outflow liquid was collected by automatic sampler.



Fig. 1. Experimental device

2.2. Experimental materials

Quartz sand with a median particle size of 250 µm was used as the infiltration porous medium in this experiment. Quartz sand is composed of 99.5% silica of quartz phase and a small amount of metal oxidation. The particle size of quartz sand is controlled between 40 and 80 mesh (average 60 mesh) using a standard screen. The quartz sand was stirred and soaked with 0.1 M HCl solution and 0.1 M NaOH solution for 24 h to remove trace metal oxides and organic matter, and then washed repeatedly with deionized water until the pH of the washing solution was close to 7. After that, the quartz was sterilized by pressure for 2.5 h and then dried at 105°C for 24 h.



Fig. 2. Size distribution diagram and online image of suspended particles of injection hole

Suspension of 1 #	Stock	Та:11: алті 4	Well	Well injection	-	-
uranium	solution	Tall liquid	pumping			
Average size/nm	1109	213	2864	4725	-	-
Suspension of 2 # uranium	Stock solution 2	Tailliquid2	Initial well washing	Middle of well washing	End of well washing	Initial well washing recovery
Average size/nm	2125	1651	2707	5999	13749	29883

In order to determine the particle size range of suspended particles, leaching circulating solution was collected from two uranium mines in Inner Mongolia (1# uranium mine and 2# uranium mine, respectively) for particle size analysis. 1# uranium mine has collected stock solution, tail fluid, well pumping fluid and well injection fluid, while 2# uranium mine has collected stock solution, tail fluid, initial well washing fluid, middle of well washing fluid, end of well washing fluid and initial well washing recovery fluid. The collected solution was sonically dispersed, and then the supernatant suspension was taken after standing for 10min. The particle size of suspended particles in the circulating solution of different parts was analyzed by laser particle size analyzer (90Plus PALS high-sensitivity Zeta potentiometer particle size analyzer, Brookhaven, USA) (Fig. 2). In addition, the Finland Pixact online particle imaging system was used to image suspended particles (Fig. 2), and the measurement results are shown in Table 1. According to the particle size measurement results, the particle size range is mainly nano scale and micro scale, the average particle size of suspended particles near the liquid

injection hole before clogging is 2.8-4.7 μ m, and the average particle size of suspended particles in the well washing stage after clogging is 10-30 μ m, so the particle size range of suspended particles in the experiment is 3-30 μ m. In order to avoid any chemical reaction between injected suspended particles and quartz sand, natural spherical SiO₂ particles were used as suspended particles in the experiment.

2.3. Experimental steps and methods

In this study, sand column leaching experiments under the conditions of suspension with different velocity, different concentrations and different particle sizes were set to simulate the process of in-situ leaching of uranium. The experimental conditions are shown in Table 2.

	- -	The second se		
The serial	Suspended particle	Suspended particle	The velocity/	Porous medium
number	size/um	concentration/mg/L	mL/min	particle size/mesh
E1	30	400	9	40-80
E2	30	300	9	40-80
E3	30	200	9	40-80
E4	30	100	9	40-80
E5	30	200	12	40-80
E6	30	200	15	40-80
E7	3	200	15	40-80
E8	3	200	25	40-80

Table 2 Main experimental parameters

The pretreated quartz sand is filled into the experimental column by the wet packing method. During each filling, ensure that the water surface is higher than the top surface of the quartz sand, and fill in layers in a step size of about 1cm. In the filling process of each layer, use a cleaning ball to beat the plexiglass column to remove the micro bubbles on the surface of the quartz sand, so as to ensure that the quartz sand in the experimental column is a saturated porous medium and uniform.

Clogging is caused by the osmotic flow of suspension in porous media, and its characteristics and action law are mainly characterized by the change of permeability coefficient of medium. In the course of the experiment, the hydraulic head values at different positions at different times were recorded by pressure sensors, and the permeability coefficients were calculated under the condition of constant flow. The permeability coefficient of the medium in the sand column was calculated according to Darcy formula:

$$K = \frac{Q \cdot \Delta x}{\pi R^2 \cdot \Delta H} \tag{1}$$

where: Q--velocity (m³/d); Δx --distance between any two pressure measuring holes (m); ΔH --head difference between two pressure measuring holes (m); R--inner diameter of sand column (m).

The final media clogging degree can be characterized by the dimensionless parameter K/K_0 , where K_0 is the initial permeability coefficient of the layer media. The smaller K/K_0 is, the greater the degree of media clogging is.

3. Results analysis

In order to protect the experimental device, the experiment took obvious clogging phenomenon as the cycle, and the experiment was terminated when the pressure data in the four pressure sensors showed

obvious multiple increase and tended to be stable, and the backwashing experiment was carried out in the case of rapid clogging to determine the type of clogging. The permeability coefficients of 0-3, 3-9, 9-15 cm sections of sand column under different concentrations were calculated and converted into the dimensionless relative permeability coefficient K/K₀according to Darcy's law, The relationship between the relative permeability coefficient and the infiltration time was constructed to observe the influence of the concentration of suspended particles and the velocity on the physical clogging in porous media.

3.1. Pore clogging mechanism

Clogging is defined as the reduction of the effective area available for fluid flow, which is defined as the macroscopic effect of the microscopic phenomenon of fine particles trapped at the pore throat (Valdes, 2002). Permeability decreases in porous media, which can be gradual or instantaneous, depending on the mechanism and type of pore clogging. There are three main types of single pore plugging mechanisms according to existing relevant studies: sieving; bridging and aggregation clogging (Dressaire et al., 2017), as shown in Fig. 3.



Fig. 3. Main mechanism of pore clogging

1. Sieving, the pores of porous media are small enough to prevent any suspended particles from passing through, or the minimum particle size of suspended particles is larger than the pore size, and most suspended particles cannot enter the porous media, generally showing the phenomenon of caking directly on the outside.

2. Bridging, when the size of suspended particles is smaller than the pore throat size, suspended particles may directly accumulate in the pore throat at the entrance, thus clogging the entire porous media inlet, and this clogging can be removed and restored by backwashing.

3. Aggregation clogging, a number of suspended particles gradually deposit and gather in the pore until the pore is filled and blocked.

3.2. The effect of suspended particles concentration on physical clogging

Under the conditions that velocity was 9mL/min and suspended particle size was 30µm, the sand column penetration experiments with suspended particle concentrations of 100, 200, 300, and 400 mg/L were carried out to study the migration and physical clogging laws of suspended particles with different concentrations in porous media.

It can be seen from Fig. 4 that the relative permeability coefficients at different depths under different concentration conditions have obvious rules. Firstly, as the concentration increases from 100, 200, 300, and 400 mg/L, the time for obvious clogging gradually decreases, which is mainly reflected in the 0-3 cm area of the sand column, which is 120 h, 57 h, 40 h, and 20 h in turn. This indicates that the higher the concentration of suspension, the shorter the time for significant clogging (especially in the surface layer) to occur. This is mainly because more suspended particles pass through the porous medium, and the chance of collision between suspended particles also increases with the increase of concentration, which makes it easier to deposit in the pores of the porous medium, and finally leads to the continuous decline of permeability coefficient.



Fig. 4 Variation curves of relative permeability coefficients in different regions under different concentrations



Fig. 5 The phenomenon of "cake forming" in filter screen

Secondly, in terms of the change of relative permeability coefficient, in the 0-3 cm area of the sand column, the relative permeability coefficient decreases the most, and the maximum reduction is 80%-90%. At the early stage of the experiment, the relative permeability of all regions in the porous medium was greater than 1, which indicated that the sudden infiltration of suspension in the initial period changed part of the pore structure inside the porous medium and even increased its porosity, thus increasing the permeability of the porous medium. Then, with the migration and deposition of suspended matter gradually filling the pores of the porous medium, the corresponding relative permeability coefficient remained relatively stable and slowly decreased. After 20 hours of the experiment, in the 0-3 cm area of the sand column with concentration of 200, 300 and 400 mg/L, the relative permeability decreased quasi-linearly, which was due to the suspended particles gradually

deposited and filled most of the pores in this surface area, and eventually clogged the surface of the porous medium, with the continuous accumulation of suspended particles in the surface layer, an obvious phenomenon of "cake formation" (as shown in Fig. 5)will occur at the filter screen in serious cases, This phenomenon has been described and characterized by multiple studies (Barkman et al., 1972; Feia et al., 2015), which is mainly due to that the suspended particles accumulate more and more on the surface of the porous medium and eventually block the pores of the filter screen, resulting in a quasi-linear decline of relative permeability coefficient in the 0-3cm area as shown in Fig. 4. However, in the sand column with a concentration of 100 mg/L, the number of suspended particles in the surface porous medium, so the migration and deposition in the whole sand column was a gradual and slow process, but the surface clogging of the sand column was still the main feature. The experimental results show that the higher the concentration of suspension is, the faster the permeability decreases and the more obvious the clogging phenomenon is.

3.3. The effect of suspension velocity on physical clogging

Under the conditions that the suspended particle size was 3 µm and the concentration was 200mg/L, the suspension penetrated through the sand column at the velocity of 15mL/min, the permeability coefficient did not decrease significantly for a long period of time (as shown in Fig. 6), it indicated that the suspended particles migrated and deposited evenly in the porous medium of the whole sand column when the velocity was low, which did not block the surface or internal pores during the experimental period. After the velocity was increased to 25mL/ min, the permeability coefficient of the experimental sand column decreased significantly. The relative permeability coefficient of each area of the whole sand column decreased with the same trend, indicating that suspended particles were uniformly deposited in each area of the sand column and gradually filled the pores of the porous medium in the whole area, it caused the overall permeability of porous media to decrease and gradually blocked.



Fig. 6 Relative permeability coefficient variation curves at different velocity at 3 μm :(a). The velocity was 15mL/min (b). The velocity was 25mL/min

Under the conditions of suspension particle size was 30µm and concentration was 200mg/L, the sand column penetration experiments of suspension velocity of 9, 12 and 15mL/min were carried out to study the migration and physical clogging laws of suspension in porous media at different velocity. As can be seen from Fig. 5, when the suspended particle size was relatively large, the relative permeability coefficient decreased rapidly in the 0-3 cm area of the sand column, which was also a typical feature of surface clogging described above. And the larger the flow velocity, the faster the time for obvious surface clogging, the final relative permeability coefficient decreased by about 95%, indicating that the flow velocity mainly affects the clogging time but cannot change the severity of clogging. It is found that the clogging can be restored by backwashing experiment, As shown in Fig. 7, the backwashing experiment data of the suspension at a velocity of 12mL/min were recorded through the pressure sensor, the original condition experiment can instantly restore the permeability coefficient of sand column 0-3 cm area to 80%, and then surface clogging appeared again in a shorter time, which indicated that the pore clogging on the surface of suspended particles in the experiment was mainly due to bridging.



Flig. 7 Variation curves of relative permeability coefficients in different regions at different velocity at 30 µm

The experimental results show that when the particle size of suspended particles is 3 µm and the suspension velocity is low, the suspended particles can migrate smoothly in the porous medium, because the internal pores of the porous medium are much larger than the particle size of suspended particles at this time, and no obvious clogging phenomenon will occur in the short term (Han et al., 2019). While the high velocity can promote the penetration of particles in the whole area of the porous medium, reduce the filtration rate, and then increase the number of particles passing through per unit time and thus accelerate clogging (Feia et al., 2015). However, when the particle size of suspended particles is 30 µm, even when the velocity of suspension is low, the porous medium will be blocked on

the surface of relatively rapid formation, and with the increase of the velocity, the formation and occurrence time of the clogging will be faster.

This indicates that when the particle size of suspended particles is small, or the particle size of suspended particles is much smaller than the internal pores of porous media, the migration of suspended particles in porous media is relatively smooth at a low velocity, and the whole permeation system can maintain a long time without obvious clogging. Higher velocity will accelerate the accumulation and deposition of suspended particles in the whole area of porous media, thus accelerating clogging. When suspended particle size is larger, or the particle size of suspended particles is close to the internal pores of porous media, suspended particles are easy to form a bridge at the entrance which resulting in rapid clogging, and flow velocity can only affect the speed of the clogging, Although the permeability coefficient can be restored by backwashing, it can be seen in the experiment that the relative permeability coefficient can be restored to 80% of the original, but the system is blocked again in about 8h, which is shorter than 12h for the first time.

3.4. System clogging type

The suspension particles we used were mainly single particle size, while the particle size of suspended particles was smaller than the pore size of porous media, and the particle size of the filter screen was larger than the pore size of porous media in this experiment, in this way can we observe some clogging phenomena and mechanisms of suspended particles in porous media. Therefore, for a single pore, the clogging mechanism in the experiment is mainly bridging and aggregation clogging, and sieving does not occur under such conditions. But the clogging types of the whole porous media infiltration system is different from the pore clogging. The experimental phenomena shows that the macroscopic types of clogging are mainly divided into two types: surface clogging and internal deposition clogging (Wang et al., 2012), as shown in Fig. 8. It can be found in the experiment that the surface clogging is obviously caused by the superposition of bridging and aggregation in a single pore, and then accumulated to the pore filled with the filter screen, which is a relatively fast process. When the first layer of clogging is relatively slow, but it is basically an irreversible process.



Fig. 8 Schematic diagram of clogging types under macro conditions



Fig. 9. Variation curve of liquid injection volume and pressure in a liquid injection well of a uranium mine

It is impossible to avoid the clogging completely in the case of in-situ leached uranium deposit running all year round, so it can only prolong the occurrence time of the clogging as long as possible. By simulating the physical clogging experiment of different concentrations and flow rates in porous media, it is found that more surface clogging occurs faster, which leads to filter clogging and even scaling, while the internal deposition clogging is usually a long-term gradual process without obvious clogging phenomenon. This indicates that surface clogging and filter clogging are more common in the ground leaching uranium mining site, resulting in a sudden increase in liquid injection pressure and a sudden drop in pumping fluid volume in a certain period of time, as shown in Fig. 9, which is consistent with the surface clogging phenomenon in our experiment, and it is necessary to be alleviated by hole washing at the situation. Without doubt long-term operation of uranium mines will also lead to internal deposit clogging, but the experimental results show that the single internal sedimentary clogging mainly occurs when the suspended particle size is much smaller than the pores of filter and porous medium and the flow rate is low, Therefore, the clogging should be mainly surface clogging when there are suspended particles of various particle sizes in in-situ leaching uranium mining (without considering aggregates such as floc), which is accompanied by filter clogging and even scaling. So the main direction of clogging in the actual process of in-situ leaching uranium mining is targeted to slow down filter clogging and surface clogging, and irreversible internal deposition clogging after long-term operation should be avoided as much as possible.

4. Conclusion

The simulation experiment of physical clogging caused by suspended particles in porous media in insitu leaching of uranium was carried out in this paper. The physical clogging law of suspended particles in porous media under different suspended particles concentrations (100, 200, 300 and 400 mg/L) and different suspension flow rates (9, 12, 15 mL/min) were studied respectively. The experimental results show that suspended particles concentration and suspension flow rate are both important factors of physical clogging in in-situ leaching of uranium., and some conclusions were obtained: (1) The clogging mechanism of a single pore can't be directly reflected as the macroscopic clogging phenomenon of porous media system, which is generally the superposition effect under the effects of various mechanisms.

(2) When the concentration of suspended particles is 100, 200, 300 and 400 mg/L, the higher the concentration, the faster the permeability of porous media decreases, and the more obvious the clogging phenomenon will be.

(3) When the particle size of suspended particles is 3 μ m and the flow rate is 15 mL/min, the porous medium will not appear clogging, while the whole porous medium will slowly appear internal deposition clogging when the flow rate is 25 mL/min.

(4) Surface clogging occurs when the particle size of suspended particles is $30 \mu m$ and the flow rate is 9, 12 or 15 mL/min, and the higher the flow rate, the faster the clogging will be. Backwashing can alleviate the surface clogging but cannot change the final clogging result.

(5) The clogging in in-situ leaching of uranium is mainly surface clogging combined with the simulation experiment and the actual situation in the field, which is accompanied by filter clogging and even scaling phenomenon. The main research direction of related clogging problem should be to slow down filter clogging and surface clogging.

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