Experimental study on the influence of CO\textsubscript{2} concentration on the crystallization law of wastewater drainage pipes

Yun Li\textsuperscript{a,b}, Zhenjiang Cui\textsuperscript{i}, Shiyang Liu\textsuperscript{a,b,d,*}, Xuefu Zhang\textsuperscript{a,b,d}, Feng Gao\textsuperscript{a,b}

\textsuperscript{a}College of Civil Engineering, Chongqing Jiaotong University, Chongqing 400074, China, email: cqjtulsy@163.com (S. Liu)
\textsuperscript{b}State Key Laboratory of Mountain Bridge and Tunnel Engineering, Chongqing 400074, China
\textsuperscript{c}Hangzhou Road and Bridge Group Co., Ltd., Zhejiang 310011, China
\textsuperscript{d}Institute of Future Civil Engineering Science and Technology, Chongqing Jiaotong University, Chongqing 400074, China

Received 11 December 2022; Accepted 1 October 2023

\textbf{A B S T R A C T}

The crystal blockage of the tunnel drainage system seriously affects the safety and stability of the lining structure of the tunnel, and there are many factors that affect the crystal blockage of the system. CO\textsubscript{2} concentration is one of the factors. This paper uses indoor model test to study the influence of CO\textsubscript{2} concentration on the crystallization of tunnel drainage pipe under different water filling conditions. First, it studies the crystallization amount of each pipe section under six working conditions. According to the results, the crystallization amounts of sections 4 and 5 of the pipe are greater than those of other sections. Section 5 has the highest amount of crystallization. In other words, the crystallization happens more at the outlet section of the horizontal drainage pipe; (2) When the CO\textsubscript{2} concentration between \(10\text{~kg/cm}^2\) and \(15\text{~kg/cm}^2\), the crystallization amount of the test pipe gets an upward trend and when the CO\textsubscript{2} concentration is around \(15\text{~kg/cm}^2\), the crystallization reaches its maximum; (3) When the test tube is filled with water, the amount of crystallization will decrease as the flow rate becomes higher. When the factor of CO\textsubscript{2} concentrations is controlled, the crystallization amount is less when all the pipe is filled with water than when it is half filled. The research results can guide the maintenance and daily monitoring of the tunnel drainage system, and support the formulation of the daily maintenance plan for the drainage system.

Keywords: Tunnel engineering; Drainage pipe; CO\textsubscript{2} concentration; Crystal plugging pipe; Groundwater velocity

1. Introduction

In recent years, China has made great progress in building infrastructure, particularly in tunnel and underground engineering. During tunnel construction, constructors often encounter traversing geological conditions like karst areas and hard water [1]. This often led to the crystallization in the tunnel drainage pipes [2]. Such blockages have severe consequences, causing the malfunction of the tunnel drainage system, the swelling and cracking of the pipe lining, and structural deterioration. It will ultimately jeopardize the safety and reduce the lifespan of the tunnel [3,4].

Currently, numerous scholars, both domestical and international, have conducted research on the patterns of groundwater crystallization. The research findings indicate that insoluble carbonate is the primary component of underground water crystallization in tunnels [5–7]. Li [8] discovered that the key chemical of crystallization is CaCO\textsubscript{3}, which is produced by the reaction between CO\textsubscript{2} and HCO\textsubscript{3} in the water. After it is precipitated from the water, CaCO\textsubscript{3} interacts with Ca(OH)\textsubscript{2} in the concrete used in engineering construction, and the crystallization in groundwater occurs. Zhu et al. [9] investigated the reaction of water and calcium carbonate and found that the dissolution and crystallization
of calcium carbonate were mainly affected by three factors: pH value, bicarbonate ions, and calcium ions. Through experiments, Vetter [10] analyzed the patterns and the influencing factors of barium sulfate’s precipitation. The results provided valuable insights to our analysis of groundwater crystallization in tunnels. Charpentier et al. [11] conducted dynamic experiments to study the scaling phenomena in underwater well equipment. The study identified CaCO$_3$ and BaSO$_4$ as the scaling substances and explained the scaling mechanism. Larsen et al. [12] conducted indoor simulation experiments to study the crystallization mechanism of carbonates. Liu [13] carried out field experiments and used MATLAB to simulate crystallization blockages of tunnel drainage pipes, and provided suggestions to solve this problem. Zhang et al. [14] studied several tunnels in Chongqing, where severe groundwater crystallization blockages occurred. By analyzing the X-ray diffraction spectra of the crystalline substances, they identified that the main component was calcium carbonate in the form of aragonite. Influenced by different environmental factors, the crystalline substances exhibited various forms on the pipe walls. Their study proposed to take preventive measure to prevent crystallization blockages. For example, a special coating can be added to the inner wall of the drainage pipes. Gryta [15], through the study of calcium carbonate crystals, demonstrated that calcium carbonate crystals can be categorized into three main types: calcite, vaterite, and aragonite. Calcite is a carbonate mineral and the most stable polymorph of calcium carbonate (CaCO$_3$) in nature. Vaterite and aragonite are less stable than calcite for their less compact structures. Although they are easy to be removed, vaterite and aragonite may be transformed into calcite and become stable under natural conditions.

After literature review we can find that few studies have focused particularly on the impact of CO$_2$ on the crystallization blockage in tunnel drainage pipes. To investigate the factors that influence the crystallization blockage in tunnels, this paper conducted an indoor model experiment based on the theories about groundwater crystallization. It took transverse drainage pipes that went to the central drainage ditch as the simulated object. The main goal is to examine the effects of CO$_2$ concentration and groundwater flow rate on crystallization.

2. Experimental design

2.1. Selection of experimental materials and parameters

The materials required for the experiment include: a 3.5 m head submersible pump, an experimental water tank, an electronic balance, a CO$_2$ concentration tester, a CO$_2$ generator, U-PVC ball valves, PVC smooth pipes, PVC elbows, PVC tees, PVC reducers, and the prepared experimental solution.

2.2. Experimental implementation

2.2.1. CO$_2$ concentration experiment

This experiment took three reagents to prepare solutions along with pure water, and they are analytical grade sodium bicarbonate (NaHCO$_3$), anhydrous calcium chloride (CaCl$_2$), and anhydrous magnesium sulfate (MgSO$_4$). Sufficient pure water from an ultrapure water machine was added into the water tank, and supersaturated solutions were prepared to reach the same ion proportions as the testing samples from the water in tunnel.

The CO$_2$ concentration experiment includes two stages. At the first stage, $\Phi$50 PVC pipes were divided into different segments, each of which measured 50 cm long. For the convenience of data recording, these segments were then labeled from 1 to 5, with segment 1 closest to the inlet and segment 5 closest to the outlet. After two control groups were established, we chose the conditions of 10 and 20 kg/cm$^2$ CO$_2$ concentrations for analysis (Fig. 1).

The second stage of the CO$_2$ concentration experiment carried out another group of small-scale experiments that focused on five concentration gradients. These CO$_2$ concentrations included 0, 5, 10, 15, and 20 kg/cm$^2$. The experiments aimed at refining the results in a quantitative way. Upon the installation of the four CO$_2$ generators, we used electromagnetic constant-pressure valves to set the CO$_2$ output concentrations to be: 5, 10, 15, and 20 kg/cm$^2$. The control group did not receive any CO$_2$ gas, so it maintained a CO$_2$ concentration of 0 kg/cm$^2$ (Fig. 2).
By integrating theoretical analysis, it is feasible to reflect the effects of CO$_2$ concentrations on the crystallization of underground water in tunnels in a quantitative way.

### 2.2.2. Water filling experiment for drainage pipes

This experiment investigated the crystallization patterns of water in the pipes, both full-pipe and half-pipe, under different CO$_2$ concentrations and water flow rates. The experimental procedures were the same with CO$_2$ concentration experiment.

### 2.2.3. Experimental duration and conditions

The experiment was conducted over eight cycles, each of which lasts 7-d. For data analysis, the experimental conditions were categorized into six groups and labeled as "a, b, c, d, e, and f" (Table 1).

### 3. Discussion and analysis

#### 3.1. Analysis of crystallization location in drainage pipe

The experimental sections were naturally air-dried. The dried pipe section was weighed and after subtracting the pipe’s original weight, we obtained the crystallization amount (Figs. 3–8).

According to data from various working conditions, crystallization blockages in the tunnel drainage pipe are more likely to occur at the outlet end of the pipe, especially in sections 4 and 5 of the pipe.

After the experiments were done, we put high-precision inspection cameras inside the pipe to capture photos of the crystallization. These photos provided a macroscopic view of the crystallization at each pipe section under different experimental conditions. The crystallization amounts increase significantly from pipe section 1 to pipe section 5 under all six conditions. Additionally, pipe sections 4 and 5 exhibit noticeably higher amounts of crystallization compared to other three sections (Fig. 9, under condition c).

#### 3.2. Influence of CO$_2$ concentration on groundwater crystallization

To examine the correlation between CO$_2$ concentration and groundwater crystallization, we conducted experiments by adopting different CO$_2$ concentrations under the same flow rate (Figs. 10–21).

### Table 1

<table>
<thead>
<tr>
<th>Condition no.</th>
<th>Specific conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>No CO$_2$ injection, half pipe flow</td>
</tr>
<tr>
<td>b</td>
<td>No CO$_2$ injection, full pipe flow</td>
</tr>
<tr>
<td>c</td>
<td>CO$_2$ injection at 10 kg/cm$^2$, half pipe flow</td>
</tr>
<tr>
<td>d</td>
<td>CO$_2$ injection at 10 kg/cm$^2$, full pipe flow</td>
</tr>
<tr>
<td>e</td>
<td>CO$_2$ injection at 20 kg/cm$^2$, half pipe flow</td>
</tr>
<tr>
<td>f</td>
<td>CO$_2$ injection at 20 kg/cm$^2$, full pipe flow</td>
</tr>
</tbody>
</table>

The relationships between the crystallization amounts and CO$_2$ concentration for full-pipe and half-pipe flow rates are shown in Figs. 20 and 21.
By comparing the crystallization amounts at three different CO$_2$ concentrations at different pipe sections, our conclusions are as follows:

1. By visually examining the crystallization amounts in both full-pipe and half-pipe flow rates, the amount of crystallization at CO$_2$ concentrations of 10 and 20 kg/cm$^2$ is significantly higher than 0 kg/cm$^2$ of CO$_2$. Thus, adding CO$_2$ to the pipe enhances the generation of crystallization.

2. Under 10 or 20 kg/cm$^2$ CO$_2$ concentrations, the difference in the crystallization amounts between at pipe sections are negligible. In fact, the crystallization amount is slightly higher at the 10 kg/cm$^2$ concentration than at the 20 kg/cm$^2$ concentration. This suggests that increasing CO$_2$ concentration from 10 to 20 kg/cm$^2$ does not further promote crystallization. We can conclude from this observation that the chemical reactions in Eqs. (1) and (2) will undergo a reversal when the CO$_2$ concentration reaches a particular threshold, as a large amount of high-concentration CO$_2$ dissolves into water. This prevents from Ca$^{2+}$ and Mg$^{2+}$ ions in the solution from crystalizing into insoluble carbonate salts.

$$\text{Ca}^{2+} + 2\text{HCO}_3^- \leftrightarrow \text{CaCO}_3 + \text{CO}_2 + \text{H}_2\text{O} \quad (1)$$

$$\text{Mg}^{2+} + 2\text{HCO}_3^- \leftrightarrow \text{MgCO}_3 + \text{CO}_2 + \text{H}_2\text{O} \quad (2)$$

The CO$_2$ concentrations adopted in the initial experiment were wide in range, so we also carried out smaller scale tests to obtain a more precise understanding of the relationship between the CO$_2$ concentration and crystallization. At this stage, the concentration gradients were set to be 0, 5, 10, 15, and 20 kg/cm$^2$. After the tests, the ion composition and content of the water samples were measured.
The concentration of $\text{HCO}_3^-$ ions in the solution is presented in Table 2. We visualize the data of the table into a graph below to show pattern of $\text{HCO}_3^-$ ion concentration in the solution as it varies with the $\text{CO}_2$ concentration (Fig. 22).

Based on the changing trend of $\text{HCO}_3^-$ ion concentration and $\text{CO}_2$ concentration, the following conclusions can be drawn. The $\text{HCO}_3^-$ ion concentration in the solution increases with the $\text{CO}_2$ concentration, causing more precipitation. This is because $\text{CO}_2$ reacts with water to produce $\text{H}_2\text{CO}_3$, which then dissociates into $\text{HCO}_3^-$ and $\text{H}^+$. $\text{HCO}_3^-$ can further dissociate into $\text{CO}_3^{2-}$. These $\text{CO}_3^{2-}$ ions serve as reactants for the precipitation of $\text{CaCO}_3$ and facilitate the precipitation. When the $\text{CO}_2$ concentration ranges from 0 to 15 kg/cm², the $\text{HCO}_3^-$ ion concentration in the solution gradually increases. However, when the $\text{CO}_2$ concentration reaches 20 kg/cm², the concentration of $\text{HCO}_3^-$ ion starts to decline. Based on our observation, the crystallization amount in the pipeline is greater when the $\text{CO}_2$ concentration is between 10 and 15 kg/cm² than when it is 20 kg/cm². As the $\text{CO}_2$ levels we set are still intermittent in the supplementary tests, it is impossible to determine the exact $\text{CO}_2$ concentration for the maximal crystallization amount. However, based on the results of the second-stage experiment, it can be inferred that the crystallization amount is maximized when the $\text{CO}_2$ concentration is around 15 kg/cm².

3.3. Influence of groundwater flow rate on crystallization

The influence of groundwater flow rate on crystallization was investigated by analyzing the effect of flow rate on the crystallization, in which the full pipe and half pipe were considered as the only variables and the $\text{CO}_2$ condition was controlled. Experimental data were presented in a line graph. Section 5 was taken as an example for its special location. As other sections followed the same pattern, we will not further elaborate here (Figs. 23 and 24).

The line graph above depicts the crystallization amount in each section of the pipe under 0 kg/cm² $\text{CO}_2$ condition. According to the graph, the crystallization amounts at the half-pipe flow rate exceeded those of the full-pipe flow rate in all sections.
Fig. 15. Crystallization quantity in full-pipe flow state of pipe section 3.

Fig. 16. Crystallization quantity in half-pipe flow state of pipe section 4.

Fig. 17. Crystallization quantity in full-pipe flow state of pipe section 4.

Fig. 18. Crystallization quantity in half-pipe flow state of pipe section 5.

Fig. 19. Crystallization quantity in full-pipe flow state of pipe section 5.

Fig. 20. Total crystallization amount at various CO₂ concentrations in half-pipe flow state.
According to our detailed analysis, under 0 kg/cm$^2$ of CO$_2$ concentration, and both in full-pipe and half-pipe conditions, the increase of crystallization amount gradually stabilized with only a marginal increment during the third and fourth cycles (from day 21 to day 35). This is because initial crystal nuclei required time to form and adhere to the inner walls of the pipe. This was a gradual change, during which the inner walls became rougher and rougher. Thus, crystallization maintained a substantial growth rate during the first three cycles (before day 21). During the fifth cycle (after day 35), the crystallization in the drainage pipe began to step up. The crystalline deposits on the pipe walls consolidating into more robust crystal blocks which attracted more crystals to gradually attach to them. The accumulation led to a continuous increase in crystallization amount. The graph clearly shows how the flow rate of the water affect the crystallization amount in the tunnel drainage pipe. It should be noted that the crystallization amount for the half-pipe consistently exceeds that of the full pipe throughout the experiment (Figs. 25 and 26).

According to observation of experimental data, a similar phenomenon to the 0 kg/cm$^2$ CO$_2$ condition has occurred. Additionally, at the end of the 8th cycle, the sections 4 and 5 of the pipe, under the 10 kg/cm$^2$ CO$_2$ concentration, crystallization increases more at the half-pipe flow rate than at the full-pipe flow rate. Therefore, CO$_2$ concentration's influence on solution crystallization is more significant at the half-pipe flow rate than at the full-pipe flow rate (Figs. 27 and 28).

The analysis of the experimental results reveals that under the condition of 20 kg/cm$^2$ CO$_2$, the outcomes are similar to the two previous cases. However, we have observed an interesting phenomenon that in the eighth cycle, both

**Table 2**

<table>
<thead>
<tr>
<th>CO$_2$ concentration (kg/cm$^2$)</th>
<th>HCO$_3^-$ (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>126</td>
</tr>
<tr>
<td>5</td>
<td>129</td>
</tr>
<tr>
<td>10</td>
<td>133</td>
</tr>
<tr>
<td>15</td>
<td>135</td>
</tr>
<tr>
<td>20</td>
<td>132</td>
</tr>
</tbody>
</table>

**Fig. 21.** Total crystallization amount at various CO$_2$ concentrations in full-pipe flow state.

**Fig. 22.** Variation of HCO$_3^-$ ion concentration with CO$_2$ concentration in solution.

**Fig. 23.** Crystallization amount of pipe section 5 at 0 kg/cm$^2$ CO$_2$.

**Fig. 24.** Total crystallization amount at different flow rates of 0 kg/cm$^2$ CO$_2$.
sections 4 and 5 in the pipe witness a noticeable decline in the growth rate of crystallization. This phenomenon can be attributed to the significant increase in CO\textsubscript{2} concentration to 20 kg/cm\textsuperscript{2} in this experiment. This influence is bigger on the full-pipe flow rate. According to previous experiments involving CO\textsubscript{2} concentration variations, when the CO\textsubscript{2} concentration is 20 kg/cm\textsuperscript{2}, the crystallization amount within the pipe is lower than the amount at a CO\textsubscript{2} concentration of 10 kg/cm\textsuperscript{2}. This discrepancy arises because a CO\textsubscript{2} concentration of 10 kg/cm\textsuperscript{2} leads to a higher concentration of CO\textsubscript{3}\textsuperscript{2–} ions in the solution compared to a CO\textsubscript{2} concentration of 20 kg/cm\textsuperscript{2}. Moreover, this phenomenon has a more evident effect on the full-pipe flow rate, thus causing the observed deceleration in the crystallization growth rate under the full-pipe flow rate state.

According to the column histogram, the overall crystallization amounts under the 0 kg/cm\textsuperscript{2} CO\textsubscript{2} concentration, both the full-pipe flow rate and the half-pipe flow rate are less than those under the conditions of 10 kg/cm\textsuperscript{2} CO\textsubscript{2} and 20 kg/cm\textsuperscript{2} CO\textsubscript{2} concentrations. This indicates that under the same flow rate conditions, variations in CO\textsubscript{2} concentration in the pipeline can significantly affect the formation of crystalline structures. This is because the dissolution of CO\textsubscript{2} in water will generate a large amount of CO\textsubscript{3}\textsuperscript{2–} ions, providing abundant ion sources for the formation of CaCO\textsubscript{3} crystals, thus facilitating the precipitation.

In summary, this paper has conducted a comparative analysis of the crystallization amounts in pipe sections under full-pipe flow rate and half-pipe flow rate conditions for three different CO\textsubscript{2} concentrations—0, 10, and 20 kg/cm\textsuperscript{2}. It reveals that the higher the flow rate of the solution within the drainage pipe, the better the inhibitory effect on crystallization. In this study, the flow rate of the solution in the drainage pipe was controlled below the full-pipe level. In other words, the flow velocities were equal under both flow rate conditions. However, when the flow rate exceeds the full-pipe level, the flow velocity will continue to increase. At higher flow velocities, the effect on crystallization may not follow the observed trend in this experiment. Subsequent research can further investigate the influence of flow velocity on crystallization of underground water in tunnels.

4. Conclusions

This study investigated the influence of CO\textsubscript{2} concentration on the crystallization of underground water in tunnel drainage pipes through indoor experiments, and the conclusions are as follows:
(1) Under six different working conditions, the crystallization amounts in pipe section 4 and pipe section 5 are found to be greater than those in the remaining sections, with pipe section 5 showing the highest crystallization amount. Therefore, it can be inferred that pipe section 4 and pipe section 5 are most prone to crystallization in this experiment, and the section near the outlet is most susceptible to crystallization in the transverse drainage pipe.

(2) The CO$_2$ concentration experiment continued with supplementary tests after the first-stage experiment. The data show that between 10–15 kg/cm$^2$ of CO$_2$ levels, the largest quantity of crystallization occurs. As the CO$_2$ levels we set are still intermittent in the supplementary tests, it is impossible to determine the exact CO$_2$ concentration for the maximal crystallization amount. However, based on the test, it can be inferred that the crystallization amount can reach its maximum at around 15 kg/cm$^2$ CO$_2$ concentration.

(3) The flow-rate experiment on tunnel groundwater controls the water flow rate inside the pipes and makes comparison experiments under different CO$_2$ concentrations. The results indicate that higher flow rate can better inhibit crystallization within the full-pipe flow rate range. By comparing the flow rates’ effects under different CO$_2$ concentrations, we find that the crystallization amount at full-pipe flow rate is smaller than that at half-pipe flow rate.

This model experiment aims to simulate the environmental conditions in tunnel drainage systems, but other factors in the field drainage pipes cannot be ignored, including the effects of sediment and debris from surrounding rocks. The experiment was conducted at room temperature without considering the influence of temperature on crystallization. Future experiments should consider these factors if conditions allow. Furthermore, this study focused on exploring fundamental theories and conducted indoor simulation experiments. The findings have not been applied in field tests of engineering projects. In the future, it would be beneficial to carry out field tests that consider the features of the tunnel project to further validate the findings and their application.

Funding

This research was financially supported by the Postdoctoral Program of Chongqing Natural Science Foundation (No. CSTB2022NSCQ-BHX0715), the Sub Project of National Key R&D Plan (No. 2021YFB2600103-01), the cooperation between Chongqing University and the Institute of Chinese Academy of Sciences (No. HZ2021009), the 2022 Chongqing Jiaotong University High Level Talent Scientific Research Start-up Fund Project (second batch) (No. XJ202204201), and the Chongqing Talent Innovation and Entrepreneurship Leading Talent Project (Grant No. CQYC202203091118).

Informed consent statement

Not applicable.

Data availability statement

The study did not report any data.

Conflicts of interest

The authors declare that there is no conflict of interest.

References