Hindawi
Geofluids
Volume 2019, Article ID 9218916, 12 pages
https://doi.org/10.1155/2019/9218916

Research Article

Experimental Study on Backfilling Mine Goafs with Chemical Waste Phosphogypsum

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Received 31 July 2018; Revised 17 October 2018; Accepted 28 October 2018; Published 16 January 2019

Academic Editor: Mohammad Sarmadivaleh

To explore the feasibility of cemented paste backfill with phosphogypsum (PG), bleeding water and rheological tests (slump and on-site pipeline loop tests) were performed with PG backfill slurry (PGBS). In the bleeding water test, the PGBS concentration with minimal bleeding water was measured between 60.87 and 67.61%; in the rheological slump test, values of 61 to 68% were determined for the on-site pipeline loop test. The rheological pipeline loop test demonstrated that the resistance coefficient is lowest when the concentration is no higher than 65%. Through industrial experiments, PG slurry with a concentration of 64%–65% backfill was successfully applied to the goaf. The experimental results demonstrate that PGBS with characteristics of "less bleeding water" and "improved pumpability" is obtained when its concentration is between 61 and 65%. Paste-like PG slurry was proven to be optimal for cemented PG backfilling technology.

1. Introduction

Phosphogypsum (PG) is a by-product of the reaction between phosphate rock ($Ca_5(PO_4)_3F$) and sulfuric acid when phosphoric acid is produced in chemical plants. Approximately 5 tons of PG will be produced when 1 ton of phosphate fertilizer is manufactured [1–3]. The structure of PG is similar to that of natural gypsum, which is mainly composed of $CaSO_4{\cdot}2H_2O$, accounting for approximately 90% of PG components [4]. However, the remaining 10% of PG components contain several deleterious impurities, such as uncomposed phosphate rock, unwashed phosphoric acid and calcium fluoride, iron aluminides, and acid insolubles. The main harmful impurity distribution is displayed in Table 1.

Owing to the harmful impurities in PG, its utilization worldwide is rare. Increases in PG globally amount to 280 million tons per year, and most of these are stacked in slag disposal pits, covering an area of approximately 2000 hectares [5, 6]. To address the problem of stockpiled PG, numerous countries devoted themselves to exploring PG resources. Certain countries use PG as a building material or soil fertilizer, accounting for 15% of the total PG [6]. However, the remaining 85% of PG is placed in stockpiles on the ground without any process applied, which not only require large amounts of ground space but also cause critical environmental problems, such as chemical or radioactive pollution [7, 8].

However, in China, PG is used in a large scale as an underground mine backfill aggregate, in addition to road construction materials, owing to its extremely low radioactivity [9]. PG as a filling aggregate has been successfully used in mine backfills in some of China's phosphate mines, which can consume a large amount of stockpiled PG [10, 11]. A sketch map of PG underground mine backfill is presented in Figure 1. However, using backfill PG in the mine stopes has two advantages, namely, (a) lower cost of backfill material because of the chemical waste and (b) significantly reduced ground environmental contamination. For example, 6 million tons of PG is produced annually in the Kailin fertilizer plant, and 1.5 million tons of PG is consumed in mine backfills, accounting for 25% of the total produced PG [12].

PG as a filling aggregate has been successfully used in some of China's phosphate mines because transport pipelines

Table 1: Main impurity content distribution in PG [4].

Impurity types	Solubility	Main form of impurities
Phosphate compounds	Dissolvable	H_3PO_4, $(H_2PO_4)^-$, and $(HPO4)^{2-}$
	Eutectic	$CaHPO_4 \cdot 2H_2O$
	Indissolvable	Phosphate complex (with Fe, Al, alkali metal, etc.)
Fluoride	Dissolvable	$(SiF_6)^{2-}$ and F^-
	Indissolvable	$CaSiF_6$, CaF_2, and Na_3AlF_6
Organics	Indissolvable	Defoaming agent and scale inhibitor
Heavy metal	—	Cr, Cu, Zn, Cd, etc.
Other impurities	Dissolvable	K^+ and Na^+
	Indissolvable	SiO_2, Fe, Sr, Mg oxide, and clathrate

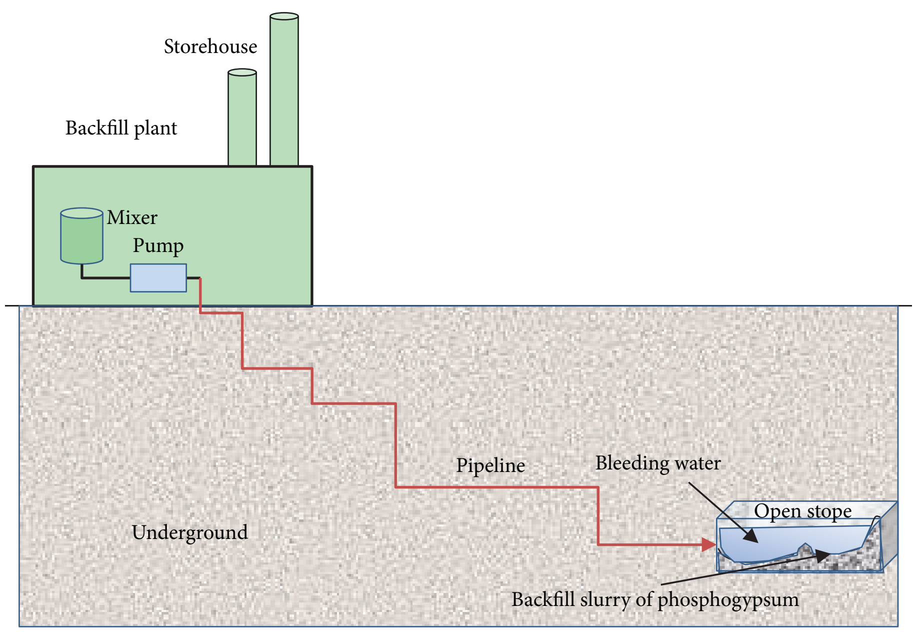

Figure 1: Sketch map of the PG underground mine backfill.

being loaded with PG slurry used for mine backfills are generally in several kilometers long. However, the mass concentration of PG backfill slurry (PGBS) is generally as low as 40%–50%. Therefore, a large volume of water with harmful substances bleeds from the goafs following the mine backfills, thus polluting the underground environment [5, 13–16].

To reduce bleeding water, the transported concentration of PGBS should be increased. However, low flowability of the PG slurry will be achieved because the frictional resistances in the pipeline increase with the increase in concentration. The pipeline will be blocked and the boost phenomenon will occur once the concentration reaches a critical value [17, 18]. Hence, the dilemma in ensuring the optimal concentration of PG backfill slurry PG is low concentration with high pumpability or high concentration with low pumpability (Figure 2). Moreover, determining the suitable concentration of PGBS with higher pumpability and less toxic bleeding water is important. In this study, the transportation performance of high-concentration (less bleeding water) PGBS in the pipeline is investigated.

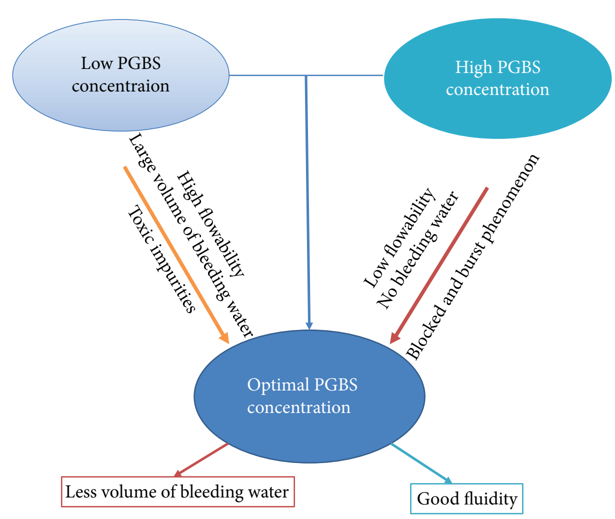

Figure 2: Optimal PGBS concentration.

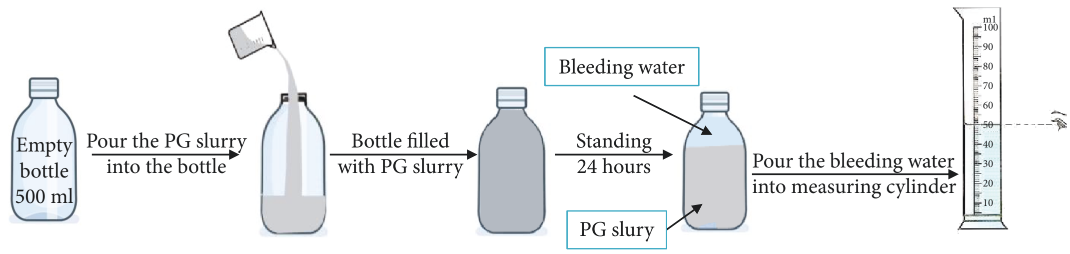

Figure 3: Testing processes of the bleeding rate [25].

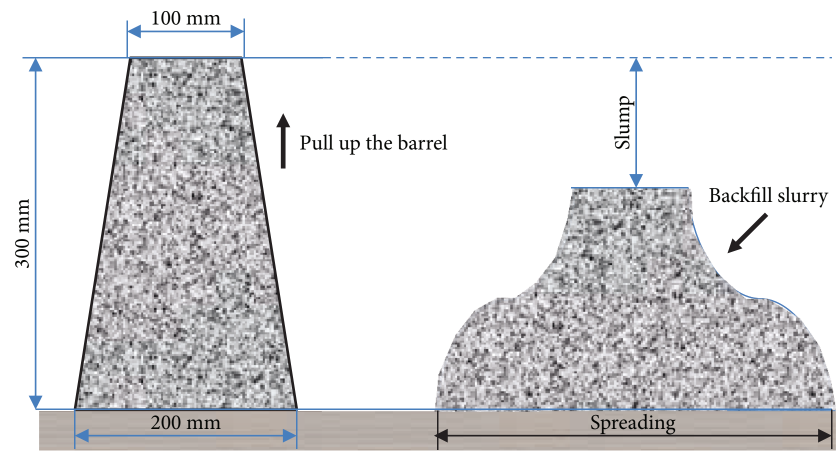

Figure 4: Schematic of the PGBS slump [22].

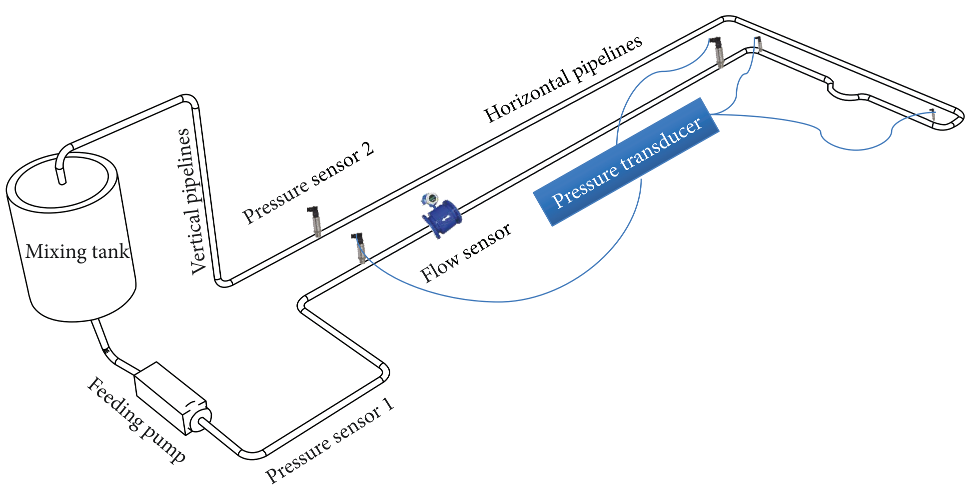

Figure 5: Sketch map of pipeline of PGBS loop test.

2. Experimental Methods

All the experiments are single-factor experiments. In order to get the bleeding water rate, slump value, and frictional resistance of PGBS, bleeding water test, slump test, and loop test are conducted, respectively.

2.1. Determination of the Bleeding Water Rate of High-Concentration PGBS. A high-concentration of PGBS, which also refers to PGBS with minimal amount of bleeding water, is defined by the bleeding water rate (BWR) of the PGBS. The BWR refers to the ratio of bleeding water and containing water of the PGBS. Low BWR is an important indicator of high-concentration PGBS. Low BWR of saturated backfill slurry is between 1.5 and 5% from the perspective of soil mechanics [16]. Some Chinese mines also used low BWR as standard of paste or paste-like backfill. Paste or paste-like backfill refers to backfill slurry that has low water contents and high concentration [19]. Transport properties of paste and paste-like backfill slurry are different from

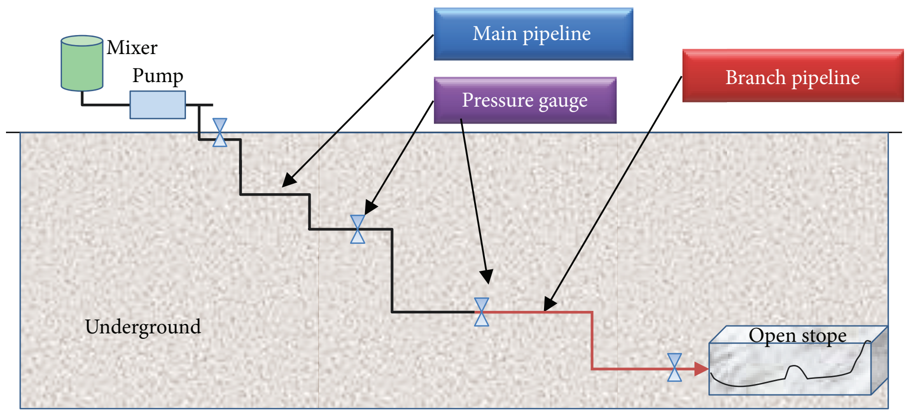

Figure 6: Sketch map of the PG backfill industrial test.

Figure 7: Bleeding water of PGBS in the bottles.

low-concentration slurries; they have the characteristics of less bleeding water and consistency [19–21]. Therefore, a high concentration (paste) of PGBS is calculated under the condition of BWR of 1.5%–5%, according to the relationship curve between the BWR and PGBS concentration. The test method is as follows. The PGBS is composed of PG (M_1), binder slag (M_2), and water (M_3). First, 500 ml PGBS (M_4) is poured into an empty bottle. All bottles have the same capacity even though the shapes of the bottles are different. The bottle is retained standing in one place for 24 hours. Second, the bleeding water in the top layer is poured into the cylinder, and its volume V measured. Finally, the BWR is calculated using equation (1). The testing processes are illustrated in Figure 3.

$$B_r = \frac{V\rho(M_1 + M_2 + M_3)}{M_3 \times M_4}, \quad (1)$$

where B_r is the bleeding rate, V is the volume, ρ is the density, M_1 is the PG mass, M_2 is the slag mass, M_3 is the water mass, and M_4 is the PGBS mass.

2.2. Determination of the Slump Value of High-Concentration PGBS. The slump value is an index of the PGBS liquidity

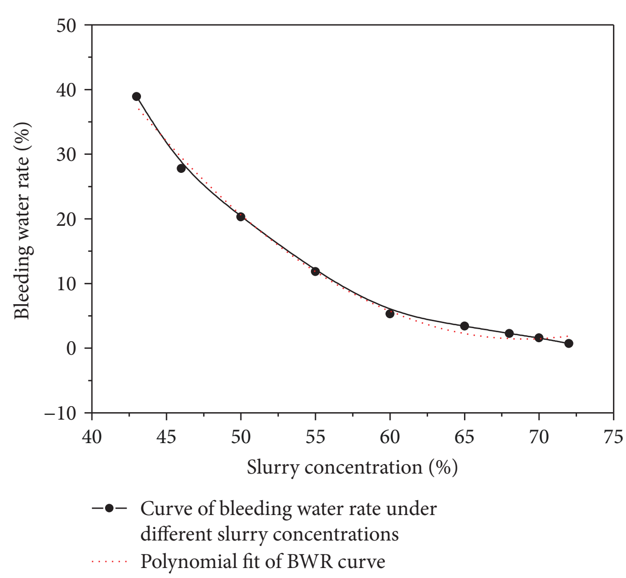

Figure 8: Relation curve of the BR and PGBS concentration.

[22, 23]. If the PGBS slump value is lower than the internationally recognized paste slump limit (i.e., 18–25 cm), the PGBS with a high concentration cannot be transported

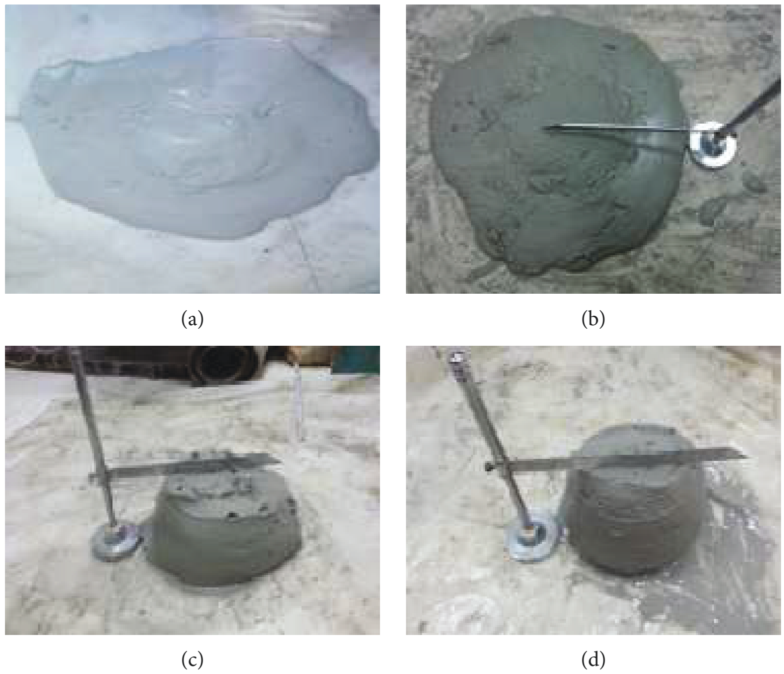

FIGURE 9: Results of slump tests at mass fractions of (a) 65%, (b) 68%, (c) 70%, and (d) 72%.

[19–21]. Therefore, to determine the high concentration of the PGBS, the relationship curve between the concentration and slump is drawn according to the slump test. The slump test method is as follows. First, the configured PGBS is poured into a slump barrel, which is pulled up after being filled with PGBS. Then, the PGBS will collapse during the process of pulling up the barrel. The slump value is equal to the height difference between the barrel and slump, as illustrated in Figure 4. Therefore, the relationship between the PGBS concentration and slump is obtained. According to the minimum paste slump value, the maximum PGBS concentration is determined.

2.3. Determination of the Frictional Resistance of PGBS. The loop test was first proposed by a U.S. mining authority in 1994, and the pipeline loop is an instrumented and closed-circuit pipeline system powered by a pump [19]. This loop is constructed to measure the transport characteristics of high-concentration backfill, such as pressure loss and flow rate, as illustrated in Figure 5. A similar pipeline loop is constructed to measure the transport characteristics of PGBS. The friction resistance loss is determined under different pump pressures and flow rates. Finally, the relationship between friction resistance and concentration is obtained.

2.4. On-Site Industrial Tests. Industrial tests were performed to verify whether high concentrations of PGBS can be backfilled to the goaf. The methods are as follows. First, high-concentration PGBS is transported to the goaf through a pipeline of several kilometers. Then, various instruments are installed on the pipeline, as illustrated in Figure 6. After the experiments, whether high-concentration PGBS can be backfilled to the goaf through the long-distance pipeline is verified according to the test data.

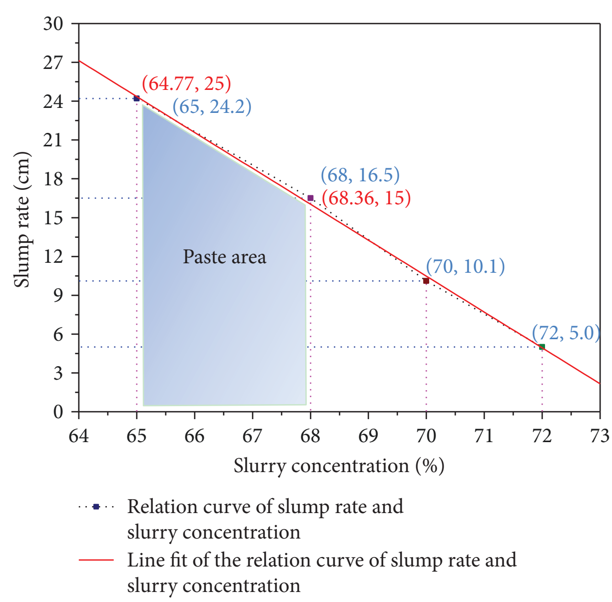

FIGURE 10: Relation curve of the slump and PGBS concentration.

3. Results

3.1. PG Backfill BWR Test. Bleeding water is water naturally separated from the backfilled filling body when the bottle stands for a short time, as illustrated in Figure 7. The products of bleeding water are caused by PG aggregate particles in the backfill slurry, which cannot absorb all the mixing water. The BR refers to the ratio of bleeding water and total water content in the backfill slurry. PGBS is composed of PG and binder slag. According to previous research, the optimal mass ratio of PG to binder slag is 4 : 1 [24]. A measuring

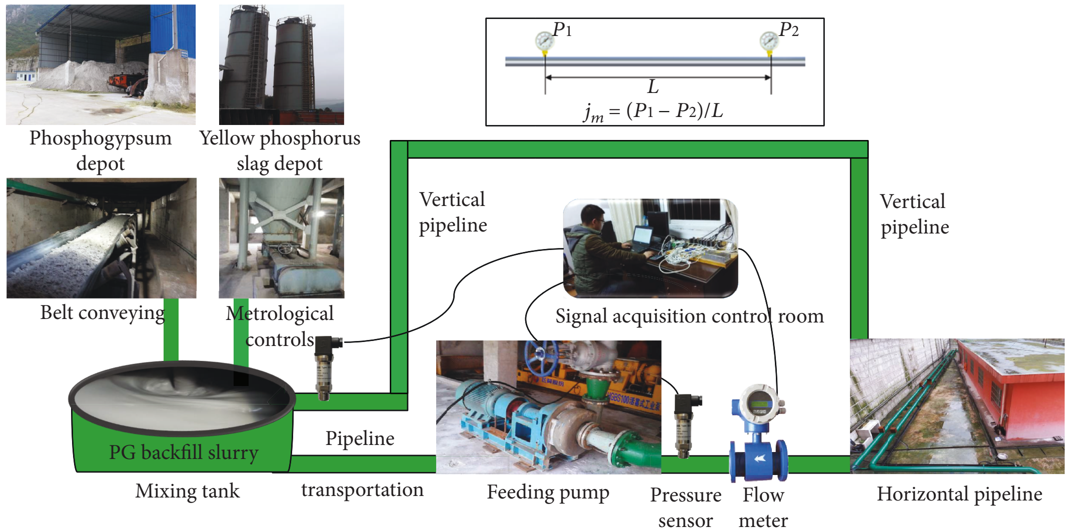

FIGURE 11: Schematic of the loop test system with several measuring apparatus [25].

cylinder is used to determine the BWR of the PGBS. Nine concentrations of the bottles are 43%, 46%, 50%, 55%, 60%, 65%, 68%, 70%, and 72%, with BR values of 38.92%, 27.80%, 20.30%, 11.85%, 5.32%, 3.42%, 2.26%, 1.58%, and 0.72%, respectively.

Through regression analysis, the relationship curve between the concentration and BWR of the PGBS can be obtained, as illustrated in Figure 8. According to the definition of paste slurry bleeding rate between 1.5% and 5% [16], a concentration of PGBS between 60.87 and 67.61% is calculated.

3.2. Fluidity Experiment of PGBS: Slump Test. The fluidity index is measured using a slump or the Vebe consistometer. The test method involves using slump barrels, with top and bottom diameters of 100 and 200 mm, respectively, to measure the height difference after the slurry collapses, as illustrated in Figure 4. The PGBS is poured into the barrel and the barrel is immediately pulled up. The height difference, also known as slump, will be generated during the process of natural PGBS collapse. The on-site PG backfill slump tests are illustrated in Figure 9 [25].

The slump values of 24.2, 16.5, 10.1, and 5.0 cm are measured according to PGBS concentrations of 65%, 68%, 70%, and 72%, respectively. To ensure the fluidity of PGBS in the transported pipeline, the PG slump value should be at least 15 cm according to the slump requirement of paste backfill, and the corresponding PGBS concentration is 68.36%, as illustrated in Figure 10. Therefore, combined with the BWR test range of 60.87%–67.61%, PGBS concentrations of 61%, 62%, 63% 64%, 65%, 67%, 66%, and 68% are selected in performing the loop test.

3.3. Loop Test. The loop test provides an effective means of investigating the frictional resistance of PGBS in pipeline transportation. Its advantages are as follows. (a) The expenditure of pipeline construction (steel tubes, etc.) and backfill materials (PG, slag, etc.) is lower. (b) Installation of various testing instruments is more convenient. (c) Control of the experimental operation is more precise. (d) The test results can be obtained rapidly. The pipeline system consists of four parts, namely, power system (the measurement range of the centrifugal pump is 0–5 MPa), mixing system (the volume of the mixing tank is $5 \times 3 \times 2$ m), testing system (various measurement gauges, such as pressure sensors, thermometers, and flow meters), and conveying system. The conveying system is a circular pipeline ($D = 200$ mm) with a 130 m length, which is composed of straight, bent, and vertical pipes. An actual underground PG backfill is simulated when the loop test systems are continuously operated. The loop test system and its measurement gauges are illustrated in Figure 11.

After the installation of the loop testing system, the PGBS transportation experiment is performed. According to a previous schedule, the concentrations of the PGBS are 61%, 62%, 63%, 64%, 65%, 66%, 67%, and 68%. The PGBS concentration is adjusted by adding dry ingredients, such as PG and slag, to the mixing tank. For different PGBS concentrations, parameters such as the flow rate and pressure are measured by gauges, which are installed on the pipeline of the loop system. According to the computational formula of hydromechanics and the measured data, rheological parameters, such as frictional resistance, are calculated. The relation curve of the friction resistance and concentration is shown in Figure 12.

The representative concentrations of PGBS, such as 62%, 64%, 65%, 66%, 67%, and 68%, are selected to analyze the relationship between the flow velocity and friction resistance. According to the formula of fluid mechanics, the PGBS flow rate is directly proportional to the friction resistance under certain slurry concentrations and pipeline diameters [19, 26]. Therefore, according to the test data, the linear relationship curve of the PGBS between the flow rate and friction resistance is fitted, and the filling resistance coefficients under different concentrations are calculated, as indicated in Table 2.

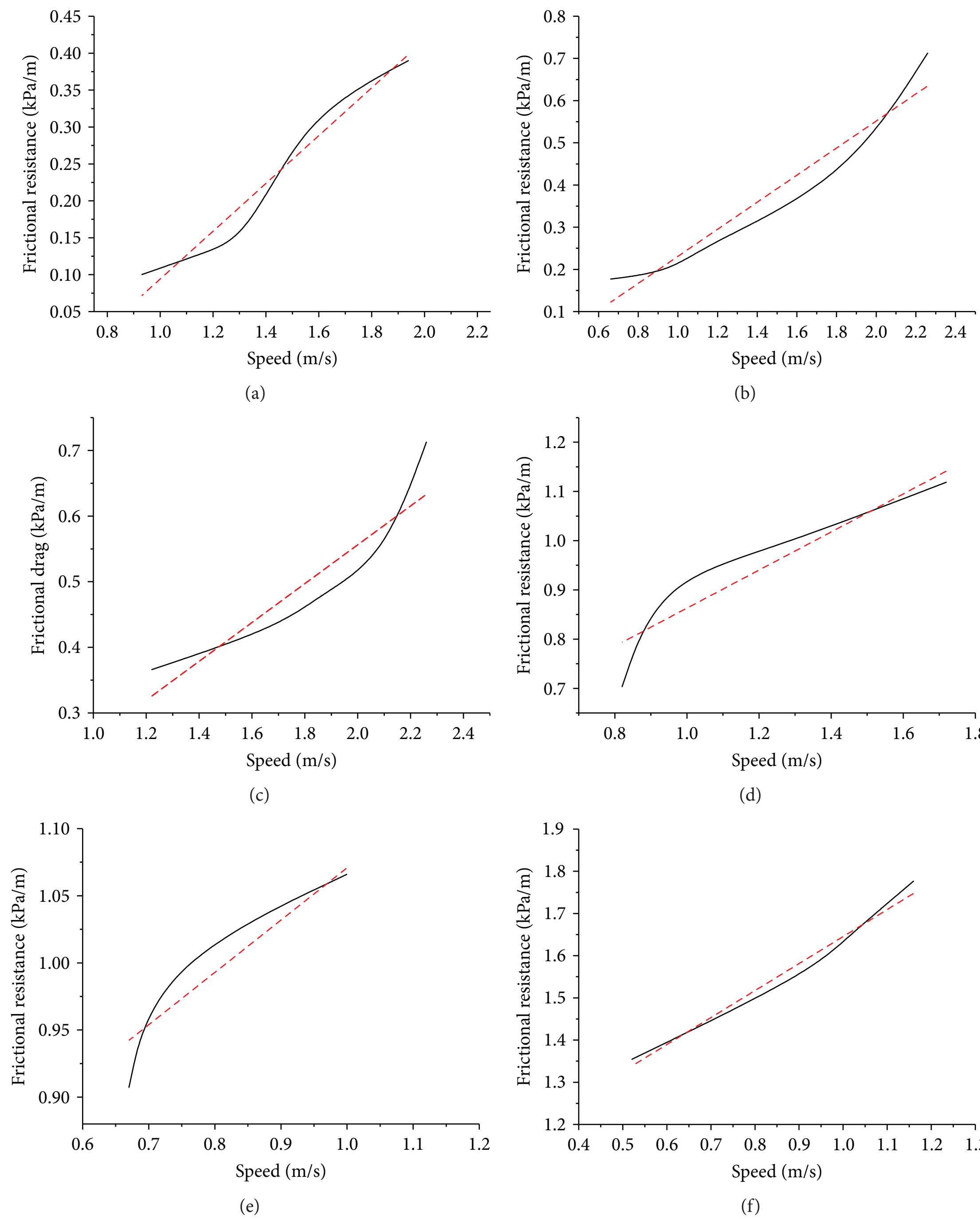

FIGURE 12: Relation curve of on-way resistance and PGBS speed in different backfill concentrations. Graphs in (a–f) indicate the PGBS concentrations of 62%, 64%, 65%, 66%, 67%, and 68%, respectively.

The results demonstrate that the friction resistance is linear with the PGBS flow rate. The resistance coefficient K (slope) reflects the friction resistance growth rate. The higher the drag coefficient is, the more difficult the backfill of the PG slurry in the stope becomes. The K value is calculated according to the linear equation fitted in Figure 12. The results of the study on the filling resistance coefficient (K) demonstrate that when the backfill concentration is greater than 65%, the backfill resistance coefficient is sharply increased (Figure 13). The results demonstrate that to ensure high-efficiency backfill, the PGBS concentration should not be higher than 65%.

3.4. Industrial Pipeline Transportation of PGBS. According to the loop test results, higher transport efficiency will be achieved when the PGBS concentration is lower than 65%. Therefore, based on the loop test, an industrial pipeline transportation of high-concentration PGBS was performed. The purpose of the industrial experiment is to verify whether

Table 2: Linear relationship curve ($y = kx + b$) of PGBS.

PGBS concentration (%)	Regression equation ($y = Kx + b$)	Slope (K)	Square relation
62	$y = 0.324x - 0.23$	0.32	0.924
64	$y = 0.320x - 0.089$	0.32	0.876
65	$y = 0.296 - 0.035$	0.30	0.752
66	$y = 0.387 + 0.476$	0.39	0.723
67	$y = 0.389 + 0.681$	0.39	0.747
68	$y = 0.640 + 1.01$	0.64	0.960

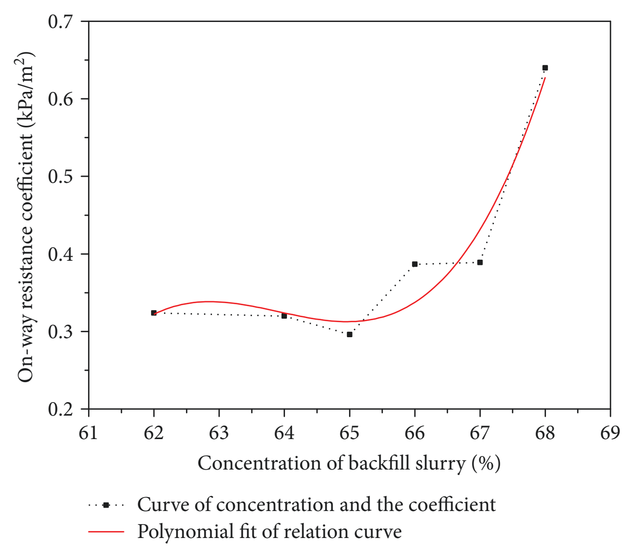

Figure 13: Relation curve of on-way resistance coefficient and slurry concentration.

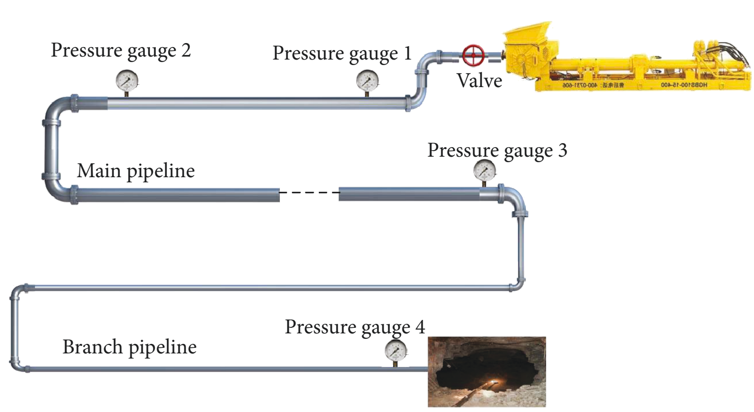

Figure 14: Backfill map of industrial experiment [25].

the optimal PGBS concentration obtained from the loop test can be used in the actual stope backfill. A practical stope is selected in the mine. The diameters of the main and branch pipes are 125 mm and 100 mm, respectively, and the corresponding lengths are 1535 m and 1236 m. A pressure gauge is installed at the outlet of the backfill industrial pump to measure the initial pressure (pressure gauge 1). Up to three pressure gauges are installed in other locations along the pipelines, as illustrated in Figure 14.

The feeding pump capacity is only 5 MPa, which cannot meet the requirements of industrial backfill. Therefore, the feeding pump is replaced by an industrial pump with a capacity of 11.5 MPa. Although the PGBS concentration is set to 65%, the actual concentration may fluctuate between 64%

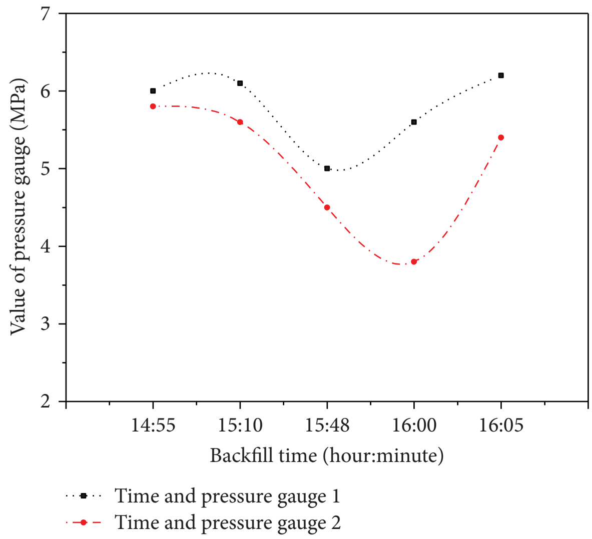

Figure 15: Measured pressure values of gauges 1 and 2 in industrial test.

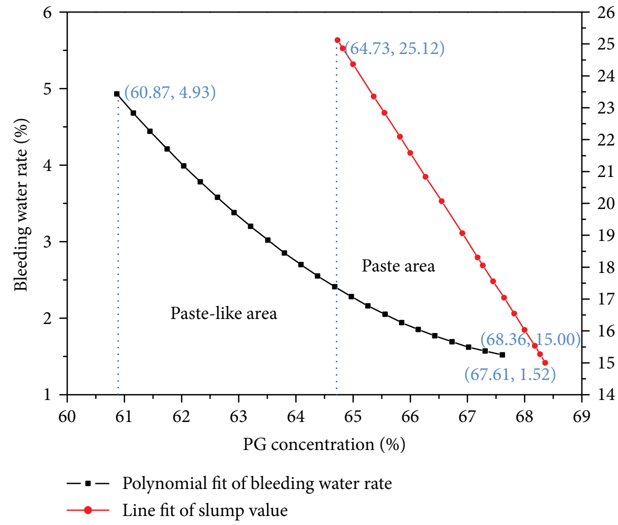

Figure 16: BWR and slump rate of PGBS.

and 65% owing to industrial errors. The backfill pressures are recorded continuously, as shown in Figure 15.

The industrial tests demonstrate that the initial pressure (pressure gauge 1) is approximately 6 MPa, which is substantially less than the maximum rated pressure of 11.5 MPa. Therefore, through the industrial pipeline transport experiment, high concentrations of PG backfill to the underground goaf are considered practical. However, if the PGBS concentration continues to increase, pipeline burst occurs owing to the limited pressure-bearing pipeline. Therefore, a stronger pressure-bearing pipeline and pump should be developed.

4. Discussion

Tailings as mine backfill materials have been applied successfully worldwide [27]. However, PG as a paste-like backfill material has never been attempted in China. Therefore, the fluidity of PGBS should be studied in view of using pipelines to transport the backfilling materials. The feasibility of PG paste-like backfill in underground goaf is discussed by the flow performance study through bleeding rate, slump, and loop tests. The experiment results are analyzed and discussed as follows.

4.1. The Difference in the PG High Concentration Value between the BWR and Slump Value Can Be Considered as Paste-Like Backfill. Slump value is commonly used globally to define the paste, and the paste slump value is often 15–25 cm [19–21]. In China, the BWR is also used to define the concentration range of paste: the BWR of the paste slurry is between 1.5 and 5% [16]. According to the polynomial fit of BWR curve as illustrated in Figure 8, the concentration ranges measured by the BWR of the PG paste are between 60.87 and 67.61%. However, according to the line fit of slump value curve as illustrated in Figure 10, the concentration range measured by the slump value of PG paste (15–25 cm) is between 64.73 and 68.36%. Given that no uniform definition according to BWR when using paste backfill in other countries is provided, PG concentrations between 60.87% and 64.73% should be considered as the paste-like backfill, as illustrated in Figure 16. Paste-like slurry is a type of paste with a certain amount of bleeding water [16], where the flow performance is close to the paste shape, as illustrated in Figure 17. Compared with paste PG, paste-like PG exhibits lower frictional resistance. Thus, it has superior fluidity than paste PG.

4.2. The Slump Value Can Be Used for the Preliminary Analysis of the PGBS Fluidity. In this study, the inflection point of the resistance coefficient is approximately 65% (Figures 13 and 18), and the slump value of 25.12 cm corresponds exactly to a PG concentration of 64.73% (Figure 18). This demonstrates the consistent results between the slump value measured by the slump test and the inflection point of the frictional resistance measured by the loop test. Therefore, a simple and convenient slump test should initially be conducted because loop tests are very complex.

4.3. Filling Resistance Data Exhibit Small Fluctuations, Which May Be Caused by the Turbulence Phenomenon Produced by the PG Slurry in the Pipeline. As shown in Figure 15, the filling resistance value of the PG slurry fluctuates between 4 and 6 MPa during the industrial test. The pressure value between gauges 1 and 2 also fluctuates, although the distance between the two pressure gauges is constant, as illustrated in Figure 19. The analysis results indicate that turbulent flow may occur during the PG slurry flowing in the pipeline. Turbulence is related to velocity, pipeline diameter, and physical properties of the filling material.

4.4. Good Fluidity of PGBS with Less Bleeding Water Is the Main Topic in This Study. PG with paste-like backfill (high concentration between 60.87 and 64.73%) is feasible through bleeding water rate, slump, and loop tests of the PGBS. Loop test results demonstrate that the friction resistance coefficient

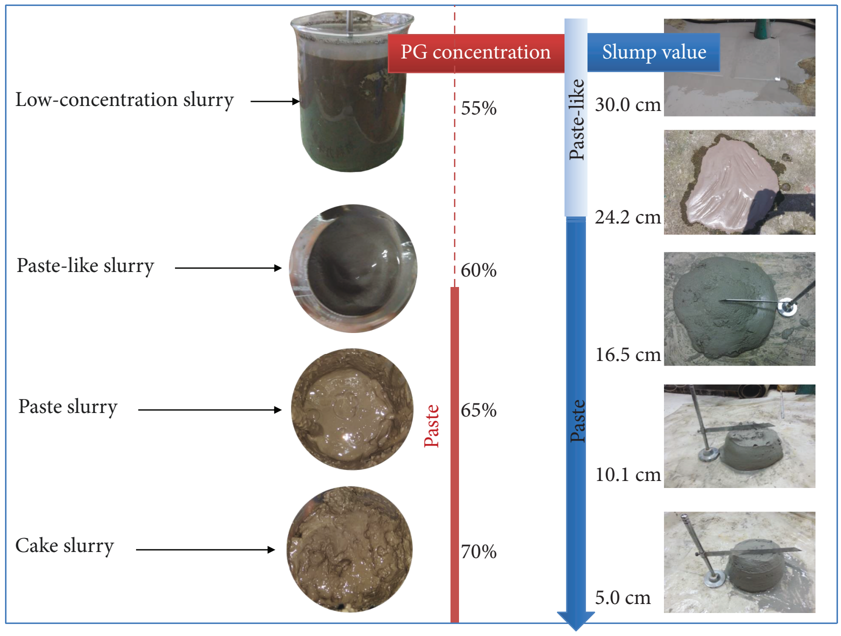

FIGURE 17: Different types of paste and paste-like slurry.

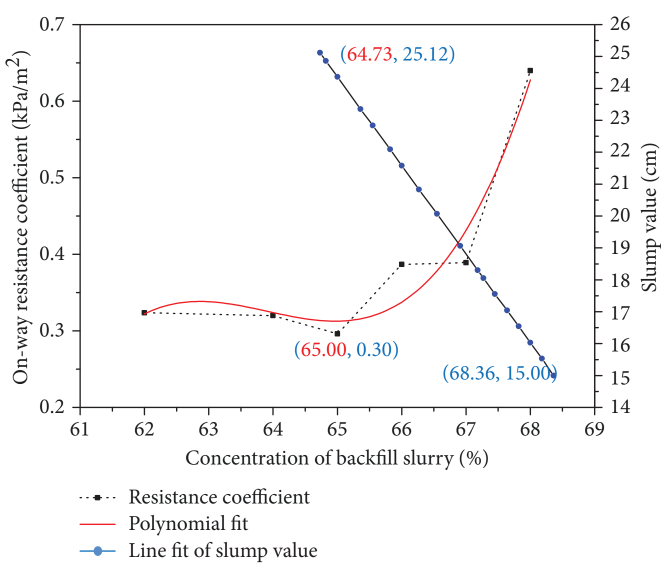

FIGURE 18: On-way resistance coefficient and slump value.

exhibits a sharp inflection point after the PGBS concentration reaches up to 65%. Industrial test results demonstrate that bad fluidity of PGBS will be acquired and the phenomena of pipe plugging and explosion are likely to occur once the PGBS concentration is over 65%.

5. Conclusions

This study discusses the feasibility of high-concentration PGBS transported into the underground goaf. The research results demonstrate that PG slurry with less bleeding water backfill to the goaf is practicable and efficient.

First, according to the BWR test, for the backfill slurry composed of PG and slag (the weight ratio of the PG and slag is 4 : 1), the measured concentration of PG slurry with minimal bleeding water is between 60.87% and 67.61%. Based on the slump test, the maximum concentration of pumpable PG slurry is 68.36%. Combined with the bleeding rate and slump tests, PGBS with a concentration of 61% to 68% is selected for conducting the loop test. The loop test results demonstrate that the resistance coefficient is the lowest when the PGBS concentration is 65%.

Second, according to the industrial pipeline transportation test of the PGBS, PGBS with a concentration of 64%–65% is used as the backfill in the goaf. High-concentration

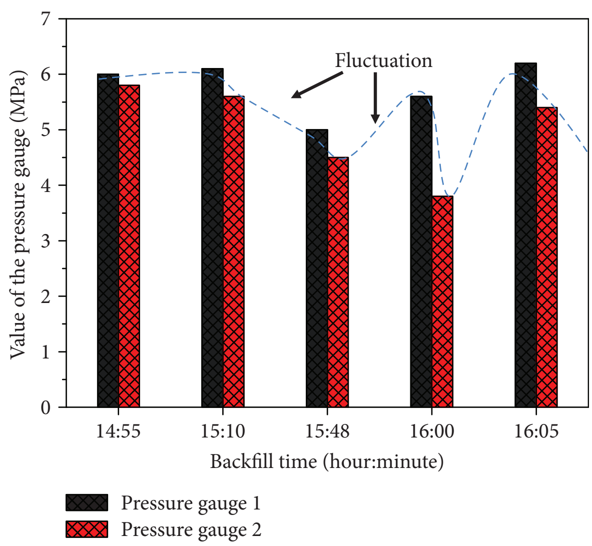

Figure 19: On-way resistance coefficient and slump value.

PGBS backfill to the goaf is feasible because the maximum friction resistance is measured as approximately 6 MPa, which is significantly lower than the capacity (11.5 MPa) of the plunger pump.

Finally, high-concentration PGBS significantly reduces the amount of bleeding water from the underground backfilled stope. It plays an important role in environmental protection and improving the backfill efficiency. Paste-like slurry as high-concentration PGBS is more suitable to backfill PGBS with a high concentration under the present circumstances. The friction resistance is substantially lower than the rated pump pressure. For the backfill slurry composed of PG and slag (the weight ratio of the PG and slag is 4 : 1), the recommended concentration is 61%–65%.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Acknowledgments

The authors wish to acknowledge the financial support received from the State Key Research Development Program of China (Grant no. 2016YFC0600706) and the National Natural Science Foundation of China (Grant No. 41630642). They would also like to thank the Qingcaichong Mine of the Kailin Group Corporation, which supported them on the on-site experiment and data collection.

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