Research Article
Valorization of Water-Based Drill Cuttings through the Bio-Carbonation Approach

Guowang Tang,1 Feng Huang,1 Guihe Wang,1 Zhengyang Song,2 Cangqin Jia,3 Peizhi Yu,1 and Yongshuai Sun4

1School of Engineering and Technology, China University of Geosciences, Beijing 100083, China
2Department of Civil Engineering, School of Civil & Resource Engineering, University of Science & Technology Beijing, Beijing 100083, China
3School of Engineering and Technology, China University of Geosciences and Key Laboratory of Deep Geodrilling Technology, Ministry of Land and Resources, Beijing 100083, China
4College of Water Resources & Civil Engineering, China Agricultural University, Beijing 100083, China

Correspondence should be addressed to Feng Huang; huangfeng@cugb.edu.cn

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This study proposed a novel bio-carbonation method to recycle water-based drill cutting (WDC) to prepare samples, which contains reactive magnesia (MgO) cement (RMC), ground granulated blast furnace slag (GGBS), and fly ash (FA), with the adoption of microbially induced carbonate precipitation (MICP). Through the investigation of some parameters (i.e., GGBS content, FA content, and curing time), the microstructures and strength development of bio-carbonated RMC-based WDC samples were evaluated. The preliminary results revealed that bio-carbonated RMC-based WDC samples outperformed the control group (i.e., without bio-carbonation) in terms of the 28-day strength (i.e., 9.8 MPa versus 4.4 MPa), which can be assigned to formation of the carbonates, that is, hydrated magnesium carbonates (HMCs). Further, in addition to the identification of HMCs, the microstructural analysis also revealed a continuous carbonate network due to the presence of HMCs, which accounts for the strength boost of samples.

1. Introduction

Drill cuttings, contaminated by drilling fluids, are soil-like waste, heterogeneous, and hazardous [1], and drilling fluids can lubricate and cool drilling tools. Meanwhile, they can stabilize the well bore and transport drill cuttings from well bores to the surface. Nevertheless, drilling fluids contain a certain percentage of heavy metals [2], water-soluble salts, and hydrocarbons [3, 4], which contaminate drill cuttings [5]. Due to the possible pollution, drilling cuttings are classified as a special waste by some countries such as US and Indonesia [6]. Hence, drill cuttings should be disposed of efficiently. Currently, according to the previous studies, there are various management options concerning drill cuttings, including the bioremediation [7], the thermal desorption [8], the disposal in landfills [9], the stabilisation/solidification [1], and the reuse in the construction sector [10, 11].

Amid the treatments as aforementioned, disposing drill cutting in landfills is feasible, as it enabled the contaminants to be metabolized, transformed, and assimilated by the microbial populations in soil [12]. At the same time, the thermal desorption is also employed to treat drill cuttings, which is supported by eliminating water, hydrocarbons [4], salinity, and clay, thereby retaining a relatively clean mixture [13, 14]. Meanwhile, in real engineering, treated drill cuttings can be reused as fillers in landfills and aggregates in concrete or brick [15]. It is to be mentioned that valorization of drill cuttings is considered to be efficient and economical. As previously reported by Asadi et al. [16], drill cuttings can be used to replace partial Portland cement (PC) [17–19], which is considered to be a major contributor to the global
greenhouse gas [20, 21]. Compared with PC, reactive magnesia (MgO) cement (RMC) is proposed recently as a sustainable construction material, which is extracted from seawater or brine (wet route) without the CO₂ emission [22]. Another advantage of RMC is that its low production temperature compared with that of PC (e.g., 700°C versus 1450°C) [23] and previous studies have confirmed that carbonated RMC could provide relatively great strength [24], and the reaction is that RMC first hydrates into brucite (Mg(OH)₂), and then it reacts with CO₂ to form various hydrated magnesium carbonates (HMCs) [23]. As a result, nesquehonite (MgCO₃·3H₂O), hydromagnesite (4MgCO₃·Mg(OH)₂·4H₂O), dypingite (4MgCO₃·Mg(OH)₂·5H₂O), and aragonite (MgCO₃·Mg(OH)₂·3H₂O) can be found in the specimens, which are identified as HMCs phases that are yielded from the carbonation process [25, 26].

To improve the mechanical properties of the carbonated RMC-based system, various supplementary cementitious materials including fly ash (FA) and ground granulated blast furnace slag (GGBS) [27] were used in mixtures, demonstrating that carbonated RMC-based mix incorporated with FA has been observed to enhance the agglomeration of carbonate samples, thereby forming more HMCs. Moreover, the formation of carbonate phases improved the morphology and interconnectivity of product phases [28]. In terms of GGBS, when it was involved in the carbonated RMC-based mixtures, the content of other hydration phases in the RMC-based system increased [29], where GGBS was activated with MgO, thus increasing the hydrotalcite content. As a consequence, the pore volume reduced that brought about improved strength. Meanwhile, the strength development was further improved in a CO₂-rich environment [30, 31].

A recent study also revealed that microbially induced carbonate precipitation (MICP) could be used to heal cracks in RMC-based systems [32] due to the formation of HMCs crystals that fill in the cracks via the bio-carbonation process. To be more specific, in the MICP process, the dissolution of RMC was used to replace Ca²⁺; thus, HMCs precipitates could be formed. (The chemical reaction is shown in the following equations:

\[
\text{CO(NH}_3\text{)}_2 + H_2O + \text{bacterialurease} \rightarrow CO_3^{2-} + 2NH_4^+ \quad (1)
\]

\[
\text{CO}_3^{2-} + Mg^{2+} \rightarrow MgCO_3 \downarrow \quad (2)
\]

Compared with RMC-based systems cured in a high concentration and pressure of CO₂, RMC-based MICP methods used urea as a carbonation source to eliminate the use of the high concentration of CO₂, which is more practical [24]. Moreover, most studies have explored the MICP process in a calcium-rich environment, so different from the conventional MICP process, the bio-carbonated RMC-based methods eliminate the use of calcium sources as an addition, which also reduces the cost of this approach.

This study innovatively investigates the microstructures and strength development of bio-carbonated RMC-based WDC samples containing GGBS and FA, and compared with carbonated RMC-based mixtures, a new bio-carbonated technology has been proposed to reuse WDCs. The strength and permeability of all samples were tested, facilitated by the uniaxial loading frame and the triaxial cell apparatus. Meanwhile, Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), and scanning electron microscopy (SEM) were used to investigate the chemical compositions and the microstructures of the samples in details. The findings from this study revealed a new approach to effectively dispose of WDCs through the preparation of the bio-carbonation of RMC-based mixtures.

2. Material and Methods

2.1. Chemicals, Mixture Composition, and Sample Preparation. The Sporosarcina pasteurii (ATCC11859) strain was used for the preparation of bio-carbonated RMC-based WDC samples in this study. The bacteria were used to provide CO₂ to react with RMC to form HMCs in this study. The mother culture consisted of a mixture of soy peptone (5 g/L), casein peptone (15 g/L), urea (20 g/L), sodium chloride (5 g/L), and 1000 mL of deionized water. The pH value of the mother culture was about 7.3 and sterilized at 121°C for 30 minutes. The bacterial solution consisted of the mother culture and Sporosarcina pasteurii. The bacterial cultivation was carried out in 28°C, at a shaking rate of 160 rpm. For 36 hours, the bacterial solution was harvested. The originally harvested bacterial solution revealed the optical density (OD600) of about 2 ± 0.1. Bacterial culture was used immediately after harvest and was not stored. Meanwhile, industrial grade urea was used.

RMC was supplied by the Japan Concorde Company and was analyzed via X-ray fluorescence spectrometry, and its chemical composition is listed in Table 1. WDCs were provided from a coal seam gas production block in Xinxiang, Henan, and their microstructure and chemical composition are revealed via SEM, FTIR, and XRD in Figures 1 and 2. Figure 1 shows flake amorphous WDC samples. Figure 2(a) displays the adsorption bands of samples. −OH group stretching vibrations were explained by peaks at 3,409 and 3,513 cm⁻¹ [36]. The peaks at 871 and 712 cm⁻¹ confirmed the presence of C-O. The peaks at 1,048, 798, 780, 526, and 470 cm⁻¹ were attributed to many Si-O functional groups. The peaks at 2,925, 2,857, 2,514, 1,795, and 1,396 cm⁻¹ indicated the existence of C-O stretch. FTIR proved that CaCO₃ and SiO₂ were the main components of WDC. Figure 2(b) displays the XRD peaks of WDC samples, confirming the existence of CaCO₃ and SiO₂ inside the samples, which is in line with the FITR results. Fly ash (FA) and ground granulated blast furnace slag (GGBS) were obtained from Boheng mineral products trading Co., Ltd. Their chemical compositions are shown in Figure 3. GGBS and FA were composed of glassy phases and mullite (3Al₂O₃·2SiO₂), respectively.

2.2. Bio-Carbonation of RMC-Based WDC Samples. Bio-carbonated RMC-based WDC samples were prepared by stirring the mixtures (i.e., WDCs, FA, GGBS,
and RMC) for 2 mins, then followed by the addition of bacterial solution or water. The solid and liquid mixture was stirred for another 1 min. The freshly prepared mixture was cast into 50 × 50 × 50 mm cubes and finished with a trowel. All samples were demoulded after 24 h and cured under an ambient temperature (30 ± 1.5°C) and relative humidity (20 ± 5%) to improve strength for up to 28 days.

**Table 1: Chemical composition of RMC.**

<table>
<thead>
<tr>
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<th>Chemical composition (%)</th>
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<tbody>
<tr>
<td>MgO</td>
<td>96.142</td>
</tr>
<tr>
<td>CaO</td>
<td>1.791</td>
</tr>
<tr>
<td>SiO₂</td>
<td>0.863</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>0.324</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>0.057</td>
</tr>
<tr>
<td>MnO</td>
<td>0.035</td>
</tr>
<tr>
<td>Others</td>
<td>0.789</td>
</tr>
</tbody>
</table>

RMC content was mixed with 20 wt. % of WDC. Three different FA concentrations (5%, 15%, and 25%) and GGBS concentrations (10%, 20%, and 30%) were studied by the WDC weight. Water content (i.e., water/solid: 30%) referred to the quality of bacterial solution was used in this study. All mixture formulations are listed in Table 2, and 9 samples were cast for each mixture under different curing durations at 7, 14, and 28 days. For control samples, the same quality of
Deionized water was used to replace bacterial culture, whose details are presented in Table 2 (i.e., Sample 0).

2.3. UCS and Permeability Tests. According to ASTM D2166/D2166M [37, 38], the strength of all samples were measured with an uniaxial loading frame. After 7, 14, and 28 days of curing, the compression test was performed at a loading rate of 1 mm/min. The permeability coefficient of all samples was also tested by the water drive with a triaxial cell apparatus [39]. The confining pressure of 3 MPa was applied to the samples, and the pressure difference was from 0 to 6 MPa. All sampling was carried out in triplicate to obtain the standard deviations.

2.4. SEM, FTIR, and XRD Analyses. Fragments extracted from UCS testing were dried for 24 h, and then they were ground into powders. The powder sample passed through a sieve size of 75 mm before FTIR, XRD, and SEM analyses. The XRD measurement was performed with an automatic XRD instrument (Brooke, Germany). The scanning speed was 0.15 s step⁻¹, and the range of 2θ is 10°~80°. The FTIR data were collected with an infrared spectrometer (FTIR8400, SHIMADZU, Japan) with a wavenumber range of 4000–400 cm⁻¹, and the scanning times and resolution is 32 4 cm⁻¹, respectively. The morphologies of the RMC-based WDC samples and bio-carbonated RMC-based WDC samples were investigated using a scanning electron microscopy (SEM, Zeiss Supra 55, Germany) with an acceleration voltage of 25 kV.

3. Results and Discussion

3.1. Strength and Permeability. Figure 4 presents the UCS of all samples after curing 7, 14, and 28 day. Compared with sample 0 and sample 8, the strength of sample varied from 4.4 to 9.8 MPa after 28 days of curing, which proved that bio-carbonated RMC-based samples was effective to treat WDC. It was worth mentioning that the maximum strength of samples reached 6.9 MPa after 7 days of curing, indicating a faster strength gain at an early age. Meanwhile, previous studies reported that urea was completely hydrolyzed to form carbonate ions by MICP in 1 day, and through the reaction between carbonate ions and brucite (Mg(OH)₂), the formed HMCs enhanced strength of bio-carbonated RMC-based samples at early age [40]. In addition, it was observed that the strength of bio-carbonated RMC-based samples increased over time from 7 to 28 days, thereby highlighting the continuation of the hydration and carbonation process [41]. This was mainly because the available
unreacted RMC in the samples continuously hydrated and transformed into Mg(OH)$_2$ during the curing process, thereby reacting with carbonate ions to form more HMCs that contributes to the further strength development after 28 day of curing.

Figure 4 also shows the effects of GGBS and FA concentrations on the strength of bio-carbonated RMC-based WDC samples. Compared with samples 2, 5, and 8 after 28 days of curing, the strength of bio-carbonated samples increased from 5.5 to 9.5 MPa when GGBS content increased from 10 to 30%. This was because GGBS has been activated by RMC, and the content of hydrotalcite increased in samples, thereby reducing the pore volume and enhancing the strength development of bio-carbonated samples [42]. The strength of samples was further improved via the bio-carbonated process due to the formation of different carbonate phases [30, 31], explaining their greater strength. When GGBS was 60 g and 120 g, the lower strength of bio-carbonated samples could be observed with an increase in FA concentrations. Nevertheless, 60 grams of FA achieved the maximum strength when GGBS concentration was 180 grams in the samples. This was because the addition of FA was considered to enhance the pozzolanic reaction and yield more hydrate phases (e.g., M-S-H and hydrotalcite), thus reducing the pore volume and elevating the strength of samples. However, the above results also confirmed that the mix design was important. In terms of the permeability, the initial permeability of pure WDC column was $1.1 \pm 2 \times 10^{-2}$ m/s without the confining pressure, while the permeability test showed that the permeability coefficient of all bio-carbonated RMC-based WDC samples and sample 0 was 0 cm/s, indicating the large bio-carbonation extent of samples, where the pore space was reduced with the formation of HMCs.

3.2. FTIR. Figure 5 depicts the FTIR spectra of bio-carbonated RMC-based WDC samples within various GGBS and FA concentrations after 28 days of curing. The molecular groups of the samples within various GGBS and FA concentrations were found to be similar. In all samples, the peak at 3,697 cm$^{-1}$ indicated the presence of the O-H functional groups, which was formed by the hydration of RMC that led to the formation of Mg(OH)$_2$ [43]. The peak of 565 cm$^{-1}$ was assigned to the presence of MgO [44]. The above results showed the presence of unreacted MgO and Mg(OH)$_2$, thereby confirming the incomplete hydration and carbonation. The peaks of 1,463 cm$^{-1}$ with an asymmetric stretch were attributed to the presence of C-O stretch, which further proved the formation of HMCs [44, 45]. The peaks of 870 and 712 cm$^{-1}$ with a bending vibration also proved the existence of C-O [46]. Furthermore, a higher carbonation band intensity was observed with an increase in GGBS concentrations, which indicated the increased formation of carbonate phases [27]. Nevertheless, the carbonation band intensity decreased when FA concentrations increased. This proved that the strength development of samples was associated with the carbonate phase contents. The peaks at 1,058 cm$^{-1}$ were attributed to the existence of Al-O bending, which was associated with the hydrotalcite formed with FA and GGBS [27]. The characteristic peak at 1,720 and 3,122 cm$^{-1}$ can be assigned to the existence of O-H bending, which was associated with the M-S-H formed within FA [46, 47].

3.3. XRD. Figure 6 displays the XRD patterns of bio-carbonated RMC-based WDC samples within various GGBS and FA concentrations after 28 days of curing. The peak positions of all samples within various GGBS and FA concentrations were found to be similar. Among some peaks of the samples, the diffraction peaks of MgO (at 43$^\circ$ and 62$^\circ$ 2θ) and brucite (at 18.6$^\circ$, 38$^\circ$, 50.8$^\circ$, and 68.2$^\circ$2θ) were observed, which were consistent with the FTIR outcomes. Meanwhile, unreacted MgO and brucite confirmed incomplete hydration and carbonation over time, indicating the prospect of bio-carbonated approach to further facilitate the strength development if the complete hydration and carbonation in the samples were to be achieved.

Figure 4: UCS of bio-carbonated RMC-based WDC samples.
The broad peaks (at 10–25°, 32–37°, and 56–65° 2θ) were attributed to the existence of amorphous M-S-H [48], which were observed in all samples since hydration occurred in the presence of water. Meanwhile, the peaks of hydrotalcite were also observed at 12.1°, 23°, and 61° 2θ. Furthermore, the main peak of hydromagnesite was observed at around 30.6° 2θ. The existence of M-S-H, hydrotalcite, and hydromagnesite was an indication of the occurrence of hydration and bio-carbonation simultaneously over time. These observations were in agreement with the strength development, highlighting the effectiveness of strength improvement via bio-carbonation.

3.4. SEM. Figure 7 shows the SEM images of RMC-based WDC samples. WDC particles were surrounded with hydration products, as shown in Figure 7(a). This was because the hydrated phases were initially formed onto the WDC particle surface and acted as the nucleation sites of hydrolysis process; thus, the WDC particles were surrounded with the reduced porosity and the boost of strength development, which was consistent with previous studies [27]. Figures 7(c) and 7(d) present morphologies of packed M-S-H structures and hydrotalcite (i.e., hydration phases), which is as a result of the hydration of MgO and SiO₂ [27]. Moreover, the addition of GGBS into the bio-carbonated RMC-based samples elevated the solubility of glassy phases, consequently achieving the formation of hydrotalcite in the samples.

Figure 8 exhibits SEM images of bio-carbonated RMC-based WDC samples. Unlike the RMC-based WDC samples, the microstructural development of bio-carbonated RMC-based WDC samples was controlled with the formation of carbonate phases (e.g., hydromagnesite). Figures 8(c) and 8(d) show the widespread presence of rosette-like crystals, which are identified as the hydromagnesite [49]. Meanwhile, compared with morphologies of RMC-based WDC samples, bio-carbonated RMC-based WDC samples presented denser inner structures (Figures 7(a) and 7(b)) owing to the formation of carbonate phases, which reduced the pore volume and formed a continuous carbonate network, facilitating the strength development. Overall, the microstructural
Figure 7: SEM images of the RMC-based WDC samples after 28 days of curing.

Figure 8: SEM images of bio-carbonated RMC-based WDC samples after 28 days of curing.
development was in agreement with the FTIR and XRD results, and they can account for the strength development of the samples.

4. Conclusion

The study aimed to investigate the use of MICP to valorize RMC-based WDC, where the relationship between the strength and microstructural development of the samples containing GGBS and FA was studied. While RMC-based WDC samples obtained the strength with the formation of M-S-H and hydrotalcite in the existence of GGBS and FA, bio-carbonated RMC-based WDC samples gained higher strength owing to the formation of an additional continuous carbonate (e.g., HMCs) network.

The strength of bio-carbonated samples increased from 5.5 to 9.8 MPa in the GGBS content from 10 to 30%, which indicated GGBS was a suitable additive in terms of the strength development. When the concentration of FA is 60 g, this was considered to be the optimal dosage. In terms of the optimal formulation (sample 8), the strength of sample revealed was up to 9.8 MPa after 28 days of curing. Therefore, the mix design of bio-carbonated RMC-based samples should be further optimized to enhance the strength development and microstructure refinement.

Overall, this study concluded that the optimum mix composition of bio-carbonated RMC-based WDC samples was 20% of RMC, 30% of GGBS, and 10% of FA at a w/s of 30%, highlighting the effectiveness of a new method for the treatment of valorization of WDC, that is, through the preparation of bio-carbonated RMC-based mixtures.

Data Availability

Some or all data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

Disclosure

Yongshuai Sun is the co-first author.

Conflicts of Interest

This is a submission from one of the editorial board of the special issue (zhengyang song). The authors declare that they have no conflicts of interest to this work. The authors declare that they do not have any commercial or associative interest that represents conflicts of interest in connection with the work submitted.

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