Iranian Journal of Mining Engineering (IJME)
Vol 17, No 54, 2022, pp 17-20

DOI: 10.22034/IJME.2021.141646.1841
DOR: 20.1001.1.17357616.1401.17.54.5.1

Iranian Society of Mining Engineering
(IRSME)

Investigation of Microbial Precipitation of Calcium Carbonate in Remediation of Building Stone

Aref Fayyazi[1], Ramin Doostmohammadi[2*]

1. Master of science, University of Zanjan, Zanjan, Iran, aref.fayyazi@znu.ac.ir
2. Associate Professor, University of Zanjan, Zanjan, Iran, ramin.doostmohammadi@znu.ac.ir

Recived: 2020/09/28 - Accepted: 2021/08/14

Abstract

Preservation and remediation of historic buildings and building Stones have always been significant challenges for mining engineers. One of the useful methods to prevent stone decade is filling the pores using microbial induced calcium precipitation (as known bio-grouting). In this paper, the effects of different conditions on the optimal use of bio-grouting are investigated by performing laboratory tests. Unprocessed travertine samples with the same mineralogical and lithological conditions were selected. Experimental pattern with four variables (concentrations of calcium chloride and urea in cemention solution, pH of washing solution and ambient temperature) were designed to determine the wave velocity ratio (as a remediation sign) at samples. The input variables were selected in 5 levels using response surface designing method and 31 laboratory tests were performed. The results indicate that the use of equal concentrations of calcium chloride and urea (about 1 M) in the cementation solution is more effective for microbial precipitation. Adjusting the pH of the washing solution about 7 and the ambient temperature about 15 Celsius degrees will lead to optimal amount of calcium carbonate precipitatation, resulting in better rock remediation. Increasing the pH from 7 to 11, reduces the wave velocity ratio by 35% and decreasing the pH from 7 to 3 reduces the wave velocity ratio by 40%. Also, increasing and decreasing the ambient temperature from 15 to 30 and 15 to 0 Centigrade degrees, respectively, reduces the wave velocity ratio by 15%. The above results show that very acidic or alkaline environments and very cold or warm temperatures are not suitable for microbial precipitation operation and the effect of acidic rains on the degradation of remediating precipitations is more than alkaline rains.

Keywords

Rock Remediation, Biogrouting, Laboratory tests, Respone Surface Method.

[*] Corresponding Author

1- Introduction

Microbial life began about 3.5 billion years ago, and the formation of calcium carbonate in the Earth's crust can be largely attributed to the action of some microbes. In the new technique of calcium carbonate microbial precipitation (MICP), this oldest biological process, microbial activity, has been used 1. The new MICP technique, also known as bio-grouting, is one of the methods to overcome the limitations of chemical slurries or cement. This low-viscosity grouting with non-chemical material can be injected into the deep depth of rock mass and inside micro-cracks 2. In general, bio-grouting requires bacteria, urea, calcium chloride and nutrients, which is also referred to as enzymatic urea due to the use of bacterial enzymes 3. In the MICP process, urease bacteria (UPBs) generally produce the urease enzyme. Urease bacteria can produce enzymes in the soil within a wide range of changes in pH, temperature and humidity. The process of bio-grouting formation through microbial decomposition of urea is as follows [1, 4].

A. Urea (CO $(NH_2)_2$) is rapidly hydrolyzed to produce ammonia (NH_3) and carbamic acid (NH_2COOH) during a urease activity in the presence of water-soluble bacterial enzymes. The product of this reaction is the spontaneous hydrolysis of carbamic acid to ammonia and carbonic acid (H2CO3) (Equations 1 and 2).

$$(1)\quad CO(NH_2)_2 + H_2O \rightarrow NH_3 + NH_2COOH$$
$$(2)\quad NH_2COOH + H_2O \rightarrow NH_3 + H_2CO_3$$

B. Ammonia forms ammonium ions (NH_4^+) and hydroxides (OH^-), and carbonic acid forms bicarbonate (HCO_3^-) and hydrogen (H^+) ions (Equations 3 and 4).

$$(3)\quad 2NH_3 + 2H_2O \rightarrow 2NH_4^+ + 2OH^-$$
$$(4)\quad H_2CO_3 \rightarrow HCO_3^- + H^+$$

C. The hydroxide ion raises the pH and changes the bicarbonate balance. In order to restore equilibrium in the reaction, the transfer of hydrogen ions from bicarbonate ions to hydroxide ions takes place, which results in the formation of carbonate ions (CO_3^{2-}) (Equation 5).

$$(5)\quad HCO_3^- + H^+ + 2NH_4^+ + 2OH^- \leftrightarrow CO_3^{-2} + 2NH_4^+ + 2H_2O$$

D. Finally, in the presence of calcium ions, carbonate ions precipitate as calcium carbonate (CaCO3) crystals (Equation 6).

$$(6)\quad Ca^{+2} + CO_3^{-2} \leftrightarrow CaCO_3$$

2- Methods

The materials used in the experiments of the present study are samples of unprocessed travertine, *Sporosarcina-Pasteurii* bacteria with a optical density of about 1, urea and calcium chloride. Also, to design the test model, the response surface method in "Minitab 19" software has been used.

If the microbial precipitation of calcium carbonate is effective in remediation and filling the voids of rocks, it is expected that the ratio of secondary to primary wave velocities in them will increase compared to pre-remediation conditions.

The Response Surface Methodology (RSM) is a set of statistical techniques and applied mathematics for constructing experimental models. The purpose in such schemes is to optimize the response (output variable) which is affected by several independent variables (input variables) 5. The main purpose of RSM is to design experiments, commonly known as DOE. Optimal selection of the test design can have a great impact on the accuracy of estimating the surface of parameters, reducing the number of tests and thus reducing project costs.

3- Findings and Argument

"Backward Elimination" model with 95% surface of reliability in "Minitab 19" software was used to construct the best model and finally a regression approximation model based on the results of laboratory tests performed in determining the ultrasonic wave velocity ratio obtained by Equation 7.

$$-0.789 - 0.003\,C + 0.224\,U + 0.6531\,pH + 0.03727\,T - 0.560\,C\times C - 0.706\,U\times U - 0.04610\,pH\times pH - 0.001278\,T\times T + 1.213\,C\times U \quad (7)$$

Where:

C: Molarity of Calcium chloride
U: Molarity of Urea
pH: pH of the Washing Solution
T: Ambient Temperature (degrees Celsius)
v: The ratio of the P-wave velocity after to before the remediation operation.

According to Figure 1, which shows the Pareto diagram in determining the effect of different parameters on the target variable (wave velocity ratio); Significant correlation between squared pH of the washing solution (pH × pH), squared ambient temperature (T × T), ambient temperature (T), multiplication of concentration of calcium chloride and urea (C × U),. squared urea concentration (U × U), squared calcium chloride concentration (C × C) and urea concentration (U) respectively with a wave velocity ratio (V).

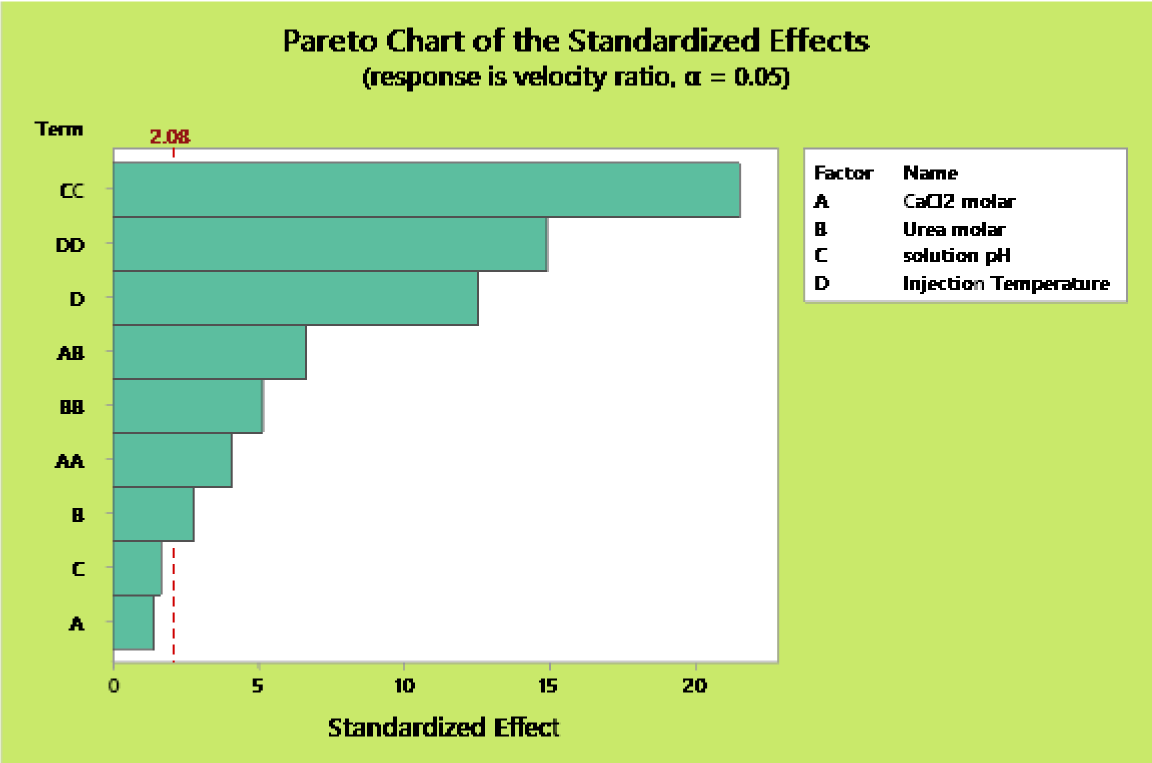

Figure 1. Pareto diagram in determining the effect of different parameters on the wave velocity ratio (A = calcium chloride concentration, B = urea concentration, C = pH of the washing solution and D = ambient temperature).

Identifying the optimal surface of input variables to achieve the desired surface of responses is a requirement of all statistical researches. Therefore, in order to optimize the model, the optimal surface of input variables (concentrations of calcium chloride and urea in the cementing solution, pH of the washing solution and ambient temperature) was performed by RSM method in experimental pattern design software (Minitab 19)

4- Conclusions

In this paper, the positive performance of the proposed method was confirmed by measuring the P-wave velocity before and after remediation of the stones with *Sporosarcina-Pasteurii* at a optical density close to 1. Using the response surface methodology, experiments were designed and performed to investigate the effect of the values of cementing solution components and environmental conditions. Studies have shown that the concentration of calcium chloride and urea, the ambient temperature of the remediation operation, as well as the acidity and alkalinity of the washing solutions are effective in the final result of the precipitation operation and the result of this effect was presented by extracting a statistical formula with a correlation coefficient of more than 96%. Then, the interaction of the effective variables in the precipitation operation was optimized and led to the conclusion that by performing the microbial precipitation operation at a temperature of 15 ° C and using a cementing solution with a concentration equal to 1.1 M for calcium. Chloride and urea can achieve the best remediation results. This result indicates that performing remediation of rocks operations in spring and autumn, when the temperature is mild, will bring the best results. Finally, the effect of rain on the stability of sediments formed in voids of rocks was measured and it was found that if the rainwater is acidic or alkaline, part of the sediments formed in voids of rocks are washed away. Examining the results of laboratory tests and the proposed equation in determining the wave velocity ratio, it was found that the effect of acidic rain on reducing the effect of remediation is more than alkaline rain. Therefore, the better performance of the method described will be higher in areas where acidic and alkaline rains are less likely to fall (non-industrial areas with lower factory densities).

References

1. Al-Salloum, Y., Hadi, S., Abbas, H., Almusallam, T., Moslem, M.A.; 2017; "*Bio-induction and bioremediation of cementitious composites using microbial mineral precipitation – A review*"، https://doi.org/10.1016/j.conbuildmat.2017.07.203 ،Engineering Geology, pp. 23-30.

2. Wu, C., Chu, J., Wu, S., Guo, W.; 2018; "*Quantifying the Permeability Reduction of Biogrouted Rock Fracture*" ،https://doi.org/10.1007/978-981-10-1445-1 ،Springer Singapore, pp. 947–954.

3. Jongvivatsakul, P., Janprasit, K., Nuaklong, P., Pungrasmi, W.; 2019; "*Investigation of the crack healing performance in mortar using microbially induced calcium carbonate precipitation (MICP) method*"،https://doi.org/10.1016/j.conbuildmat.2019.04.035 ،Construction and Building Materials, pp. 737-744.

4. Morales, L., Garzón, E., Romero, E., Sánchez-Soto, P.J.; 2019; "*Microbiological induced carbonate (CaCO3) precipitation using clay*" ،https://doi.org/10.1016/j.clay.2019.03.018 ، Applied Clay Science, pp. 1–24.

5. Myers, R. H., Montgomery, D. C., Geoffrey, V. G., Borror C. M. & Kowalski S. M.; 2004; "*Response Surface Methodology: A Retrospective and Literature Survey*" ، https://doi.org/10.1080/00224065.2004.11980252 ،Journal of Quality Technology, pp. 53-77.