Third International Conference on Sustainable Construction Materials and Technologies http://www.claisse.info/Proceedings.htm

Bacterial Calcification for Enhancing Performance of Low Embodied Energy Soil-Cement Bricks

Abhjit Mukherjee^{1*}, Navdeep Kaur Dhami², B.V.V. Reddy³, and M. Sudhakara Reddy²

¹Indian Institute of Technology Gandhinagar, VGEC Complex, Ahmedabad 382424, India

²Department of Biotechnology, Thapar University, Patiala 147004, Punjab, India

³Department of Civil Engineering, Indian Institute of Science, Bangalore 560062, India

^{*}*Prof. A. Mukherjee*, ¹*Indian Institute of Technology Gandhinagar, VGEC Complex, Ahmedabad 382424, India*

Tel: +918758095500, *Fax:* +917923972324. *Email: abhijit.mukherjee@iitgn.ac.in, dhami_navdeep@yahoo.co.in, venkat@civil.iisc.erent.in, msreddy@thapar.edu*

ABSTRACT

Soil–cement bricks are vastly more energy efficient than fired clay bricks. Although they have adequate strength, they absorb high levels of moisture and in humid conditions they become soft and non-uniform expansion leads to excessive deformation and cracking. In this research a barrier layer on their surface that impedes moisture ingress is developed by depositing calcite using bacteria. Soil-cement bricks (230 mm x 110 mm x 60-75 mm) were prepared by mixing bacteria (*Bacillus megaterium*) and cured by spraying a nutrient media for 28 days. The specimens were tested for water absorption, porosity and compressive strength and compared with control specimens. Results showed that the rate of water absorption and porosity were significantly reduced and compressive strength was enhanced in bacteria treated bricks. The results suggest that the barrier layer created by bacterial activity greatly alleviates the weaknesses of energy efficient soil-cement bricks enabling their large scale use.

Key words: microbial calcite, alternative building materials, bacteria, porosity, compressive strength

INTRODUCTION

The emerging economies have an imperative of rapidly building their infrastructure while controlling the energy consumption and emission. Thus, they must select building materials that satisfy the performance needs of the user as well as the development needs of the society, without causing any adverse impact on environment (Reddy and Jagdish 2003). Present manufacturing processes of building materials such as steel, cement and brick emit huge quantities of greenhouse gases like CO₂ to the atmosphere. Studies related to energy consumption for production and processing of different building materials and the CO₂ emissions and the implications on environment have been reported from New Zealand (Buchanan and Honey 1994), Japan (Suzki et al. 1995, Oka et al. 1993) and India (Debnath et al. 1995; Reddy and Jagdish 2003). Cement, steel and bricks are the largest and bulk consumption items in construction industry and minimizing the embodied energy of these materials can result in considerable energy savings as well as reduction of CO_2 emission. In the former SCMT Conference the present authors (Achal et al. 2011) reported a microbial concrete that improves durability of conventional concrete and thus makes it sustainable. However, the technology has potential for other forms of construction such as roads, masonry walls and dams. In this meeting we report out work on a sustainable brick.

Fired clay bricks have around 3MJ/Kg of embodied energy, far higher than concrete or stone blocks. Generally, burnt clay brick (230 mm x 110 mm x 60-75 mm) needs either 0.20 kg coal or 0.25-0.30 kg of firewood for the burning process which translates into a thermal energy of 3.75 - 4.75 MJ (average 4.25 MJ) per brick. Reddy and Jagdish (2003) reported a soil-cement brick that consumes only one-fourth of the energy of burnt clay brick. Manufactured by compacting a mixture of locally available soil, sand and a small quantity of cement, these bricks have a very low embodied energy and CO₂ emission. Moreover, it uses local materials and inexpensive equipment. These bricks have a cement content of about 6-8% with energy content of 2.75-3.75 MJ per brick and have 45% lesser embodied energy as compared with burnt clay brick masonry (Reddy and Jagdish 2003). Though these bricks have adequate strength and durability, they absorb high levels of moisture. In humid conditions, they become very soft and non-uniform expansion leads to excessive deformation and cracking. A barrier layer on their surface that impedes moisture ingress would significantly improve their usefulness.

Microbially induced calcium carbonate precipitation (MICCP) is the process by which microorganisms deposit carbonates as part of their basic metabolic activities (Stocks Fischer et al. 1999; Achal et al. 2009). Precipitation of calcium carbonate crystals occurs by heterogeneous nucleation on the bacterial cell wall. Once supersaturation is achieved and these crystals precipitate inside the pore spaces. The bacteria not only initiate calcite precipitation, but also serve as nucleation sites for calcite crystals in association with other factors such as Ca^{2+} ions, dissolved inorganic carbon, pH, and temperature in the medium (Stocks Fischer et al. 1999; Hammes and Verstraete 2002). These carbonates/ calcite crystals act as cloggers in the building materials by filling the voids and thereby reducing permeability (Dhami et al. 2012).

The authors have carried out extensive studies on carbonate crystal precipitation in building materials. In cement mortar cubes bacterial calcite led to enhanced compressive strength along with reduction of water and chloride ion permeability (Achal et al. 2010; 2011). It was also effective in reducing corrosion in reinforced concrete (Achal et al. 2012). All these observations clearly indicated that bacterial calcite precipitation proceeds as long as nutrients for microbial growth and ingredients for calcite precipitation are available and seals the

pores. Moreover, it supports the notion that this technology can lead to self-healing materials.

The objective of this paper is to evaluate the potential of bacterial carbonates/ calcite crystals as biogenic surface treatment to remediate excessive absorption and anomalous expansion of low energy, low CO₂ emitting, green building materials i. e. soil cement bricks. The effect of bio-cementation by bacterial calcite on varying densities soil cement cylinders has also been checked in order to find out an optimal combination.

Precipitation of calcium carbonate by bacteria

Bacillus megaterium isolated from calcareous soil was used in this study. The culture was chosen because of its efficacy to produce urease and calcite crystals in the experimental system. The strain was cultivated in sterile nutrient broth medium along with 2% urea and 25 mM CaCl₂ (NBU media) The final pH of the medium was 8.0. The culture was incubated at 37 °C in rotating shaker at 120 rpm for 96 hrs. The crystals precipitated at the bottom of each flask were collected through filtration, washed and dried at 37°C for 48 hrs for analysis of morphology and mineralogical composition. X- ray Diffraction was used to identify the types of carbonate polymorph that were precipitated. Bacillus megaterium was able to produce maximum urease (650 U/ml) on fourth day and precipitated carbonate crystals in presence of urea and CaCl₂. Carbonate crystals precipitated by *B. megaterium* increased significantly with increase in time and about 180 mg crystals/100 ml medium was observed after 3 weeks at 37 °C. No considerable amount of crystals precipitated in control. Carbonate crystals precipitated were identified as calcite and vaterite through X- ray diffraction. The precipitation of various types of carbonate crystals by different bacteria is related to the optimal growth conditions required (Del Moral et al. 1987; Rivadeneyra et al. 1998). Zamarreno et al (2009) reported that unstable carbonate polymorphs like vaterite and aragonite are precipitated under unfavourable growth conditions and stable polymorphs such as calcite and dolomite are produced under optimal growth conditions.

Microbial plugging of soil-cement bricks

Soil-cement bricks of size 230 mm x 110 mm x 60-75 mm were prepared as described by Reddy and Gupta (2005). Manually operated brick making machine was employed. The bacterial culture with density 10^8 cells/ml was mixed while making the bricks. The brick density was set at 1.8 Kg/m³. The bricks were cured by spraying NBU media for 28 days. After 28 days of curing, the bricks were kept inside the laboratory for drying for more than 30 days before using them for experiments. Control bricks were made without bacteria.

Water absorption test

Experiments were performed to determine the water absorption and rate of moisture absorption of the bricks with the time of soaking in water. The bricks were dried in an oven at 60°C until they attained constant weight. The weights of dried bricks were recorded. For determination of water absorption, the bricks were immersed in fresh water for 24 hrs, removed, wiped with damp cloth and final weight was measured as per IS: 3495. To calculate the rate of moisture absorption, the bricks were immersed in water for different intervals of time (0, 0.5, 1, 1.5, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25, 30, 35, 40, 50, 60, 70, 80, 90, 100, 120, 140, 160, 180, 210, 240, 600, 720, 960, 1220 and 1440 mins) and the weight of wet brick was measured. The percentage saturation of the brick was calculated with respect to dry weight.

Bricks when soaked in water absorbed water rapidly during first few minutes as shown in figure 1a. The rate of water absorption was significantly reduced in bacteria treated bricks. The saturated water content of bacteria treated bricks was found out to be 6.6 % while in case of untreated bricks, it was found to be 9.9% which showed 33.6 % reduction (Fig. 1b). The reduction in the extent of water absorption may be attributed to the fact that due to bacterial calcification, carbonate crystals were deposited and the pores have been reduced on the surface of bricks leading to protective film coating which prevented ingress of water. Accumulation of bacterial biomass, insoluble bacterial slime, and poorly soluble biogenic gas bubbles (produced by bacterial cells) on the other hand, may increase permeability (Vandevivere and Baveye. 1992; and Bonala and Reddi. 1998). As the large amount of pores is plugged due to precipitation, the pore connectivity and water accessible porosity is decreased (De Muynck et al. 2010). Because microorganisms are mainly retained in the pores that are 2 to 5 times larger than the cells (Samonin and Elikova. 2004), precipitation, therefore, will initially occur in pores with diameter ranging from 2- 20 µm. However, as a result of the alkalinisation of the environment, heterogenous precipitation can also occur on the surface of the pores. This could result in the plugging of smaller pores (De Muynck et al. 2009). Achal et al. (2010) and De Muynck et al. (2008a,b) observed significant decrease in water absorption rate in cement mortar cubes upon treatment with bacteria as compared to control.

Wet compressive strength

The compressive strength of bacterial treated (B-BI) and control soil-cement bricks (C-BI) was obtained by testing them in compression testing machine. The frogs were filled with rich cement mortar. Bricks were soaked in water for 48 hours prior to testing. Saturated bricks were mounted in compression testing machine such that there is thin plywood sheet on both faces of the brick between thick rigid platens. The ultimate crushing load was noted down to calculate wet compressive strength of the bricks.

The compressive strength of bacteria treated soil- cement bricks was found out to be 6.69 MPa while that of control bricks was 6.02 MPa which showed 10% improvement (Fig. 1c). The improvement in strength is also due to biomineralized calcium carbonate on bacterial cell surface and within soil cementitious material. The plugging of pores in between the bricks leads to enhanced strength. Measurable increase in compressive strength by application of *S. pasteurii* has also been reported in cement mortar cubes (Ramachandran et al. 2001; Achal et al. 2011), limestone (Le Metayer-Levrel et al. 1999) and fly ash bricks (Dhami et al. 2012).

Linear Expansion on saturation

Experiments were performed to determine the linear expansion on saturation of soil-cement bricks. Length comparator fabricated in the lab was used. Initial length of the oven dried bricks was determined and then soaked in water for 24 h. Length of saturated bricks was determined. The difference in the initial and final readings gave linear expansion of the brick, which was expressed as percentage of actual length in dry state to get linear expansion value.Linear expansion on saturation of <0.10% generally leads to stable long term performance (Reddy 2002). Linear expansion in case of bacterial bricks was found out to be 0.05% while in case of untreated bricks it came out to be 0.09% (Fig. 1d). This also proved the effect of bacterial calcite on performance enhancement of soil-cement bricks.



Fig. 1. Influence of microbial calcite deposition on (a) water absorption rate, (b) saturated water content (c) compressive strength and (d) linear expansion of soil cement bricks treated with *Bacillus megaterium*. Error bars show standard deviation

Porosimetry analysis

Changes in porosity and pore size determination following clacification were studied using mercury intrusion porosimetry (MIP) (with a Micromeritics Autopore 5510 device). Bacterial treated and control brick specimens (1 cm³ geometric volume) were dried over night in an oven at 80°C prior to MIP analysis. The samples were degassed under vacuum under pressure of 50 μ m Hg. Mercury surface tension value of 485 dynes/cm and contact angle of 130° was used, respectively. Mean pore size in the range of 0.006 μ m to 338 μ m was determined. Porosity (% total porosity) was then determined as weight normalized volume of mercury intruded in the sample.

Parameter	Control Specimen	Bacterial treated Specimen
Total Intrusion Volume (mL/g)	0.15 <u>+</u> 0.00	0.09 <u>+</u> 0.00
Total Pore Area (m ² /g)	10.63 <u>+</u> 0.55	6.20 <u>+</u> 0.29
Median Pore Diameter (µm Volume)	0.37 <u>+</u> 0.01	1.23 <u>+</u> 0.06
Median Pore Diameter (µm Area)	0.017 <u>+</u> 0.00	0.01 <u>+</u> 0.00
Average Pore Diameter (µm 4V/A)	0.058 ± 0.00	0.05 ± 0.00
Bulk Density at 0.60 psia (g/mL)	1.62 <u>+</u> 0.06	1.89 <u>+</u> 0.07
Apparent (skeletal) Density (g/mL)	2.16 <u>+</u> 0.11	2.28 <u>+</u> 0.13
Porosity (%)	25.30 <u>+</u> 1.72	17.50 <u>+</u> 0.92

 Table 1. Mercury intrusion porosimetry analysis of control and bacterial treated soilcement bricks

Mercury intrusion porosimetry data (Table 1) of bacterial and control specimens also reflected the effect of MICP on reduction of total porosity. The total porosity of control specimens was found to be 25.3% while upon calcification; it was estimated to be 17.4% which showed 31% reduction. The porosity of the samples was calculated using cumulative intrusion volume and relevant mass measurements. The reduction in porosity is attributed to filling of the voids and clogging of pores due to calcite binders. Calcium carbonate forms an impervious layer on the surface. Whiffin et al. (2007) observed 90% decrease in porosity of sand column upon bacterial calcite formation. Both porosity and permeability are related to each other. If porosity is high and pores are well connected, permeability is high. If porosity is low or pores are badly connected, permeability is low. In this case, precipitation of calcium carbonate in the pore spaces on the surface of specimens resulted in reduction of pore space volume, lowering of permeability, porosity and enhancement of performance.

SEM, EDX and XRD analysis

The layer of white deposits on the upper portion of bacteria treated bricks and upper layer of control bricks was analyzed with SEM, EDX and XRD. Samples were completely dried at room temperature, then examined at accelerating voltages ranging from 30-35 kV by a SEM (Zeiss EV050), which was equipped with energy dispersive X-ray analyzer (Bruker AXS, QuanTax 200) for elemental analysis. Samples were gold coated with a sputter coating Emitech K575 prior to examination. XRD spectra were obtained using an X'Pert PRO diffractometer with a Cu anode (40kV and 30mA) and scanning from 3° to 60° 20. Each sample was crushed and ground using a mortar pestle before mounting a glass fiber filter using a tubular aerosol suspension chamber (TASC). The components of the sample were identified by comparing them with standards established by the International Centre for Diffraction Data.



Fig. 2a) Scanning Electron Micrograph of calcite crystals on the surface of soilcement brick b) Rod shaped impressions of bacterial cells (BC) on calcite crystals c) Energy dispersive X ray analysis d) X ray diffraction pattern of crystalline layer present on the surface of bacterial sand-cement brick SEM investigations revealed interesting phenomenon. The involvement of bacteria in calcite biomineralization was very evident as bacterial cells in close contact with calcite crystals were visible. Rod shaped impressions of bacterial cells within calcite crystals proved that they had been occupied by bacteria at some stage of crystallization or the cells had completely colonized by the crystals (Fig. 2a, 2b). The presence of calcite associated with bacteria proves that bacteria served as nucleation sites during the mineralization process.

EDX analysis further clarified high amount of calcium in case of bacterial treated samples (Fig. 2c) while in case of control untreated samples, there was very low calcium content (Fig. 2d). XRD analysis was done to reveal the form of mineral constituents formed in bacterial bricks which showed that majority of carbonate deposits were present as calcite while no calcite peaks were visible in untreated bricks (Fig. 2e). The newly formed bacterial cement is highly coherent and ensures high physicochemical resistance (Rodriguez Navarro et al. 2012). Bacterially induced calcite crystals are assumed to be more resistant to dissolution since it has been experimentally demonstrated that biomineralized calcite is less soluble than inorganically precipitated calcite (Morse 1983).

Conclusions

The present study reports the success of biogenic calcite crystals formed by *B. megaterium* in reducing the porosity of low energy soil cement bricks. The carbonate crystals strongly adhere to the surfaces and within the pores of soil cement bricks and cylinders by acting as biosealant which leads to reduction in water absorption, porosity, permeability and ultimately enhances the strength of energy efficient soil cement bricks. The brick can successfully replace the fired clay bricks and greatly reduce CO_2 emission. Further use of the technology can be in construction of various built environments using natural materials such as earthen dams, soil stabilization, and mud road construction. Further studies are necessary to evaluate the effect of urea and calcium chloride that are used in this system.

REFERENCES

- Achal, V., Mukherjee, A., and Reddy, M.S. (2011). "Effect of calcifying bacteria on permeation properties of concrete structures". J Ind Microbiol Biotechnol, 38:1229– 1234.
- Achal, V., Mukherjee A., and Reddy, M. S., (2011). Microbial Concrete: A way to enhance the Durability of Building Structures, ASCE J. Materials for Civil Engg, Vol. 23 (6), 730-734.
- Achal, V., Mukherjee, A., and Reddy, M.S. (2010). "Biocalcification by *Sporosarcina pasteurii* using Corn steep liquor as nutrient source". *J Ind Biotechnol*, 6: 170-174.
- Achal, V., Mukherjee, A., Basu, P. C., and Reddy, M.S. (2009). "Strain improvement of Sporosarcina pasteurii for enhanced urease andcalcite production". J Ind Microbiol Biotechnol, 36: 981–988

- Achal, V., Mukherjee, A., Goyal, S., and Reddy, M. S. (2012). "Corrosion Prevention of Reinforced Concrete with Microbial Calcite Precipitation". ACI *Mater J*, 109: 157-164.
- Bonala, M. V. S., Reddi, L. N. (1998). "Physicochemical and biological mechanisms of soil clogging: an overview." ASCE Geotech Spec Publ 78: 43–68.
- Buchanan, A. H., and Honey, B.G. (1994). "Energy and carbon dioxide implications of building construction". *Energy and Buildings*, 20: 205–217.
- De Muynck, W., De Belie, N., and Verstraete, W. (2010). "Microbial carbonate precipitation in construction materials: a review". *Ecol Eng*, 36: 118-136.
- De Muynck, W., Cox, K., De Belie, N., and Verstraete, W. (2008a). "Bacterial carbonate precipitation as an alternative surface treatment for concrete". *Constr Build Mater*, 22: 875–885.
- De Muynck,W., Debrouwer, D., De Belie, N., and Verstraete,W. (2008b). "Bacterial carbonate precipitation improves the durability of cementitious materials." *Cem Concr Res*, 38: 1005–1014.
- Debnath, A., Singh, S. V., and Singh, Y. P. (1995). "Comparative assessment of energy requirements for different types of residential buildings in India." *Energy and Buildings*, 23: 141–146.
- Del Moral, A., Roldan, E., Navarro, J., Monteoliva-Sanchez, M., and Ramos-Cormenzana, A. (1987). "Formation of calcium carbonate crystals by moderately halophilic bacteria." *Geomicrobiol J* 5: 79–87.
- Dhami, N. K., Mukherjee, A., and Reddy, M. S. (2012). "Improvement in strength properties of ash bricks by bacterial calcite." *Ecol Eng*, 39, 31-35.
- Hammes, F. and Verstraete, W. (2002). "Key roles of pH and calcium metabolism in microbial carbonate precipitation". *Rev Environ Sci Biotechnol*, 1, 3–7.
- IS 3495. (1992). Methods of Test of Burnt Clay Building Bricks.
- Le Metayer-Levrel, G., Castanier, S., Orial, G., Loubiere, J.F., and Perthuisot, J.P. (1999). "Applications of bacterial carbonatogenesis to the protection and regeneration of limestones in buildings and historic patrimony." *Sediment Geol*, 126: 25–34.
- Morse, J. W. (1983). "The kinetics of calcium carbonate dissolution and precipitation." In R. J. Reeder (ed.), Carbonates: mineralogy and chemistry. Reviews in mineralogy, 11: 227–264. Mineralogic Society of America, Washington, D.C.
- Oka, T., Suzuki, M., and Konnya, T. (1993). "The estimation of energy consumption and amount of pollutants due to the construction of buildings". *Energy and Buildings*, 19: 303–311.
- Ramachandran, S.K., V. Ramakrishnan, and S.S. Bang. (2001). "Remediation of concrete using microorganisms." *ACI Mater J*, 98: 3-9.
- Reddy, B. V. V. (2002). "Progress of Stabilized Mud Brick Construction in India." Proc National Workshop on Alternative Building Methods, Bangalore, India, 84-94.
- Reddy, B. V. V., and Gupta, A. (2005) "Characteristics of cement-soil mortars." *Mater Struct*, 38: 639–650.
- Reddy, B. V. V., and Jagadish, K. S. (2003). "Embodied energy of common and alternative building materials and technologies". *Energy and Buildings*, 35: 129–137.
- Rivadeneyra, M. A. G., Delgado, A., Ramos-Cormenzana and Delgado, R. (1998). "Biomineralization of carbonates by *Halomonas eurihalina* in solid and liquid media with different salinities: crystal formation sequence." *Res Microbiol*, 149: 277–287.

- Rodriguez-Navarro, C., Jroundi, F., Schiro, M., Ruiz-Agudo, E., and González-Muñoz, M. T. (2012). "Influence of Substrate Mineralogy on Bacterial Mineralization of Calcium Carbonate: Implications for Stone Conservation." *Appl Environ Microbiol*, 78: 4017–4029.
- Samonin, V. V., and Elikova, E. E. (2004). "A study of the adsorption of bacterial cells on porous materials." *Microbiology*, 73: 696–701.
- Stocks-Fischer, S., Galinat, J. K., and Bang, S. S. (1999). "Microbiological precipitation of CaCO₃." Soil Biol Biochem. 31:1563-1571.
- Suzuki, M., Oka, T., and Okada, K. (1995). "The estimation of energy consumption and CO₂ emission due to housing construction in Japan". *Energy and Buildings*, 22: 165–169.
- Vandevivere, P., Baveye, P. (1992). "Relationship between transport of bacteria and their clogging efficiency in sand columns." *Appl Environ Microbiol* 58: 2523–2530
- Whiffin, V.S., Van Paassen, L., and Harkes, M.P. (2007). "Microbial carbonate precipitation as a soil improvement technique." *Geomicrobiol J*, 24: 417–423.
- Zamarreno, D. V., Inkpen, R., and May, E. (2009). "Carbonate crystals precipitated by freshwater bacteria and their use as a limestone consolidant." *Appl Environ Microbiol*, 75: 5981-5990.