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Review Article

Influence of Natural Zeolite and Mineral additive on Bacterial Self-healing Concrete: A Review

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Abstract

With time, the development of micro-cracks in concrete is a frequently reported problem in the structures due to the ingress of harmful substances, leading to the degradation of its quality and strength, which ultimately declines the construction. The present work is a review paper based on enhancing the self-healing property of concrete by inducing different bacteria alone or incorporating different mineral additives. It has been seen that various rehabilitated methodologies are in queue to surmount concrete's weaknesses and to increase its strength and durability. The latest methodology includes using non-pathogenic microbes in concrete as Microbial induced Calcium Carbonate Precipitation (MICCP). The property of precipitating calcium carbonate ($CaCO_3$) crystals by their metabolic activities helps repair the cracks in harsh conditions and improve their strength. Ureolytic bacteria like Bacillus pasteurii/Sporosarcina pasteurii, Bacillus subtilis, Bacillus megaterium, etc., have a specific property by which they can excite urea when integrated with a calcium source and help in sealing the cracks by $CaCO_3$ precipitation. Different studies have observed that specimens having a bacterial concentration of 10^5-10^7 cells/ml with Natural Zeolite (NZ) replacement (10%) represents better interaction of the microstructure of concrete because of the formation of calcium silicate hydrate (CSH) gel. Further, the reduction in CH bond with reduced pore space has also been observed. NZ alone enhances micro-structural property, but it shows $CaCo_3$ precipitation and more densification of microstructure under bacterial combination. XRD also confirms an increase in the calcite composition when the bacterial concentration of 10^5-10^7 cells/ml is used. The overall properties of standard and high-strength bacterial concrete (10^5-10^7 cells/ml) with 10% Natural Zeolite replacement can provide a better option for the future of sustained and strong concrete.

Keywords: Micro-cracks; Bio-mineralization; Encapsulation; Self-healing Properties; Calcite Precipitation.

1. Introduction

Concrete is the second most used material after water and is one of the chief essential ingredients of the construction industry, either in structuring bridges, underground parking, or residential construction. Its usage has been expanded globally from 2.8 to 4.08 billion tonnes in 2019 [1]. Concrete structures always remain exposed to environmental stress, so it is needed to protect them from retaining and preserving them. Though it can withstand compressive forces, tensile forces are tough to resist, so cracks develop quickly when exposed to tension. With time, the depth of cracks increases, and as it prolongs, it meets the reinforcement area, leading to disastrous conditions.

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Similarly, motorway bridges are also prone to such disasters, as salt is used to de-ice the roads and penetrate the cracks, exaggerating steel reinforcement corrosion [2]. Such cracks affect the concrete's durability, i.e., corrosion provides an easy passage for water entry and impurities like Cl^- and SO_4^{2-} in the concrete matrix [3]. Such frightening collapses need to be controlled, by minimizing cracks in the concrete and providing Autogenous healing in them as early as possible, along with the recovery of water tightness [4]. As it is a basic indirect need of man, issues with concrete durability and sustainability demand a practical, economical, feasible, and early solution by the researchers. So, the prime target is to enhance the mechanical properties and improve its durable properties under the manufacturing process. At the same time, the next step ahead will be to focus on reducing cement consumption (i.e., by replacing mineral additives) under the improved methodology for self-healing of concrete.

Being an age-old problem in the construction industry, various techniques can repair initial cracks. Traditional repairing methodology is applied in most countries, which is time-consuming and has multiple limitations; they are based on natural means and can only restore the cracks of concrete from 0 to 100 μm widths [5]. Several modern technologies for healing the cracks are on trial to control such situations in the present scientific era. So, to control the cracks and improve the healing capacity of concrete nowadays, the introduction of new technology, "microbial calcite precipitation" (MICCP) with and without mineral additives (Natural Zeolite 'Clinoptilolite') in concrete is an emerging helpful methodology to repair the cracks autogenously (beyond 100 μm) and improve its durability. This logistic method of bio-mineralization in collapsing sites has attracted the researcher's attention, as microbial self-healing is expected to elongate the service life of concrete structures and enhance their durability and sustainability [6].

The self-healing concrete is designed to extend its lifetime and repair the initial cracks using the Autogenous self-healing technique MICCP before fracture initiation. This technique has been used by Saifee et al. [7], and it was observed that microbial metabolic activity in the concrete leads to enhancing the durability performance of the concrete. Another study on Bacillus pasteurii bacterium was used by Mokhtar et al. [8] indicates that the Bio-calcium carbonate fills a certain amount of void, making the surface texture more compact and resistant penetration. But the bacillus pasteurii gives slightly lower strength than the Bacillus Sphaericus [9]. A study by Manikandan et al. [10] showed that the temperature variation on bacterial strain used could survive from -30°C to 700°C [10]. Sporosarcina pasteurii bacteria used in light aggregate concrete increase the resistance of penetration of chloride ions [11]. It was also found that the overall increase in strength is a result of the appropriate quantity of organic matter in the matrix derived from the biomass of microorganisms at 10^5 cell/ml [12-14]. Strength as a self-healing property as derived by Xu and Wang shows the ability of water absorption in the presence of bacteria with nutrients was assessed by Sorptivity coefficient indicates the recovery after healing was 95% [15]. Healing of artificial crack in the presence of bacteria solution has been observed by Jongvivatsakul et al. [16] under the curing of urea-$CaCl_2$ heal the specimen for about 85% with increasing compressive strength.

Further, the maximum healing of concrete or survival rate gained by freeze-drying technique introduced by Pungrasmi et al. [17] encapsulating the bacteria spores using sodium alginate gives a 100% survival rate after microencapsulation. This phenomenon is also seen in clay soil, showing the strength and stiffness property improved 278% & 177% due to the formation of bacterial precipitations [18]. Self-cementing materials such as silica fume, flyash, metakaolin, and recycled aggregates are frequently used in the studies [19-23]. The self-healing process by encapsulation has been studied by Dai et al. [24]. Incorporating microcapsules with a higher slenderness ratio in concrete beam causes minor bending stiffness, deform easier and slow down the damage propagation along with the height before breakage. The experimental research program is shown in Figure 1.

2. Properties of Natural Zeolite (Clinoptilolite)

2.1. Pozzolanic Activity

Zeolites represent a vast family of alumino-silicates in nature that have been studied by mineralogists for more than 200 years [23]. The Silicate mineral exhibit pozzolanic properties in the presence of water by reacting $Ca(OH)_2$ to form the product CSH, which has the prominent and influential binding ability. This property depends upon the amount and rate by which $Ca(OH)_2$ binds with the active ingredients of pozzolans (SiO_2+Al_2O_3). Clinoptilolite is one such economical alumino-silicate having cage-like structures which ease the ion exchange. Due to the isomorphic interchange of Al^{3+} with Si^{4+} in structure, a lack of +ve charge arises in the framework. That deficit has balanced mono and divalent exchangeable cations such as K^+, Mg^{2+}, Na^+, and Ca^{2+}, forming bonds with the specified number of water molecules and completing the channels framework [25-27]. Zeolite channels are predominantly occupied by Sodium, Calcium, Potassium, and water, as well as traces of Magnesium, Tin, Potassium, Lead, and Barium [28, 29], out of which Na^+, K^+, Ca^{2+}, and Mg^{2+} are easily interchanged with ammonium ions. These exchangeable cations easily find their place in the cavities of Zeolites in hydrated form. If they are removed, regeneration of new cells structure and removal of amorphous Al-species may occur [4].

Thus, the classification and density of exchangeable cations play an influential role in determining the stability of pores and the thermal behavior of Natural Zeolite [30-33]. With an increase in the ratio of Si/Al in K+ ions, crystal structure stability also gets enhanced. Figures 2 and 3 show crystal structure and SEM images. Figures 4 and 5 represent the sieve and XRD images of Clinoptilolite.

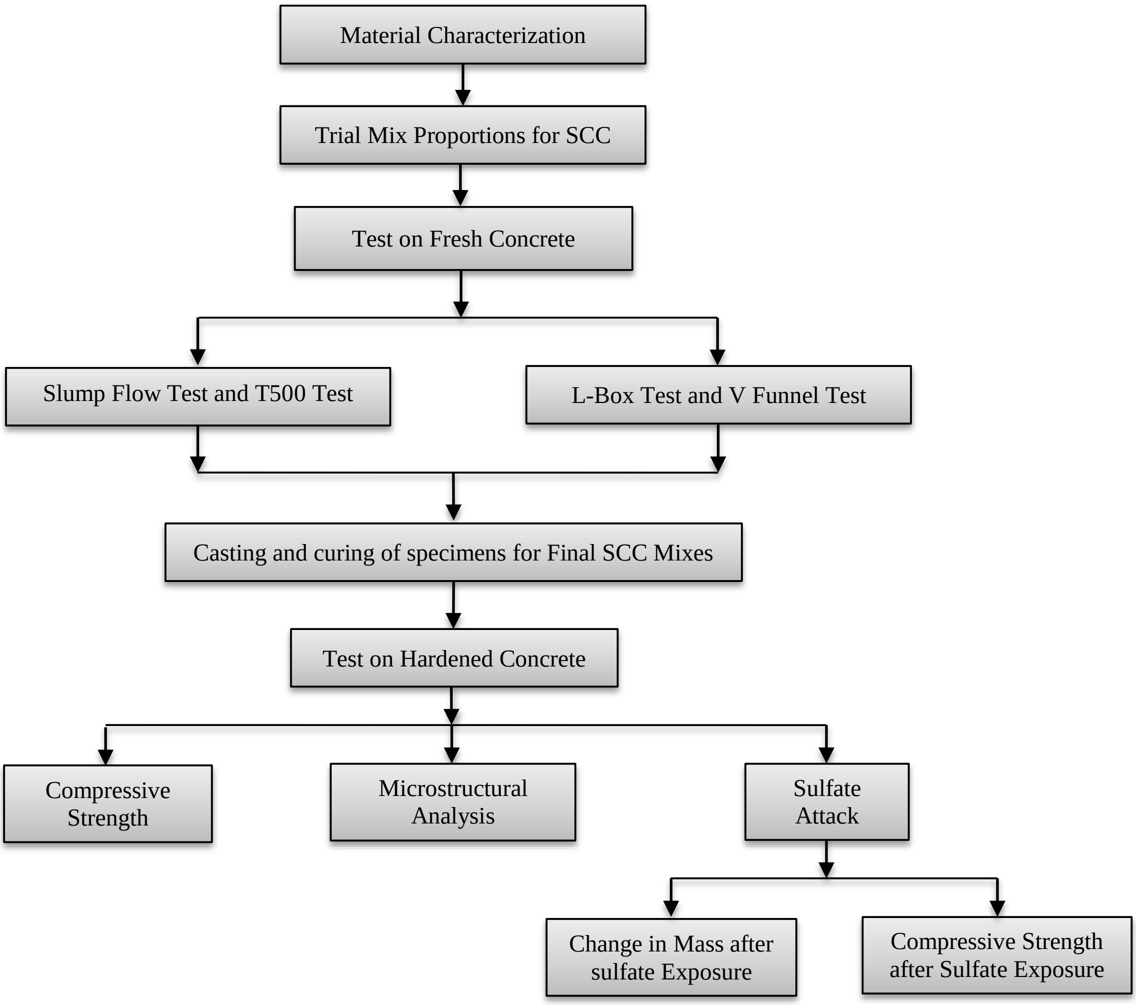


Figure 1. Experimental research program [22]

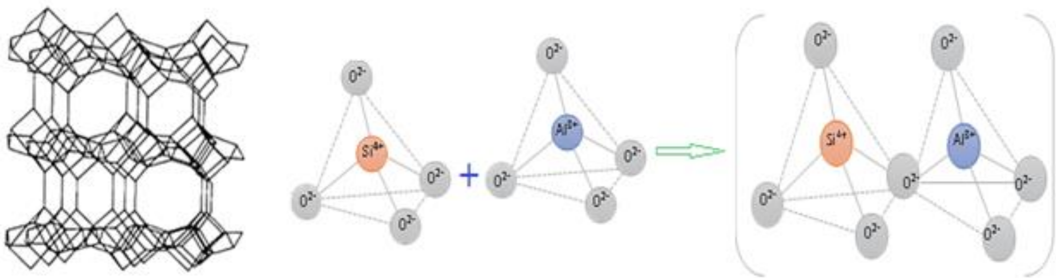
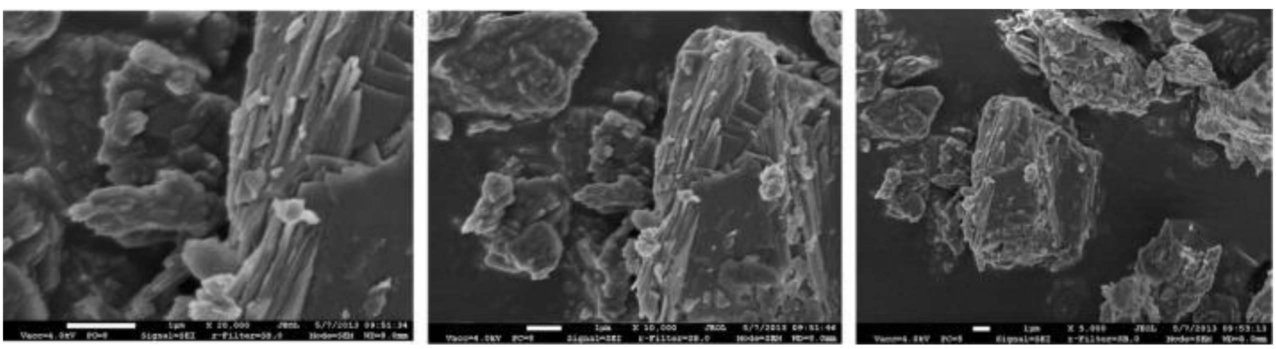


Figure 2. Crystal Structure & Aluminosilicate Structure of Clinoptilolite [31], Primary Building and Secondary Building unit of Natural Zeolite [32]



Magnification up to 20,000 | Magnification up to10,000 | Magnification up to 5,000

Figure 3. SEM. Natural Zeolite 'Clinoptilolite' [31]

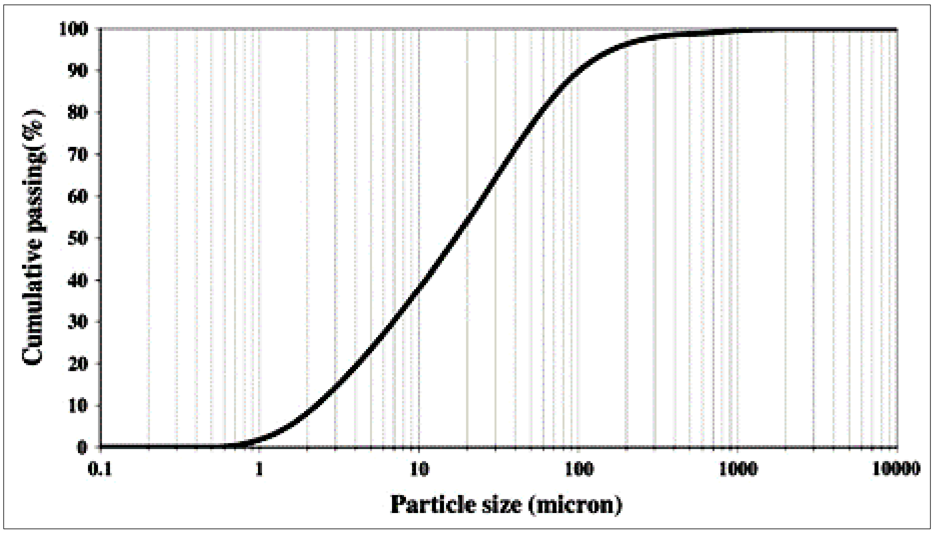


Figure 4. Sieve Analysis Zeolite 'Clinoptilolite' [30]

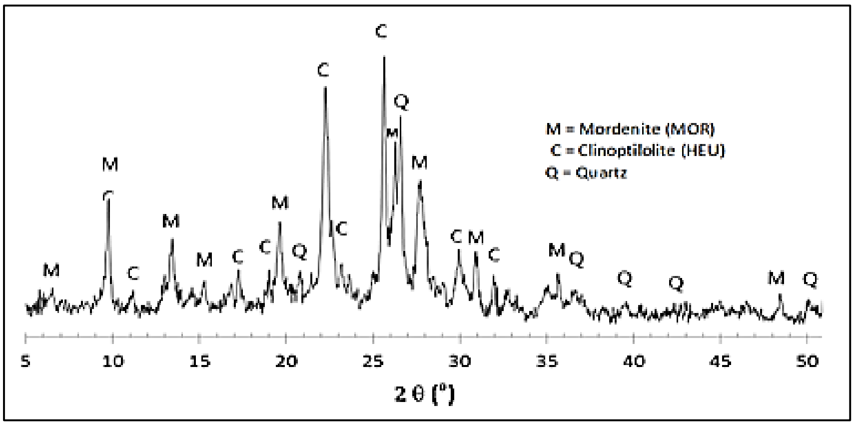


Figure 5. XRD Image of Natural Zeolite (Clinoptilolite) [31]

2.2. Chemical Analysis

Zeolite samples were examined to determine their chemical composition using the usual analytical method for silicate material due to the high proportion of silica (70%) in Zeolite [34-36]. Standard wet chemical analysis/instrumental method is preferred for chemical analysis. Specifically, the concentration of Al_2O_3, Fe_2O_3, CaO, and MgO has been analyzed by titrimetric methods, SiO_2 by gravimetric methods, and Na_2O/ K_2O has been explored by flame photometry [37]. Various studies have been done to date to identify the chemical composition physical properties of NZ shown in Table 1.

Table 1. Properties of 'Clinoptilolite' (wt percent)

Chemical Composition	(wt) Percent	Physical Properties	(wt) Percent
SiO_2	65-72	Appearance porosity	35-45
Al_2O	10-13	Appearance density	2.1-2.3 (g/cm^3)
Fe_2O_3	0.7-1.9	Weight per unit volume	1.2-1.35 (g/cm^3)
CaO	2.0-3.7	Water absorption 'original'	25-35
MgO	0.8-1.2	Water absorption 'grinding'	90-105
Na_2O	0.1-0.65	Oil absorption	51 'g oil/$_{100}$ g sample.'
K_2O	2.1-3.5	Whiteness	68
SO_3	0-0.10	Original bleaching	1.95 'g sample/g tonsil.'
H_2O	3.5-6.80	Active bleaching	1.92 'g sample/g tonsil.'
Si/Al	3-5	pH	6.0-8.0
Cr_2O_3	0-0.01	Specific gravity	2.0-2.4 (g/cm^3)
Loss of Ignition	9-14	Specific surface	22-40 (m^2/gm)
Cations Exchange Capacity	1.5-2.1	-	-

2.3. Thermal Behaviour

The stability index determines the thermal stability of Zeolite. It depends upon the disintegration rate of Zeolite with temperature and can be confirmed by XRD studies. The relation instability index and Si/Al confirm the significance of the latter ratio to manage the thermal stability of NZ shown in Table 2.

Table 2. Thermal behavior of Natural Zeolite Clinoptilolite (NZ) [34]

Si/Al ≥ (more significant than equal to) 3.80	Very stable
Si/Al ≤ (less than equal to) 1.28	Quite Unstable
Intermediate (In between ≥ & ≤) Si/Al	Zeolite stability cannot be directly predicted from Si/Al ratio
The inverse relationship between thermal stability and the ionic potential, $(Z/r)_{wt}$	
$(Z/r)_{wt}$ ≤ (less than equal to) 0.072	Very stable
$(Z/r)_{wt}$ > (greater than) 0.187	Unstable
Intermediate region	$(Z/r)_{wt}$ does not allow discrimination

Volume contraction of Zeolite unit cell depends upon weighted ionic potential. Different factors correspond to framework topology and the expected relationship between the framework density and stability index.

3. Self-healing Process

Many factors are considered in the natural way of healing, such as temperature, degree of damage, freeze-thaw cycles, age of the concrete, and the mortar state. As for the artificial way to repair cracks in concrete is an artificial self-healing process that was first invented in 1994. The primary method was used as a healing agent (adhesive) encapsulated inside a microcapsule. Once a crack forms, it causes the microcapsules to break, releasing the healing agent, hence healing the crack. The adhesives can be stored in short fiber or long tubes [38, 39]. There are many approaches to create innovative concrete and enhance its properties while reducing the cost of widespread use of the material. The bacteria to be used as a self-healing agent in concrete can in such way, i.e., they can be used to perform long-term effective crack sealing, up to the total construction lifetime.

Among which 'bacterial-based self-healing' has been found more capable and easier to handle. Other has certain limitations in maintaining strength and durability of the site for longer duration; this issue can be rectified by adding bacteria in the concrete mix, which have effective crack sealing properties for a long time and can also restore the efficacy of total constructions lifetime. Figure 6 shows crystal formation at crack surface and XRD image of Bacteria Precipitations can be seen from Figure 7.

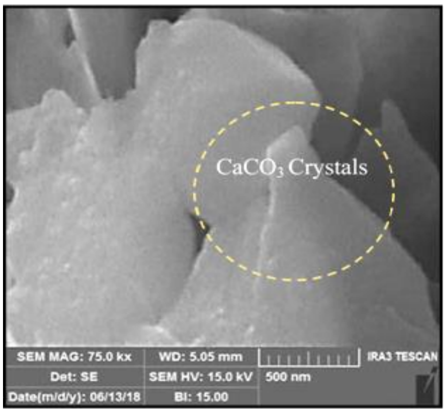


Figure 6. $CaCo_3$ Crystal Formation at Crack surface [40]

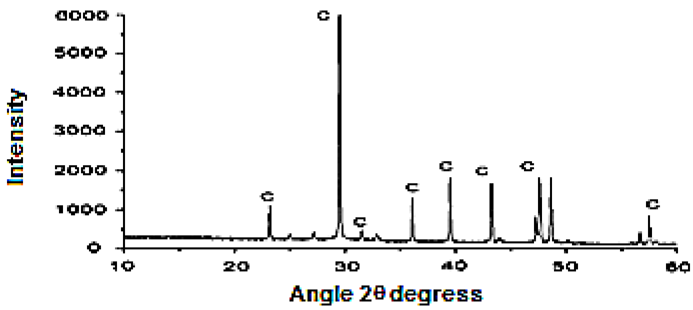


Figure 7. XRD image of Bacteria Precipitation [41]

3.1. Healing Agents in Concrete

Bio-concrete utilizes a simple mechanism to close the micro-crack formation. It involves (*i*) a bearer like calcium lactate (Ca ($C_3H_5O_2)_2$) *(ii)* and bacteria left inside micro-capsules/ encapsulated/immobilized/ just introduced in the mixture, which grows as the water reaches the crack. When bacteria grow, they multiply the metabolic end-products from limestone ($CaCo_3$). Thus, incorporated bacteria in concrete provides an extra protective layer that prevents corrosion in steel [42]. Spore-forming and alkali-resistant bacteria are used in such processes (Table 3), which can also live the entire construction life in dry conditions [43]. The longer life of bacteria aids the healing mechanism due to sustainable organic properties. It has been considered a practical, economical, and more extended self-healing method in construction.

Table 3. Self-healing approach*/techniques and measured variables

S.No.	Approach/Techniques	Crack width & depth	Reference
1	Natural Process*	Crack heals up to 60 µm wide was reported	Parks et al. (2010) [44]
2	Polymer*	Crack up to 138 µm wide filled	Snoeck et al.(2014) [45]
3	Supplementary Cementitious materials (SCMs) *	Crack below 200 µm wide could be filled	Hueng et al. (2014) [46]
4	Bacteria Immobilization /Encapsulation*	Maximum healing of crack width 0.970 mm wide was reported	Wang et al. (2014), Wang et al. (2014), Qian et al.(2015) [47-49]
5	Others (Biological/Chemical) *	Crack healing width up to 0.22 mm was reported	Stuckrath et al. (2014) [50]
6	Micro-encapsulation	Maximum depth of 35 mm crack was filled	Mostavi et al. (2015) [51]
7	Bacteria (Direct)	A maximum depth of 27.2 mm was filled	Achal et al. (2013) [52]
8	Bacteria (liquid 30 ml)	Average crack width of 0.8mm was filled	Luo et al. (2015) [53]
9	Carbonated steel slag	Crack healed was 5 mm in length	Pang et al. (2015) [54]

3.2. Hydration Behavior

3.2.1. Bio-concrete-mechanism

Calcite precipitation occurs with the decomposition of urea by bacteria in the presence of urease enzyme during metabolic activity. Bacteria species have urease enzyme that catalyses urea to ammonia and carbonate. Further, these components get hydrolyzed to carbonic acid and ammonium chloride, leading to calcium carbonate 'calcite crystal'[52].

$$CO(NH_2)_2 + H_2O \rightarrow NH_2COOH + NH_3 \tag{1}$$

$$NH_2COOH + H_2O \rightarrow NH_3 + H_2CO_3 \tag{2}$$

$$H_2CO_3 \rightarrow HCO_3^- + H^+ \tag{3}$$

$$2NH_3 + 2H_2O \rightarrow 2NH_4^+ + 2OH^- \tag{4}$$

$$HCO_3^- + H^+ + 2NH_4^+ + 2OH^- \rightarrow CO_3^{2-} + 2NH_4^+ + 2H_2O \tag{5}$$

Bacteria surface plays a key role in calcite precipitation, as it is negatively charged with neutral pH. Calcium ions with a positive charge easily combine with the bacterial surface and thus encourage nucleation [53].

$$Ca^{2+} + Cell \rightarrow cell - Ca^{2+} \tag{6}$$

$$cell - Ca^{2+} + CO_3^{2-} \rightarrow Cell - CaCO_3 \tag{7}$$

3.2.2. Calcium Carbonate Precipitation

The encapsulation process by MICCP holds the ability of the bacterium to elaborate extra $CaCO_3$ at the cellular level by metabolic activity [55-57]. Other species also show such properties of crystal formation reduces sulfate attack as urea degrading bacteria, unicellular cyanobacteria & silicate consolidated bacteria [57]. A higher concentration of Ca and CO_3 ions are needed for $CaCO_3$ precipitation. As the precipitation is completed, ion activity exceeds the solubility constant (Kso) (as given by Equations 8 and 9. Comparison of ion activity outcomes with solubility, the system's saturation state (Ω) is defined as follows:

If Ω > 1= the system is oversaturated & precipitation would likely appear [58].

$$Ca^{2+} + CO_3^{2-} \leftrightarrow CaCO_3 \tag{8}$$

$$\Omega = \alpha(Ca_2^+) \propto (CO_3^{2-})/Kso \; with \; Kso \; calcite, 25^{\circ} = 4.8 \times 10^{-9} \tag{9}$$

Carbonate ion concentration is linked with bicarbonate, carbonate ions, dissolved carbon dioxide, and the surrounding pH range. While dissolved inorganic carbon (DIC) concentration depends on the temperature and partial presence of CO_2 (as it is vulnerable to the atmosphere), the equilibrium reaction and constant governing the dissolution of CO_2 in aqueous media (25ºC at 1atmp) as shown in Equations 10 to 13 [59]. When $H_2CO_3^* = CO_2\,(aq) + H_2CO_3$, Two different metabolic pathways in the approach of bio mineralization associated with microorganisms may occur as autotrophic pathways & heterotrophic pathways.

$$CO_2(g) \leftrightarrow CO_2(aq) \qquad (PK_H = 1.468) \tag{10}$$

$$CO_2(aq) + H_2O \leftrightarrow H_2CO_3^+ \qquad (PK = 2.84) \tag{11}$$

$$H_2CO_3 \leftrightarrow H^+ + HCO_3^- \qquad (PK1 = 6.352) \tag{12}$$

$$HCO_3^- \leftrightarrow CO_3^{2-} + H^+ \qquad (PK2 = 10.32) \tag{13}$$

3.2.2.1. Autotrophic Pathways

Precipitation of $CaCO_3$ by bacterium converts CO_2 in the presence of Ca^+ in an instant environment. CO3 includes non-methanogenesis, methylotrophic, oxygenic photosynthesis, & oxygenic photosynthesis. Carbon dioxide is used as a carbon source [55, 60].

3.2.2.2. Heterotrophic pathway

$CaCO_3$ precipitation occurs either by the SO_4 or NO_2 cycle. SO_4 cycle is done in the presence of sulfur-reducing bacteria via dissimilate reduction of sulfate [61]. Organic matter degrades by sulfur-reducing bacterium HCO_3^- and H_2S is produced using SO_4^{2-} a terminal electron acceptor Equation 14 [32, 59]. According to [58], gypsum represents the cavities that provide Ca^+ for crystalline calcium carbonate precipitation and SO_4^{2-} for the metabolic process of Sulfur Reducing Bacteria (SRB) Equation 15. Deterioration of organic matter in anaerobic conditions contributes to increased alkaline conditions and facilitates the formation of aragonite crystals.

$$2CH_2O + SO_4^{2-} \rightarrow H_2S + 2HCO^{3-} \tag{14}$$

$$CaSO_4.2H_2O \rightarrow Ca^{2+} + SO_4^{2-} + 2H_2O \tag{15}$$

Introduces a process in which 1mole of urease is separated within a cell to produce an equal amount of NH_3 and CO_3, which spontaneously hydrolyses to form NH_3 and H_2CO_3 [62] as shown in Equations 16 and 17;

$$CO(NH_2)_2 + H_2ONH_2 \xrightarrow{Bacteria} COOH + NH_3 \tag{16}$$

$$NH_2COOH + H_2O \rightarrow NH_3 + H_2CO_3 \tag{17}$$

These products equilibrate with water to form bicarbonate ions and two ammonia and hydroxide ions (Equations 18 and 19).

$$2NH_3 + 2H_2O \leftrightarrow 2NH_4^+ + 2OH^- \tag{18}$$

$$2OH^- + H_2CO_3 \leftrightarrow CO_3^{2-} + 2H_2O \tag{19}$$

Generation of NH_3 on hydrolysis of CH_4N_2O causes pH rise and creates an alkaline micro environment around the bacterial cell. Ca^+ ions over bacteria cell walls start to precipitate as $CaCO_3$ super-saturation reached in Equation 20.

$$CO_3^{2-} + Ca^{2+} \leftrightarrow CaCO_3 \tag{20}$$

Bacterium cell surface plays a vital role in the precipitation of $CaCO_3$ originating site (Equations 21 to 23).

$$Ca^{2+} + Cell \rightarrow Cell - Ca^{2+} \tag{21}$$

$$Cl^- + HCO_3^- + NH_3 \rightarrow NH_4Cl + CO^{2-} \tag{22}$$

$$cell - Ca^{2+} + CO^{2-} \rightarrow Cell - CaCO \tag{23}$$

4. Self-healing Properties in Concrete

4.1. Evolution in Compressive Strength

The maximum stress under gradually applied load a given solid material will sustain without fracture. A study by Valipour et al. [63] gives max compressive strength as the w/b ratio decreases (i.e., 0.35), parallel study with NZ (10%) exhibited the highest strength at 7 & 28 days at w/b ratio 0.4. [33] Concrete (M_{35}) under Zeolite combination found some promising results with 15% replacement, gives a negligible reduction in strength (3.7%, 2.7% & 6.9%) as compared to the control mix at 28, 90 & 365 days. Another study [61] is based on normal and fiber-reinforced mortar under bacteria action (i.e., Sporosarcina pasteurii "10^6 cfu/ml") that retain the compressive strength of 15.38 % and 45% at 270 days as compared to the control mix (55MPa) at 28 days. When limestone replaces (30%) gives a reduction of 19.23% in strength as compared to the control mix, that enhanced up to 22% with the addition of NZ (i.e., 10%) & Sporosarcina Pasteurii (10^7cells/cm^3) [40]. The addition of Silica fume (i.e., 10%) & Alkaliphilic bacteria (10^5cells/ml) retain the strength up to 21.98% as compared to the control specimen [41, 61-65]. The combination of mineral additive with Natural Zeolite introduced by Sreeharsha & Ramana [52] as found the blast furnace slag (BFS) improves the negative effect of NZ at lower percentage replacement in strength at early ages, which gives better result by replacement of NZ (i.e., 10%) at an early age. Modified concrete with Natural Zeolite workout by Nagrokiene & Girskas noted that the strength of modified concrete alone NZ (i.e., 10%) increases up to 13.3% and 15% compared to the control mix 7 and 28 days. The increase of compressive strength in modified concretes can be related to active SiO_2 and Al_2O_3 presence in Zeolite additives [28]. According to previous studies, it has been identified that amorphous silica dioxide is the key factor to enhancing the compressive strength due to a reduction in water absorption [57].

Table 4. Different types of bacteria and their strength results

S. No.	Bacteria used	Best results	Bacterial concentration	Reference
1	Bacillus subtilis	Improvement of 12% in compressive strength as compared to controlled concrete specimens with light weight aggregates	2.8 ×10^8 cfu/ml	Khaliq & Ehsan (2016) [66]
		Compressive strength enhancement of about 14.92% at 28 days with better acid attack factor	10^5 cfu/ml	Rao et al. (2013) [67]
2	Bacillus megaterium	Maximum rate of strength development was 24% achieved in highest grade of concrete 50 MPa	30 ×10^5 cfu/ml	Andalib et al. (2016) [68]
3	Bacillus aerius	Increase in compressive strength by 11.8% in bacterial concrete compared to control with 10% dosage of RHA	10^5 cfu/ml	Siddique et al. (2016) [69]
4	Bacillus sphericus	Tendency to heal the cracks and improve strength and durability. Also seen higher bacterial concentration produced more $CaCo_3$	10^5-10^8 cfu/ml	Wng et al. (2010) Gavimath et al. (2012) [70, 71]
5	Bacillus cereus	Improve compressive strength by 38% and high chloride penetration resistance	10^5-10^7 cfu/ml	Maheswaran et al. (2104) [72]
6	Bacillus flexus	Perform better results in terms of compressive strength and durability	--	Mohari & Kumar (2019) [73]
7.	Bacillus Pseudoforms	Potentially acts as self-healing agent without effecting strength	10^9 cfu/ml	Jonker (2007) [55]
8	Sporosarcina pasteurii	Compressive strength 35% more than the control concrete	10^5 cfu/ml	Chahal et al. (2012) [74]
		Immersion in Phosphate buffer shows better performance concerning strength and durability	--	Ramakrishnan et al. (2005) [75]
9	AKKR5	10% increase in compressive strength as compared to control concrete	10^5 cfu/ml	Siddique et al (2016) [76]
10	Shewanella Species	25% increase in compressive strength of cement mortar compared with the control mortar	10^4 cfu/ml	Siddique & Chahal (2011) Ghosh et al. (2005) [77, 78]

4.2. Water Absorption, Sorptivity/Water Penetration

Water absorption & penetration are two primary components that directly affect concrete durability and inversely illustrate the presence of permeability and reflex details about the concrete specimen's cavity structure. According to Markiv et al. [29], the Natural Zeolite (i.e., 10%) with an air entering agent with superplasticizer showed the lowest penetration depth as 3.5 mm. Characteristic of high dose Natural Zeolite (15 & 30%) identified by Nuruddin et al. [76] indicates a smooth reduction in water penetration/water absorption of about 13% and 40% compared to the control mix at 28 days. Another same study [41] with silica fume under Alkaliphilic bacteria (10^5 cfu/ml) played a vital role in lowering the water penetration depth. The Alkaliphilic bacteria (10^5 cfu/ml) and Silica Fume (i.e., 10%) resulted in a reduction of porosity was about 50 to 55% compared to corresponding control specimens at 28 & 56 days. Another study by S. Bhaskar et al., [64] was based on two different species with separate rates of Sorptivity in three different modes, namely, (i) S-pasteurii + Natural Zeolite, (ii) S-urease + Natural Zeolite & (iii) Nutrient Solution + Natural Zeolite. The maximum reduction was seen in combination one as 78-91% at 4-8 months, whereas the second combination has a reduction of 58.97%. The lowest reduction was seen in the third combination as 14.02% compared to the control mix at different time intervals. In addition of two separate bacteria (S-pasteurii & S-urease), the Calcium Carbonate formation and water penetration depth of cementation seem denser for S. pasteurii than S. urease because of increasing ureolytic activities of enzyme in S. pasteurii.

4.3. Freeze and Thaw Resistance

According to American Cement Manufacturers, it expands about 9 percent when the water freezes. If moist concrete freezes, it produces a specific pressure within the cavity/pores of the specimen. When the pressure exceeds the concrete's bearing capacity, the cavity/pore dilates and ruptures. The accumulated effect of succeeding freeze-thaw cycles and interference of paste and aggregate can finally cause expansion and cracking, scaling, and crushing of concrete. To overcome its effect or attenuate Natural Zeolite's effect, a substitution of cement must be utilized in modern concrete. The latest study by Markiv et al. [32] replacing NZ (10%) in concrete sustains superior resistance in freezing and thawing damage after 100 cycles compared to the control mix. The secondary layer of C-S-H gel formations is because of active Calcium Aluminates Hydrate (C-A-H) reaction leads to lower the capillary porosity effect in concrete incorporated with Natural Zeolite [32]. Najimi et al. [33] highlighted that the closed pores/cavities are the critical factors that increase the sturdiness of concrete. Through repeated freezing, more water penetrates the pores of the conglomerate and retains spoil conglomerate structure until the failure. The highest and lowest values were recorded as 728 and 219 cycles at Natural Zeolite 10% & 0%, respectively [33].

4.4. Drying Shrinkage of Concrete

Shrinkage retains a key position amongst the distortion properties of concrete specimens & influences the quality, durability, and facilities of structures. According to Markiv et al. [33], particle ranges higher than 0.1µm size are most appropriate for deep curing. The higher dose of Natural Zeolite (15 & 30%) showed slightly lower drying shrinkage than the control mix. Another study by Najimi et al. [33] highlighted drying shrinkage of Natural Zeolite at 10%, 20% & 30% replacement level & a new representation of drying shrinkage vs. moisture loss has been seen. No change has been seen in drying shrinkage at moisture loss (i.e., 3.3%) in the control mix, but the addition of NZ (15 & 30%) showed a lower shrinkage level than the control mix. An Introduction of 4% moisture loss with Natural Zeolite (15 & 30%) showed the enhancement in drying shrinkage of about 67% and 50% more than the control mix. Jana, [80] also highlighted Natural Zeolite at 30% caused drying shrinkage 20% more than the control mix.

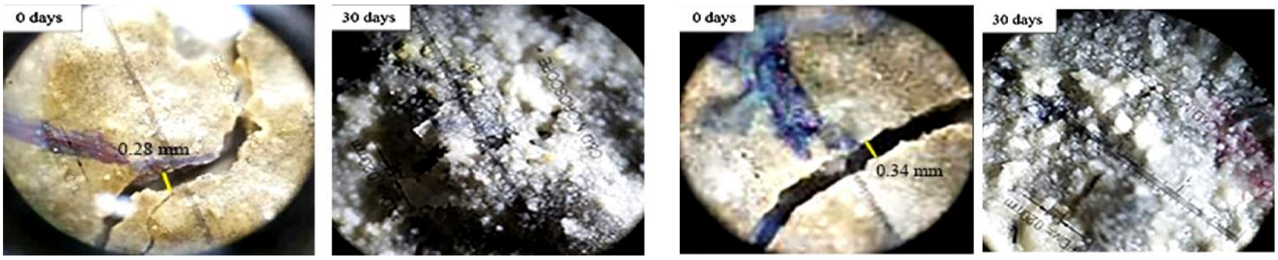
4.5. Sulfuric Acid Deterioration

The penetration of H_2SO_4 explains the weight gain of the concrete specimen. A study by Najimi et al. [33] concluded weight measurement results on control and concrete with Natural Zeolite at early and later ages (i.e., 56 & 300 days). The control concrete enhances the weight continuously at 300 days when immersed in acid (i.e., penetration of H_2SO_4), which was the cause of weight increase. As the decline surface paste was brushed off the Natural Zeolite specimen, the sample lost its weight early (i.e., 56 days). Soon after deteriorating the surface layer of paste and reaching the aggregates, the weight of NZ samples (15% & 30%) started to increase due to the penetration of acid solution at a later age of 300 days. An evident decomposition can be seen on the surface of NZ after 300 days of sulfuric acid exposure so that the aggregates become visible. This kind of deterioration was not seen in Control specimens. Subhashini et al. [81] worked on bacterial culture specimens with different fiber content. The concrete specimen with bacterial culture (10ml) in the presence of steel fiber (0.25%) and polypropylene (0.75%) exhibited better results in a percent reduction in weight was 2.65% at 28 days.

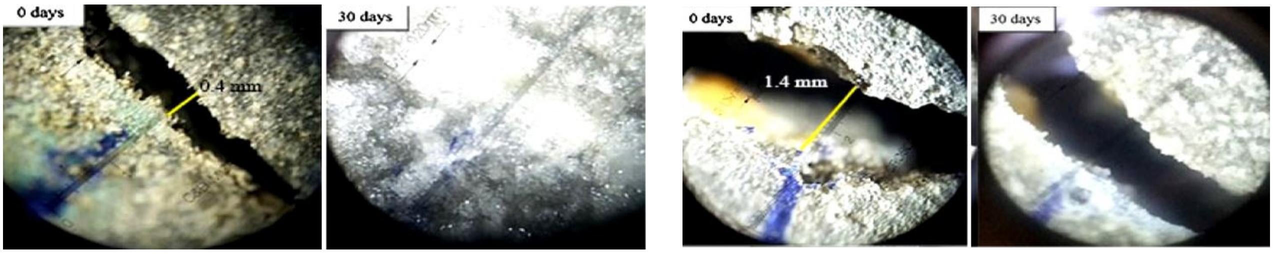
4.6. Rapid Chloride Permeability Test (RCPT)

It is popularly called a column test, which is conducted to understand & evaluate the resistance of chloride ions penetration in concrete as an indicator of its porosity. A study by [41] compares the total charge passed through normal concrete or concrete with mineral additives at different ages. Initially, the SF (i.e., 10%) achieved a minimum coulomb charge transfer (840) compared to the control mix (2525) at 28 days. Also, observe that the drop in coulomb charge departs through concrete because of enriching micro-pores structure of hydrated matrix by addition of silica fume. Additionally, Bacteria (10^5 cfu/ml) with SF (i.e., 10%) displayed an 18% lower charge passed as compared to 840. A Combined effect of bacteria and Nutrient solution highlighted by Bhaskar et al. [64] on chloride permeability. Specimens with (Sporosarcina pasteurii + Nutrient Solution + Zeolite) gives minimum coulomb charge was 4946, 3930 & 3174 as compared to control mix (7189) at 120, 180 & 240 days, respectively. The highest value of percentage reduction in chloride ion penetration was 55.85% at 240 days. Calcite precipitation could edge minor capillary & fewer blocking of pores, which has substantially minimized chloride ions perforation. Betterment with aggregate cement paste link because of calcite precipitation also plays a vital role in lowering chloride ion permeability. Second study by Eskandari et al. [82] based on Zeolite content (i.e., 5% & 10%) at w/c ratio 0.45 & 0.4. A minimum value of Chloride permeability of 1510 & 1220 was recorded under NZ (10%) at w/c ratio 0.45 compared to control mix (2953) at 28 & 90 days, respectively. Further, the lowest reduction of charge (716) has been seen on NZ (5%) at 90 days with the lowest w/c ratio (i.e., 0.4). Advance study on Micro nanobubbles introduced by Mohsen Zadeh et al. [80] with Natural Zeolite & Metakaolin. Micro Nanobubbles of water (100%) with NZ & MK (i.e., 10%) give the minimum value of charge passed (939.50 & 468.75) as compared to the control mix 4164.66 at 28 and 90 days, respectively. Another study by Toklu et al. [81] highlighted the chloride permeability of concrete under Natural Zeolite & Blast furnace slag at different combinations. Experimental result shows lowest charge 221 & 651 achieved by two different combinations at NZ (i.e., 20%) alone and BFS+NZ (i.e., 10%) compared to control batch 3780 at 90 days. Figure 8 Shows SEM Micro Structure/Self-healing of Concrete specimens.

(a) Slica Fume (10%) [68] (b) Bacteria + silica fume (10%) [59] (c) Bacteria + NZ (10%)[40]



(d) Bacteria Specimen without Mineral additive healing up to 0.28 mm (e) Bacterial specimen NZ10% & Lime Stone 20% healing up to 0.34 mm



(f) Bacteria Specimen Curing Condition Urease-$CaCl_2$ at 5 days healing up to 0.4 mm (g) Bacteria Specimen Curing Condition Urease-$CaCl_2$ at 28 days healing partially up to 1.1 mm

Figure 8. SEM Shows Micro Structure/Self-healing of Concrete specimen [40]

Table 5. Uses of Natural Zeolite in cement, cement mortar, and concrete composite

Test Perform	**Natural Zeolite Clinoptilolite replacement % age**				
	Up to 10	**≥ 10 ≤ 20**	**≥ 20 ≤ 30**	**≥ 30 ≤ 40**	**≥ 40 ≤ 50**
Strength properties	Markiv et al. (2016), Ahmadi & Shekarch (2010), Vejmelkova et al. (2015) [32, 85, 87]	Ahmadi & Shekarch (2010), Vejmelkova et al. (2015) [85, 87]	Jana (2007), Nas et al. (2018), Vejmelkova et al. (2015) [80-86, 87]	Jana (2007), Karakurt & Topcu (2011) Vejmelkova et al. (2015) [80, 87, 89]	Uzal et al. (2011) Karakurt & Topcu (2011) Vejmelkova et al. (2015) [87, 89, 90]
Transport properties	Ahmadi &Shekarch (2010), Nas et al. (2018), Toklu.(2021), Karakurt & Topcu (2011), Vejmelkova et al. (2015) [84-87, 89]	Ahmadi & Shekarch (2010), Karakurt & Topcu (2011), Maheswaran et al. (2014) [72, 85, 89]	Jana (2007), Toklu (2021), Bilim (2011), Karakurt & Topcu (2011) [80, 84, 89, 91]	Jana.(2007), Vejmelkova et al. (2015) [80, 87]	Uzal et al. (2011), Vejmelkova et al. (2015) [87, 90]
Alkali–silica reaction	Ahmadi & Shekarch (2010), Jana (2007), Karakurt & Topcu (2011) [80, 85, 89]	Ahmadi & Shekarch (2010), Jana (2007), Karakurt & Topcu (2011) [80, 85, 89]	Jana (2007), Karakurt & Topcu (2011) [77, 86]	Ahmadi & Shekarch (2010), Karakurt & Topcu (2011) [85, 89]	Karakurt & Topcu (2011) [89]
Freeze and thaw resistance	Mohari & Kumar (2019) [73]	Siddique et al. (2015) [76]	–	–	–
Sulfate and acid resistance	Jana (2007), Karakurt & Topcu (2011) [80, 89]	Jana (2007), Karakurt & Topcu (2011), Janotka & Stevula (1998) [80, 88, 89]	Jana (2007), Karakurt & Topcu (2011), Janotka & Stevula (1998) [80, 88, 89]	Karakurt & Topcu (2011), Janotka & Stevula (1998) [88, 89]	Karakurt & Topcu (2011), Janotka & Stevula (1998) [88, 89]
Drying shrinkage	Markiv et al. (2016), Jana (2007) [32, 80]	Jana (2007) [80]	Jana (2007) [80]	–	–

Heat of hydration	Vejmelkova et al. (2015), Krolo et al. (2015) [87, 92]	Vejmelkova et al. (2015) [87]	Vejmelkova et al. (2015) [87]	Vejmelkova et al. (2015) [87]	Vejmelkova et al. (2015) [87]
Corrosion of reinforcement	Vejmelkova et al. (2015) [87]	Vejmelkova et al. (2015) [87]	Vejmelkova et al. (2015) [87]	Vejmelkova et al. (2015) [87]	Vejmelkova et al. (2015) [87]
Carbonation	Krolo et al. (2005) [92]	Krolo et al. (2005) [92]	Krolo et al.(2005) [92]	–	–

4.7. Scanning Electron Microscopy and XRD Analysis

Scanning Electron Microscopy is used to identify the morphology of the deposited materials in the cracks and demonstrate the microstructure properties of concrete, well-developed $Ca(OH)_2$ and C-S-H crystal formation. The particles of Natural Zeolite has identified by Najimi et al. [33] are overlapped or plate-shaped with an average size of 11.82µm, crucial for pozzolanic activity. Another investigation by Nagrockiene & Girskas [31] revealed that the maximum and lowest density by SEM shows as 2404 and 2380 (kg/m^3) at Natural Zeolite (10% & 0%). A new combination of Micro nanobubbles was introduced by Mohsen Zadeh et al. [80] with Zeolite & Metakaolin. SEM shows Zeolite & Metakaolin (i.e., 10%) in the presence of Micro nanobubbles of water (i.e., 100%) improves the property of concrete in transfer region, decreases fraction, and enhances the cohesion compared to control mix. The high coherence & density of concrete specimen having a magnification of 100 µm is related to the sample containing Silica Fume & Metakaolin (i.e., 10%) mixed with Micro nanobubbles of water (i.e., 100%). Temperature effect on Natural Zeolite by Nas et al. [87] focused on SEM investigation for the inner structure of the concrete (i.e., 200-800 ºC). Concrete unexposed to the high-temperature effect is dense, and CSH crystals consist of vicious blocks. Zeolite specimens contain pozzolanic properties, resulting in dense and low porosity microstructure with reduced CH content and intense CH content CSH crystals. The combination of fiber and Bacteria in mortar specimens has initiated by Bhaskar et al. [64].

Through SEM, the presence of S-pasteurii shows a rich amount of minerals capable of sealing freshly formed cracks. A thin lining of minerals precipitate is seen at the crack surface wall in the presence of nutrients alone. This thin lining of minerals precipitation disappeared or was absent in control concrete. The presence of other calcite through silica fume is considered by Siddique et al. [41] because of its active pozzolanic property. SEM shows the presence of Silica Fume (i.e., 10%) represents better microstructure interaction because of a secondary formation of CSH gel. Compared to the control mix, a reduction in the C-H bond with minor pores space has been found. Further addition of S-pasteurii (105cell/ml) clearly showed calcite precipitation & densification of microstructure, as the cause in the enhancement of strength & decrease in porosity Figures 8-a and 8-b. Limestone powder has introduced by Jafarnia et al. [40] with Sporosarcina pasteurii (10^7cell/ml) and Natural Zeolite combination. Natural Zeolite (i.e., 10%) is capable of enhancing the Microstructural properties alone, but with a combination of S-pasteurii, PTCC 1645 (10^7cell/ml) showed a more dense structure Figure 8-c. Crack healing by S-pasteurii (10^7cells/ml) without any additive shows 0.28mm cracks get completely cured at 30 days Figure 8-d. The enhancement in Crack healing identify in combination (S-pasteurii + NZ (10%) + limestone (20%)) that a width of 0.34 mm completely healed at 28 days. Further under Curing Environment ('Urease-$CaCl_2$') observed that up to 0.40 mm wide cracks gets completed healed formerly at 5 days Figure 8-f. Despite that it has seen that 1.1 mm (crack width) has partially cured in 28 days Figure 8-g.

Secondly, the X-Ray Diffraction device displays the crystalline stages existent in the concrete and thereby reveals chemical composition information. For this, the study by Siddique et al. [41] shows that the chemical composition of bacterial concrete with and without silica fume (10%) imprints the summit of quartz (Q), calcite (C), Calcium Silicate Hydrate (CSH), ettringite (E) and larnite (L). The quantification analysis of Silica Fume with Sporosarcina pasteurii (10^7 cell/ml) shows the calcite composition enhanced significantly under Sporosarcina pasteurii; amorphous content was also present in crystalline stages with the rise from 25º to 28º (Figure 5). Two different bacteria (i.e., S. Pasteurii & S. Urea) with zeolite have been initiated by Bhaskar et al. [64]. This indicates calcite's dominant crystal morphology. The percentage of $CaCO_3$ in the residue was significantly higher in the bacteria-incorporated mix than in those without bacteria.

5. Summary and Discussion

A giant crack is an alarming signal, but the hidden micro-cracks are the big dragons inside; even a minor crack may lead to ruinous conditions. Cracks can shake the site to disturb the structural integrity, but minor cracks may cause terrible durability issues. Even a crack of a sub-millimeter size may cause the connecting links to increase their size and the permeability of the concrete matrix. It acts as a doorway for the entry of water and chemicals into it, ultimately causing pre-mature matrix deterioration and corrosion of embedded steel reinforcement. Also, strength and durability always remain integral and demanded factors in any concrete used for construction, so minor cracks should always be kept in mind, which usually develop with time in the structure. It demands standard maintenance and restoration of concrete constructions at different times, which is too costly and unavailable. In this scenario, an economical, feasible autonomous self-healing repair mechanism such as Microbial induced calcium carbonate precipitation (MICCP) seems to be highly beneficial in reducing maintenance charges. It will also help to increase the material's durability.

A lot of work has been done to explore bio-mineralization. However, still, there is much more to be done to ensure the reproducibility and durability of the self-healing mechanism. It demands long-term study to affirm the claim of having hard concrete for decades by using a mixture of bacteria and natural zeolite in the concrete mix. Identifying the desirable transition zone in filler and crack-edge consolidation, which helps prevent crack formation developed in the process, has another future objective of approving the strength.

Though it has been claimed to be economical compared to other traditional approaches, it still affirms that bio-concrete production is another area of study through a solid-based medical approach. Investigation into reducing the costs of the specific microbe, its nutrients, maintenance, and labor is also needed. This strategy would encourage users to adopt bio-concrete as an innovative material as a future aspect to encourage self-healing concrete on an industrial scale.

6. Conclusions

Building materials are always in demand for construction in terms of their strength and durability criteria. Self-healing techniques may benefit in maximum parameters of acclaimed concrete. It succeeds the traditional tactics simply by mixing healing treatment agents with the concrete and other materials during casting. The following key points are concluded in the present study:

- Researchers have found that the amorphous silica dioxide is critical in enhancing the morphology of concrete specimens to a significant level, as an increase in the compressive strength/tensile strength, a reduction in water absorption, and permeability.
- The crack healing ratio is directly related to the width of the crack and the cracking age. A decrease in the crack healing capacity has been seen with the progress in cracking age. More wide cracks show the decrease in the healing ratio due to the release of $CaCo_3$ in the water environment from there.
- The reduction in pore size has been seen in dense microstructure that declines the bacteria's viability and is responsible for decreasing the self-healing properties, also investigated by SEM, EDS, and XRD.
- Bacterial concrete replacement of 100% natural aggregate by recycled aggregate increases the damage level due to the inferior mechanical properties present in recycled aggregate. Metakaolin and bacteria play an important role in reducing this level of damage. Due to the deposition of bacterial calcite precipitation and the acceleration of the hydration reaction of OPC by Metakaolin, the damage level was seen to decrease with age.
- During the research, it has been concluded that the spore-forming ability and high ureolytic activity of zeolite immobilized bacteria (i.e., Sporosarcina Ureae and Sporosarcina pasteurii) sustain them in a high pH environment and also produce self-healing compounds.
- SEM and EDS studies showed that the incorporated bacteria produce a copious amount of minerals that can seal the freshly formed crack in the concrete.
- Self-combination of natural zeolite and silica fume (i.e., 10%) enhanced the compressive strength up to 15%, further increased by 20-22% by adding S-Pasteurii (10^5-10^7 cells/cm^3) in a different mode. A mixture of zeolite, silica fumes, and blast furnace slag (i.e., 10%) presents much better results. Still, the presence of bacteria (S-Pasteurii) among all strengths proves to be best at a concentration of 10^5 cells/ml.
- Studies point out that the utilization of natural zeolite (i.e., 10%) in concrete sustains prominent resistance to freezing and thawing damage after 100 cycles compared to the control mix.
- The lowest penetration (i.e., 3.5mm) was seen at the substitution of natural zeolite (i.e., 10%), which can further be seen as a reduction in water penetration of 40% at a higher dosage of natural zeolite (i.e., 30%). The combination of Silica fume (i.e., 10%) and Alkaliphilic Bacteria (10^5 cfu/ml) reduced porosity by 50-555%, whereas Natural Zeolite (i.e., 10%) with S. Pasteurii (10^5 cfu/ml) reduced porosity by 78-91%. It has been observed that bacteria and Zeolite combination highlighted the maximum reduction in water penetration.
- Applying a higher dose of natural zeolite (i.e., 30%) enhances the drying shrinkage of standard concrete by up to 20%, which can further be seen in increasing mode (i.e., 67%) by using 4% moisture loss. It shows a higher dose of natural zeolite is effective alone and with moisture loss.
- The penetration of H_2SO_4 in concrete with bacterial culture (10ml) exhibited a better result in a percent reduction in weight.
- The permeability of concrete having silica fume (i.e., 10%) alone exhibited a reduction of 66.7%, which was further recorded at 72.71% after the incorporation of bacteria (10^5 cfu/ml). The second combination, bacteria + zeolite (i.e., 10%), was recorded as 55.85% and MK + NZ (i.e., 10%) + micro nano bubbles of water (i.e., 100%) caused a maximum reduction of 88.74%.

7. Declarations

7.1. Author Contributions

Conceptualization, J.N.A.; methodology, J.N.A.; validation, J.N.A.; investigation, J.N.A. and M.N.A.; data curation, J.N.A., M.N.A., and J.K.N.; writing—original draft preparation, J.N.A.; writing—review and editing, M.N.A.; visualization, J.N.A.; supervision, R.A.K. and R.A.K. All authors have read and agreed to the published version of the manuscript.

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Data sharing is not applicable to this article.

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7.5. Conflicts of Interest

The authors declare no conflict of interest.

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