REVIEW



Self-healing concrete for sustainable buildings. A review

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Received: 7 November 2021 / Accepted: 17 December 2021 / Published online: 5 January 2022 © The Author(s), under exclusive licence to Springer Nature Switzerland AG 2022

Abstract

A total of 12 billion tons of concrete materials are produced annually, about 2 tons per person. More sustainable buildings are thus needed to decrease the carbon footprint of concrete infrastructures in the context of climate change. Crack formation is a major flaw of concrete structures. Although cracks are usually small and do not necessarily induce building collapse, cracks reduce the life span and sustainability of buildings. Therefore, research has developed self-healing materials that are capable of repairing narrow cracks automatically. Here we review self-healing technologies such as adding mineral mixtures, bacteria, and adhesive liquids. Mineral healing is economically positive, yet relies on suitable conditions such as the presence of water, and is less efficient to heal larger cracks. By contrast, the bacterial encapsulation is promising due to the uniform characteristics of bacteria in the alkaline environment of concrete.

Keywords Concrete · Bacteria · Healing mechanism · Sustainability

Introduction

Crack development in concrete is a major factor in impairing the functional integrity of the structure, hence, limiting its life span and sustainability, as a consequence of unwanted

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incidences such as thermal deformation, external stresses, and shrinkage development (Kim et al. 2021; Sirtoli et al. 2020). The cracks cause potential damage to concrete structure, in particular, when harmful liquids and gases (e.g., sulphide, chloride) can penetrate into the matrix through these cracks (Van Tittelboom and De Belie 2013). As a consequence, cracks may become grower and reinforcement starts to contact with environment. This phenomenon can cause a total collapse of the structure when reinforcement starts to corrode (Fang et al. 2021; Van Tittelboom and De Belie 2013). Thus, it is clear that the repair of cracks needs a rigorous inspection and maintenance to improve the durability of concrete structures. Such an incidence happened recently in Ikoyi, Lagos, Nigeria, where a 21-storied building suddenly collapsed (The BBC 2021; The Sahara Reporters 2021). The experts initially suspect that the collapse may occur due to greater magnitude of cracks resulting from improper structural design, bad materials, stress, and poor maintenance of structure (The Vangurad 2021). However, repairing cracks remains challenging when it is difficult to identify their precise locations (Basheer and Cleland 2006; Zhang et al. 2020). In addition, the cost associated with repairing work is estimated to be equivalent to half of the yearly construction budget (Cailleux and Pollet 2009). Therefore, developing self-healing materials that are capable of repairing narrow cracks automatically have attracted a great deal of attention from researchers and practitioners (Fernandez et al. 2021; Islam and Bhat 2021; Joshi et al. 2017; Kim et al. 2021; Sidiq et al. 2019; Sirtoli et al. 2020; Tang et al. 2015; Vijay et al. 2017).

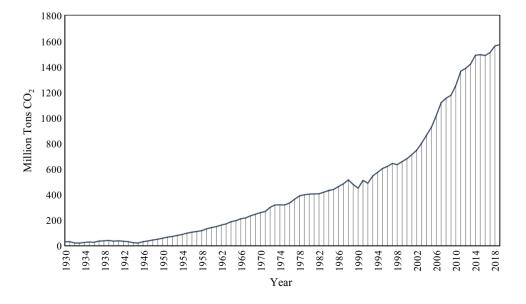
The most consumed construction material in the world, concrete, is used in many infrastructures such as bridges, roads, railway lines, and other buildings as building blocks. Today, concrete is chosen because of its high resistance to water, low energy input, easy casting, and cost-efficient characteristics compared to other construction materials (Rodrigues and Joekes 2011; Zhang et al. 2017). Yet, the production of concrete itself consumes a lot of energy (i.e., 0.95 MJ/kg) and leads to the emission of large amounts of CO₂ (i.e., 0.35 kg C/kg) (Sangadji 2017). The concrete cement industry has been considered as the third-largest source of CO₂ emissions after fossil fuel combustion and land-use change. Andrew (2019) presented a global estimate of CO₂ emissions from concrete cement production during 1930–2018 (Fig. 1). In 2018 alone, 1.50 ± 0.12 Gt of CO₂ emissions were emitted worldwide, equal to 4 percent of fossil fuel emissions (Fig. 1).

In addition, due to rapid urbanization and industrialization, with increasing consumption rate concrete-made structure requires a large amount of material cycling and possess high energy demand during each stage of lifecycle that leads to the high potential of environmental risk (Alves and Sanjurjo-Sánchez 2015; Stanaszek-Tomal 2020). However, to be fitted in the definition of sustainable building material, these environmental burdens should be minimized (Stanaszek-Tomal 2020). Therefore, an efficient design process is required that can ensure sustainable use of materials, can reduce operational costs, save total energy, and minimize the environmental impact of building structures (Sangadji 2017).

Furthermore, concrete, a low tensile strength material (Wang et al. 2019), is responsive to environmental stresses and with time may develop cracks that will further weaken and lessen the durability of the infrastructure (Gupta et al. 2017; Khaliq and Ehsan 2016; Mauludin and Oucif 2019; Vijay et al. 2017; Wiktor and Jonkers 2011). However, concrete usually has micro-cracks that are formed at the early stage of service due to shrinkage and thermal expansion (Guo and Chidiac 2019). These micro-cracks can form a continuous network and finally develop macro-cracks. This can happen also in cases where reinforcement bars have been embedded, where the development of cracks will make them prone to corrosion when they are exposed to air and water (Zhang et al. 2017). Moreover, the presence of cracks in concrete allows the passage of gases & liquids and deleterious chemicals (Guo and Chidiac 2019; Magaji and Yakubu 2019; Sangadji 2017; Zhang et al. 2020); and it is undesirable that waste penetrates through concrete in conditions where it must act as a barrier. Difficulties may also arise if cracks develop in the concrete used in an underground infrastructure because it might be impractical or impossible to repair. Repairing these cracks is also very expensive even can be more than the manufacturing cost (Gupta et al. 2017; Sangadji 2017; Vijay et al. 2017; Zhang et al. 2017). Nowadays, countries and nations are investing a large amount of money in the maintenance and development of infrastructures (Gupta et al. 2017; Joshi et al. 2017; Vijay et al. 2017). For example, the Netherlands spent one-third of the annual budget for major construction works on inspection, maintenance, and repair, while the UK accounts for more than 45% (Sangadji 2017).

As a solution to the challenge of cracking in concrete, self-healing concrete can be used as suggested by many studies (Bekas et al. 2016; Danish et al. 2020; Gupta et al.





2017; Kumar and Lakshmi 2020; Sangadji 2017; Tang and Xu 2021; Vijay et al. 2017; Wu et al. 2012; Xue et al. 2019; Zhang et al. 2017). Self-healing concrete is widely recognized for its intrinsic autogenous healing powers. When hydration in the mixture is preserved, the mixture may heal, thus creating a stable structure. But, auto-healing is restricted to minor fractures and is beneficial only when water is available. However, concrete is modified to allow it to function autonomously. Furthermore, concrete may be changed to create an autonomous crack healing mechanism (Van Tittelboom and De Belie 2013). This technology possesses the ability of self-repair once cracks develop by being blocked or sealed with secondary products of physical and chemical processes. Therefore, this review article critically assessed the sustainability of different self-healing technologies and their opportunities and barriers for industrial-scale implementation.

Self-healing mechanisms

Concrete is somewhat capable of repairing cracks naturally, as referred to autogenous self-healing (Hearn 1998). This technology has been used as a successful indicator of crack treatment which can work independently irrespective of the crack position. The use of healing materials in concrete technology is now gaining popularity due to its significant advantages. The main concept of autogenous healing is that materials used in this technology can self-heal by nature (Ayobami Adebola et al. 2020). The mechanisms have been investigated in several studies, and later on, summarised by Sangadji (2017) who found four major mechanisms. The autogenous self-healing mechanism can firstly be by swelling of the cement matrix close to the cracks due to the absorbed water through hydrated cement paste. Secondly, by accelerating the hydration process through hydration of water-deficient hardened cement which in turn occupies the voids. Thirdly, during the hydration process, the formation of calcium carbonate takes place through the reaction of calcium ions (Ca^{2+}) with carbonate (CO_3^{2-}), which precipitates in the cracks. This approach is believed to play the most pronouncing contribution in the healing mechanism (Joshi et al. 2017; Vijay et al. 2017). And, the final mechanism is to make clogging of small particles that disintegrated from crack surfaces or are transported by ambient water (Sangadji 2017). This reveals that a crack should develop first for selfhealing to take place.

Self-healing processes either can be by autogenously or autonomously. However, Huang et al. (2016) classified the healing processes into four groups i.e., autogenous selfhealing, mineral admixture-based self-healing, self-healing based on bacteria, and adhesive-based self-healing.

Autonomous self-healing

Autonomous healing processes are purposefully engineered to achieve better and sustainable self-healing results (Guo and Chidiac 2019; Rajczakowska et al. 2019). The healing process is by supplying healing agents to the cracks such as mineral admixture, bacteria, adhesives (Gupta et al. 2017; Huang et al. 2016; Nasim et al. 2020), and encapsulated materials with pozzolan (De Belie et al. 2018; Stanaszek-Tomal 2020; Van Belleghem et al. 2018).

Mineral admixture-based self-healing

Concrete mixed with mineral admixtures is healed when water ingress into the cracks and reacted with the mineral additives present. This can be done either by an expansive or crystalline mineral admixture (Chang et al. 2021; Huang et al. 2016; Sisomphon et al. 2012; Wu et al. 2012). The expansive additives expand to a larger volume within the concrete matrix by reacting with water. The investigated additives are free lime (CaO) and anhydrite (CaSO₄), calcium sulfoaluminate ($Ca_4(AlO_2)_6SO_4$) (Kishi et al. 2007) and some geo-materials contain silicon dioxide, montmorillonite clay, and sodium aluminum silicate hydroxide (Ahn and Kishi 2010). The expansion transpires inside the concrete matrix and fills up the crack, however, it can also lead to damage in case of larger expansion (Huang et al. 2016). The crystalline additives consist of crystalline catalysts and reactive silica, which form crystals when react with Ca(OH)₂ and water (Sisomphon et al. 2012).

Self-healing based on bacteria

Microbial metabolic activity-induced mineralization to fill up cracks was suggested in one of the first studies of its kind by Gollapudi et al. (1995) which later lead to bacteriabased self-healing approaches. However, only a few species of bacteria survive the concrete 'casting' process and can adapt and even form spores in the high alkaline environment of concrete. The spores remain inactive and survive high temperature and pressure, dehydration, and chemical processes, and can become metabolically active when water (ingress) and nutrients become available to them. Through various metabolic pathways, these bacteria start to produce calcium carbonate by using Ca^{2+} , CO_2 and water from the surrounding environment. The metabolic pathways include urea hydrolysis (Gollapudi et al. 1995), conversation of organic compounds (Jonkers and Schlangen 2009), oxidation of organic acids (Huang et al. 2016), or denitrification (Sangadji 2017). The most studied mechanism of concrete self-healing has been on the use of bacterial species such as Bacillus pseudifirmus, Sporosarcina pasteurii (Khaliq and Ehsan 2016; Sangadji 2017), and Bacilus cohnii (Zhang et al. 2017), and other species used are *Bacillus lentus* (Dick et al. 2006), *Bacillus alkalinitrilicus* (Wiktor and Jonkers 2011), *Bacillus subtilis, Bacillus sphaericus, Escherichia coli*, and *Bacillus balodurans* (Stanaszek-Tomal 2020). The significantly larger cracks (0.46 mm-wide) were reported to be healed in concrete after 100 days of healing (Wiktor and Jonkers 2011) by using bacteria-based healing methods (Dinesh et al. 2017; Justo-Reinoso et al. 2021) (Table 1).

Adhesive-based self-healing

The last approach is by supplying one or multi-component adhesive healing agent to the concrete. This is done either by encapsulation or vascular techniques. In the capsule-based technique, an adhesive is sequestered inside distinct capsules which are released upon rupture whereas, in vascular techniques, the healing agent is supplied through the network of hollow tubes. The hollow tubes networks ideally connect both the interior and the exterior of the structure (Magaji and Yakubu 2019; Van Tittelboom and De Belie 2013). The most studied adhesive agents are cyanoacrylate, silicon (Dry et al. 2003); tung oil (Cailleux and Pollet 2009); Methylmethacrylate (MMA), polymethylmethacrylate (PMMA), and triethylborane (TEB) (Yang et al. 2011).

Sustainability assessment

Globally, 12 billion tons (2 tons per person per year) of concrete materials are produced annually (Sangadji 2017). In addition, the cost for repair and maintenance of crack has been estimated at 147 USD per cubic meter of concrete, even though production costs are around 70 USD per cubic meter (Seifan et al. 2016). Moreover, albeit European Standards consider that any concrete should have a service life of 50 years, the contractors are generally responsible for damages only until 10 years, and usually, cracks are not included, so maintenance and repairs must be done (Silva et al. 2015). This is seen in the impact of durability-related problems on a national-economy scale due to large sums addressed to fixing concrete structures, such as in the United States, where 4 billion dollars are invested yearly in repairing concrete highway bridges due to corrosion of materials (Jonkers et al. 2010). In this way, it is seen that it is of extreme importance to find new sustainable alternatives such as self-healing concrete that can reduce concrete costs and maintenance needs.

In this respect, as mentioned by several authors, a sustainable efficient process of self-healing can be able to sense damage and cracks by itself first and then free the healing agent, with the least of external inputs (Silva et al. 2015; Vijay et al. 2017). Hence, the first comparison of sustainability of the available self-healing technologies should be done based on determining which method requires the least effort while providing a better outcome.

In this set of ideas, the first option for self-healing concrete is its autogenous self-healing capacity. It is based on the natural capacity of concrete to heal itself by 1) swelling of the cement matrix, 2) hydration, 3) production of calcium carbonate (CaCO₃) through the reaction of calcium ions (ca^{2+}) with carbonate (CO_3^{2-}) from the surrounding environment, and finally, 4) physical clogging of small particles into the surface of the crack. However, this autogenous method is unreliable since concrete composition is highly variable, being that certain compositions favor the healing while others do not. This is true especially with variable water contents in concrete, where it has been seen that more water in the mixture promotes autogenous healing. Additionally, autogenous healing occurs only in very tiny fractures (around 0.05 mm of width) and is very difficult to determine, control, and assess, particularly considering the wide variety of potential cracks and stresses to which concrete may be exposed (Sangadji 2017). Therefore, even if this method is economically viable (no additional investment), naturally, it will not provide good effectiveness, and additional reparations and maintenance works would be required, which is not desirable.

Table.1 B	Bacterial species	s used for self-heal	ng in concrete (modified from A	Ayobami	Adebola et al. 20	20)
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Bacteria types	Cement replacement	Significance	Healed crack width (at 28 days)	References
Bacillus pasteurii	Rice husk ash and dust	Compressive strength	_	Ameri et al. (2019)
Sporosarcina pasteurii	Fly ash	Strength	_	Achal et al. (2011)
Bacillus subtilis	_	Compressive strength	0.90 mm	Mondal and Ghosh (2018)
Bacillus sphaericus	Microcapsules	Durability	0.97 mm	Wang et al. (2014a, b)
Bacilus cohnii	Organic mineral com- pounds	Compressive strength	0.79 mm	Zhang et al. (2017)
Bacillus pasteurii	Lightweight aggregate	Durability	0.50 mm	Chen et al. (2019)
Bacillus sphaericus	Biochar	Compressive strength	0.70 mm	Gupta et al. (2018)
Bacillus mucilaginous	Brewers yeast	Strength	0.50 mm	Chen et al. (2016)
Bacillus halmapalus	_	Compressive strength	0.40-0.60 mm (56 days)	Palin et al. (2017)

In contrast, there is the option of adding a healing agent (either chemical or biological) that flows out when there is a breakage or cracking process (Joshi et al. 2017; Vijay et al. 2017). Firstly, even if chemical addition has proven good results, it is relevant to outline that their healing activity does not work equally in all concretes, and sometimes they can influence the workability and mechanical properties of the mixture, hence, promoting crack formation and the propagation of cracks (Seifan et al. 2016). Concerning bacterial addition, direct application of the bacteria by curing the concrete has shown less effectiveness compared to micro-encapsulation of the bacteria, since there is a maximum depth of healing of 27.2 mm for the first approach as to the healing of 35 mm of the second one. In this set of ideas, several authors have identified that the most effective and sustainable way to produce self-healing concrete is through this bacterial encapsulation, in particular, due to the protection mechanism and uniform characteristics of bacteria in the alkaline environment of concrete (Muhammad et al. 2016; Stanaszek-Tomal 2020; Vijay et al. 2017).

Secondly, a sustainable option should consider the costs of implementation. Today, bio-based additives with encapsulated spores to be mixed in the production process cost 5670 Euro/m³, mainly due to the requirement of aseptic conditions to produce them and their expensive encapsulation process. This results in a price of 30 to 50 Euro/kg of spores, which significantly contributes to the total price of the final product (Silva et al. 2015). Therefore, since prices are currently high enough to make the process profitable or cost-effective, it is required to develop more research based upon mixed bacterial cultures and industrial processing of the material which could lead to potential theoretical reductions of costs of the encapsulated spores to 714 Euro/m³ or 15 Euro/kg of spores (Silva et al. 2015). On the other hand, research on clay encapsulation claims results of 130 Euro/ m³ of concrete, however, it could compromise the material's strength (Hartsock 2011).

Thirdly, to be sustainable, the process itself should have the least possible environmental impacts. As it was mentioned before, the microbial hydrolysis of urea to create the calcium carbonate, that seals the cracks, also produces ammonium. This is reported by several authors that the process can produce nitric acid which could harm the concrete, and possibly add excessive amounts of nitrogen oxide to the atmosphere (Guo and Chidiac 2019; Joshi et al. 2017; Magaji and Yakubu 2019). It is observed that 1m² of concrete requires a 10 g/l of urea for remediation which generates 4.7 g of potentially harmful nitrogen. This quantity is equivalent to one-third of nitrogen that each human produces on a regular basis (Seifan et al. 2016). In this sense, a most sustainable solution for self-healing concrete is to better use calcium lactate instead of urea as a carbon source, which also demonstrates positive and related effects in CaCO₃

production (Jonkers et al. 2010; Joshi et al. 2017; Sangadji 2017; Vijay et al. 2017).

Finally, as a sustainable option, self-healing concrete should reduce greenhouse gases emissions to the atmosphere. This can be achieved indirectly with lower CO_2 emissions, since cement production alone is estimated to account for 7 percent of the total anthropogenic CO_2 , particularly due to the high temperatures in the production process (around 1500 °C) (Jonkers et al. 2010). In this set of ideas, even though still many of the healing mechanisms in concrete itself are questionable and some results are far from satisfactory, if inevitable cracks could be sealed without needing reparation, concrete structures will serve a longer service life, making it sustainable and reducing CO_2 emissions on maintenance, repair, and production of new material (Sangadji 2017).

In short, the industry should focus on the most effective healing approach to increase the viability of self-healing concrete, for which it should mainly work with encapsulation of microorganisms as a healing method. Besides, it also should consider choosing the most efficient species of bacteria to perform the repairing task, which has been identified by several studies to be the *Bacillus sphaericus* (Jonkers et al. 2010; Joshi et al. 2017; Seifan et al. 2016; Stanaszek-Tomal 2020).

Furthermore, since emissions should be avoided, reducing ammonia emissions by changing the hydrolysis of urea into calcium lactate should be considered in order to increase the viability of the concrete. Secondly, it is a fact that promoting the usage of self-healing concrete will reduce CO₂ emissions, however, when it is not possible to only apply this technology as a solution, governments and agencies should also promote the reduction of CO2 emissions from production, maintenance, and repairing of concrete with sustainable solutions, such as new materials added into the cement production (e.g., recycling of waste), recycle of demolished concrete into new one and enhancement of the durability and self-repair processes of existing structures (Sangadji 2017). This should be done by creating programs in which producers are mandated to extend the warranty of products (and where the crack formation is included) and to innovate in the production processes to achieve a longer life span for the concrete (Silva et al. 2015).

Finally, further research on cheaper methods that could provide lower costs for bacterial healing processes is required to enhance the efficiency of self-healing concrete, since currently, prices of self-healing technology are too high to be considered for large-scale implementation.

Applications, opportunities and barriers

Self-healing techniques prevent passages in fluids from entering the concrete for dissolved particles, acidic gasses, and water as the bacteria form a pervading surface on the cracks. A self-healing mechanism can also sense any damage or cracks in concrete which results in the release of the healing agent that triggers rehabilitation of micro-cracks. This system has been reported to have the capacity to cause over 100% recovery in compressive resilience (Van Tittelboom and De Belie 2013). Besides, it's a pollution-free technique, which is based solely on biological activity.

Self-healing concrete, although it is one of the most promising techniques, is still unable to repair wide cracks or potholes on roads. Currently, the technology can repair cracks of up to 32 mm deep and 0.97 mm wide (Dong et al. 2013; Mostavi et al. 2015; Qian et al. 2015; J. Wang et al. 2012, 2014a, b). In addition, most self-healing systems are currently being tested in laboratory settings, but one fit for all mechanisms is yet to be invented. Therefore, a largescale demonstration is required before implementing this idea commercially. The problem of nutrient media optimization also needs to be addressed. The properties of concrete shrinking, corrosion, and carbonization are still not studied in detail (Vijay et al. 2017). Furthermore, this concept could be promoted among contractors and owners by investigating the precise estimation of service life through a comprehensive understanding of the efficacy and variability of self-healing.

The US alone spends \$ 18–21 billion annually on infrastructure repair and upgradation (Li and Herbert 2012). If self-healing concrete were in use, America could have saved that huge amount of money every year though the initial cost would be higher. Again, despite the benefits of selfhealing concrete universally, some particular environments would benefit from the self-healing technology more than the others. These environments, including areas vulnerable to earthquakes, high in corrosive materials, enable cracks to develop in concrete at a higher rate where frequent repair of structures is not feasible (Hartsoc 2011). Self-healing concrete has the capacity to contribute to the infrastructure crisis of those environments. This technology also has the ability to reduce the costs of daily concrete maintenance to the environment, showing its value as sustainable resources.

Based on the literature, it has been shown that the biological processes are efficient in significantly enhancing the longevity of the concrete structure by effectively fixing cracks and cavities. In contrary to the literature on durability, biological healing materials have shown conflicting results on concrete performance. Some studies reported that the concrete strength decreases up to 35% because of the mixing of bio-healing agents (Achal et al. 2013; Wang et al. 2014a, b).

The technology is still at a nascent stage as the focus is on developing effective materials that can satisfy the critical needs of targeted applications. Several studies have recommended the use of microbial concrete as a cost-effective, environmentally safe alternative, and high-end sealant which eventually increases the durability of construction materials. This approach has been proven successful so far, however, still, some gaps exist that have also been addressed in many research findings. One of the major drawbacks is nitrogen oxide emission into the atmosphere through ammonium ions (NH_4^+) production by ureolytic activity (De Muynck et al. 2010). Also, the presence of high amounts of ammonium converted to nitric acid in the concrete matrix increases the risk of damage to salts. And no optimization has yet been found to avoid excessive emission. Besides, excessive bacterial growth in the surface can cause unregulated production of biofilm and uneven surfaces (Ghaz-Jahanian et al. 2013).

There is still doubt on how far the healing process can take place in the concrete structures. However, there is still a chance to contemplate the process if unavoidable cracks due to fragility of underlying concrete could be self-healed/ sealed/repaired. As a result, the concrete structures will certainly provide a long service life by making it more durable and sustainable.

Conclusion

Self-healing capability of the concrete structure is the demand from the construction sector at present and will be the transforming factor for the future. Mechanisms of self-healing, i.e., autogenous, mineral admixture-based, selfhealing based on bacteria, and adhesive-based self-healing and their sustainability have been reviewed in this study. Researches on self-healing concrete show promising results but in laboratory settings. Despite positive results, the technology is still limited for the healing of micro-cracks in the concrete and cannot heal big cracks (wider than 1 mm). The biggest barrier for this technology is economic feasibility as lab processes and associated costs substantially increase the price of this most promising technique. Moreover, this technology might require additional reparations and maintenance which is undesirable. We, as researchers, believe that there is a huge opportunity for self-healing concrete to get extra money for 12 billion tons annually in addition to increased sustainability of concrete structures and decreased carbon footprint of the construction industry. The technology is being demanded by the construction industry and especially in developing countries where the concrete structures are poorly managed. There is a demand of further research and cracking the economic barrier sooner so this novel technology can become common and all concrete structures in the

future become more sustainable, maintenance-free, and safer than in present.

Acknowledgements The authors did not receive any financial support from any authority. The authors would like to thank Ravi Kumar for reading the manuscript.

Authors' contributions All authors contributed equally to the article and approved the submitted version.

Funding The authors did not receive any financial support from any authority.

Availability of data and material The data used in this study are available on request from the corresponding author.

Declarations

Conflict of interest The authors declare that there is no conflict of interest.

Consent for publication The authors have full consent for publication.

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