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Self-sealing cementitious materials by utilizing superabsorbent polymers

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A B S T R A C T

Currently scientific research is searching for technological solutions that provide highly reliable building materials. Incorporating self-sealing agents into the design phase makes concrete more eco-friendly by reducing the maintenance processes and extending concrete's lifespan which decreasingthe need to produce cement and emits a lot of CO_2. Consequently, this investigation describes an attempt to assess the performance of superabsorbent polymer (SAP) as a self-sealing agent. During the experimental work two different particle sizes were taken into consideration SAP_1 (425–600µm) and SAP_2 (> 600 µm), the ratios of SAP were 0.3%and 0.4% wt. to cementn respectively. Regular tests were performed for the pre-crack phase to generate the initial crack which started after 28 days. Two series of sealing periods wet-dry cycles ,Cy4th (8 days) and Cy10th (20 days), were taken to promote the sealing process. The DZM (Digital Zoom Microscope) system was used to detect the self-sealing phenomenon and the width of the sealing on the concrete cracks' surface through the use of special software imaging processes. The outcomes demonstrated the positive effect of using SAP as a sealing agent as SAP_2 was able to heal the crack up to 0.15mm depending on the particle's size. Also, the percentage of partially at Cy4th was 64% compared to the other mixes.

1. Introduction

Concrete has been regarded as a reliable building material for many centuries. However, concrete is vulnerable to cracking when tensile stresses are generated inside of it. The proper repair of cracks has become a subject of worry since these fissures allow harmful substances to infiltrate deeper matrix layers, compromising the durability and ultimately the mechanical performance [1]. Therefore, continuous mixtures of concrete must be developed continuously to meet the demand for more resilient and effective building materials [2]. The ability of cementitious composites to seal and repair cracks by autogenous sealing is primarily caused by the continuing hydration of unhydrated cement particles and the precipitation of $CaCO_3$. The mechanism of rehydration continues to hydrate until all of the unhydrated cement has been consumed. Water must be present inside the fractures for these processes to start and, ultimately, for autogenous sealing to occur [3]. Therefore, superabsorbent polymers have been investigated as a way to retain the humidity in the cracks. SAP has gained popularity as a way to improve cementitious materials' capacity for reducing the price of traditional repair methodical repairs ways [4]. Broadly in the literature, SAPs are hydrogel networks made of cross-linked water-soluble polymers. One of the most significant aspects is the SAP's swelling capacity. Watery solutions can be absorbed by SAP, which can hold for hundreds of times their weight [5]. Polymers take up water from the surrounding environment during the wet period. At this time, it is working like containers for water. When it's dry, they slowly release it and accelerate the process of autogenous healing [6]. The carbon dioxide from the air dissolves in the water produced by the SAP, and calcium carbonate begins to form [7]. As a result, the crack entry is physically blocked, resulting in a self-sealing [8]. It should be noted that the insertion of SAP increases the macro porosity, which has a detrimental impact on mechanical performance despite their advantage for the stimulation of self-healing [9]. Thus, the ratio of SAP that is added to cementitious mixtures should therefore be carefully considered [10]. advantage for the stimulation of self-healing [9]. Thus, the ratio of SAP that is added to cementitious mixtures should therefore be carefully considered [10]. Researchers have extensively demonstrated the use of various agents such as superabsorbent polymers as a sealing and sealing agent. Gruyaert et al. [11] added the in-house-made SAP < 400 µm with (0.5% and 1%) content to cement.

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Nomenclature:

C.A	Compressive strength of concrete Mpa	Cy	Wet-dry cycles
w/ca	Additional water/ cement	OPC	Ordinary Portland Cement
SAP	Superabsorbent polymer	SSD	Saturated surface dry
DZM	Digital Zoom Microscope	UTM	Universal testing machine

Their system demonstrated an increase in flexural strength of 8% and 6%, respectively. Van Tittelboom et al. [12] described another survey that incorporated polyurethane (PU) and superabsorbent polymers. Van Tittelboom et al. [13] described another study that involved glass tubes for polyurethane (PU) and SAP < 600 μm with 1% by weight of cement to mixtures, separately. The compressive strength of the polymer was reduced by 18% after 28 days compared to control mixes, while polyurethane had no effect. To check the sealing ability, the cracks were developed by a four-point bending test; the processing of crack width was from 50 to 250 μm. These cracks healed due to the swelling characteristics of SAP and water release that continued hydration and produced $CaCO_3$ [14]. Despite the extensive studies on the study self-sealing phenomenon by using different agents of healing like SAP polymer, there is no specific research to study the states of specimens in pre-created (generated initial crack as demonstrated in the methodology of work) by using different particle sizes of SAP on the sealing phenomenon with and without polymer. On the lab scale, one of the evaluation techniques employed to assess the efficiency of self-sealing is microscopic analysis. Therefore, this paper describes an attempt to create self-sealing concrete by using superabsorbent polymers as a self-sealing agent. Digital Zoom Microscope (DZM) of SAP-based concrete is scanned and compared with traditional concrete (without a sealing agent).

2. Experimental plan

2.1. Material

On the laboratory scale, three different concrete mixtures were made to compare the results of self-sealing concrete. The properties of materials used in preparing mixtures are shown in Table 1 the essential compounds of mixtures were:

2.1.1. Cement

Ordinary Portland Cement (OPC) made the mixture with grade 53 and a 3.15 g/cm3 density.

2.1.2. Coarse Aggregate

The locally accessible crushed material was employed with a nominal maximum diameter of 10mm and IS examined a specific gravity of 2.65: 383-1970 specification [15], in saturated surface dry (SSD) conditions.

2.1.3. Fine aggregate

In the current experiment, the locally accessible river sand was utilized as fine aggregate which was examined following IS: 383-1970 instructions and was in zone II with a specific gravity of 2.54 and fineness coefficient of sand 2.6.

2.1.4. Superabsorbent polymer (SAP)

In the laboratory work, SAP has a high absorption capacity and can swell significantly to produce a pliable, insoluble gel as shown in Fig.1. Table 2 demonstrates the characteristics of the superabsorbent polymer (SAP) employed in the experimental work in this study. SAP particles exhibit less swelling when combined with fresh concrete because their ability to expand is strongly dependent on the solution's ionic content and alkalinity [17]. So, the tea bag method was conducted to quantify the water absorption by the SAP within the concrete as shown in Fig. 2.

This method was described by RILEM technical committee [18]. The results were 23 g of water per gram of dry SAP particle size (425-600) μm and 25 g of water per gram of dry SAP particle size >600 μm. Once the cement has hydrated, the SAP will release the water it has absorbed, contract, and leave behind tiny macropores [19]. Cracks are more likely to spread through the pores when they first appear, and moisture that enters via the crack causes the SAP to re-swell [20]. In times of dryness, SAP re-releases its water content, promoting autogenous repair. The mechanism of autogenous sealing is the production of calcium carbonate ($CaCO_3$) or calcium hydroxide Ca $(OH)_2$ through hydration of unhydrated cementitious particles and calcium-silicate hydrate (CSH) gel swelling or expansion [21]. The mechanism of SAP sealing is the production of calcium carbonate ($CaCO_3$) or calcium hydroxide $Ca(OH)_2$ through hydration of unhydrated cementitious particles and calcium-silicate hydrate (CSH) gel swelling or expansion [22]. Fig.3 shows the mechanism of SAP sealing on the outer faces of the concrete.

Table 1. Properties of materials That was used in preparing mixtures at 75% humidity at 22 C^0

Property	**Cement**	**Sand (Zone II)**	**Gravel**
Specific gravity	3.15	2.54	2.65
Water absorption %	-	0.80%	1.50%
Size of aggregate (mm)	-	> 4.75	10 *mm*

Figure 1. Superabsorbent polymer (SAP) in a dry and wet state

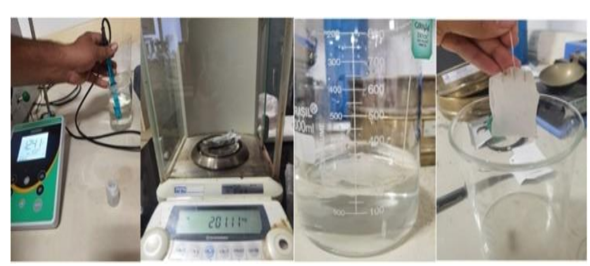


Figure 2. Method of conducting the water absorption by the SAP

2.2. Methodology

One of the key objectives of this investigation is to consideration of a rapid (accelerated) self-sealing mechanism by using the SAP agent to reduce damages caused by the pre-cracking phenomenon. There is no specific standard method investigated in previous studies to elucidate the behaviour of SAP concrete. This section shows the steps of experimental work performed throughout this investigation.

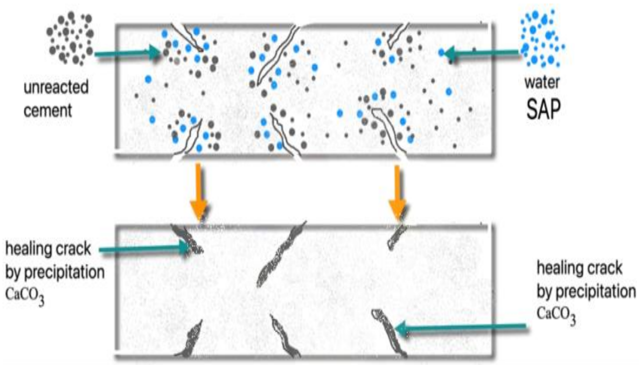


Figure 3. The mechanism of SAP within the concrete

Table 1. Properties of SAP

Items		SAP_1	SAP_2
Particle size (mic)		425-600	>600
Bulk density		0.8	0.8
Mean absorption of SAP(g)	In distilled water	177.5	263
	In cement solution	23	25

2.2.1. Samples preparation

In batching concrete, the quantity of cement, fine aggregate(sand), coarse aggregate(gravel), and SAP is determined by mass as shown in Table 3 to obtain a concrete composition for sealing assessment. The Indian standard code IS:10262 (2009) has been used to select suitable ingredients for the concrete design [23].

The blending process commences with the dry mixing of composites of concrete (cement, sand, gravel, and SAP) for at least 3 minutes. After that, the wet mixing was initiated by adding water to obtain a homogeneous mixture for the moulding process, which was done by compacting the mould with 25 manual blows per layer (three for each mould) as shown in Fig 4. To prevent the loss of wetness from the concrete, the curing process began 24 hours after the casting process. The curing period was 28 days for all specimens under lab conditions, 75% humidity and at 22°C.

2.2.2. Pre-cracked phase (generated initial crack)

After a 28-day curing time, the second set of samples was loaded at a steady rate of 0.2 mm/min by using a universal testing machine (UTM). As shown in Fig 5, four interconnected microlenses were applied to simulate pre-cracking, observe the initial cracks, and control the crack mouth opening to obtain the width of the crack less than 0.5mm for a faster sealing process. Prince et al. [24] found that Initial cracks can be obtained by loading specimens to 75% of the ultimate strength, using compressive strength and bending test (flexure).

2.2.3. Promote the sealing process

After pre-cracking, three sets of samples were subjected to 10 cycles of wetting and drying mode (one cycle: in water for 24 h and in the air for another 24 h) under the lab conditions of 75% humidity and at 22°C. to enhance the sealing process of the fractures at the concrete interface as shown in Fig 6.

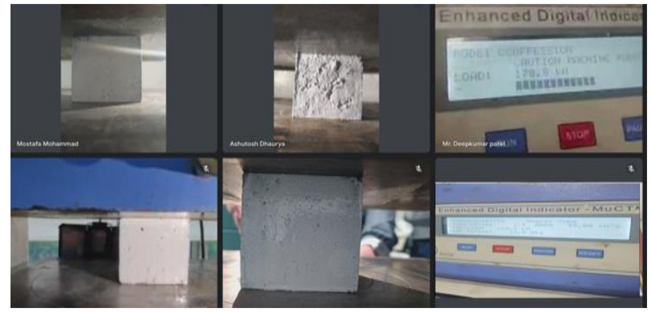


Figure 4. Samples preparation

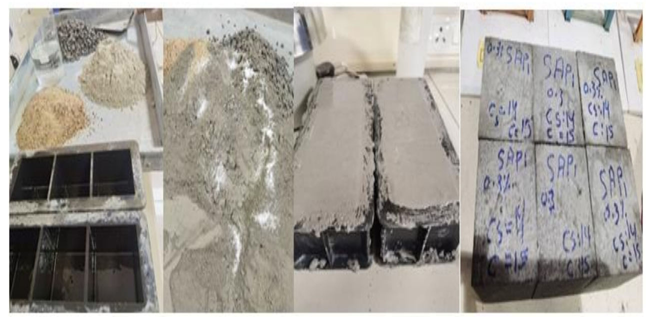


Figure 5. Generation initial crack in the sample

2.2.3. Promote the sealing process

After pre-cracking, three sets of samples were subjected to 10 cycles of wetting and drying mode (one cycle: in water for 24 h and in the air for another 24 h) under the lab conditions of 75% humidity and at 22°C. to enhance the sealing process of the fractures at the concrete interface as shown in Fig 6.

Table 3. Mix proportion of cement concrete. Material Consumption of Cubic Concrete (Kg/m^3)

Item	w/c	Cement	Sand	Gravel	water	w/c_a g/g
R	0.4	550	706	794	238	0
SAP_1	0.4	550	706	794	238	23
SAP_2	0.4	550	706	794	238	25

Concrete grade (MPa)= M25, dimensions of the cubes: (70*70) mm and curing age: 28 days, $w/c_{a:}$ additional water

2.2.4. Evaluation sealing process

Estimating the sealing process was done by measuring the width of the cracks before and after the periods of wet and dry cycles by using a digital zoom microscope as shown in Fig 7. On the lab scale, one of the evaluation techniques that has been employed to assess the efficiency of self-sealing is microscopic analysis due to the difficulty of using visual observation to estimate the performance of superabsorbent polymer as a self-sealing agent. The DZM are characterized as macro zoom lens systems with two lenses that offer two distinct viewing angles to see the reflected light, producing a three-dimensional (3D) image of the material. Applications of DZM include monitoring purposes for material that requires detailed studies of material surfaces for larger samples.

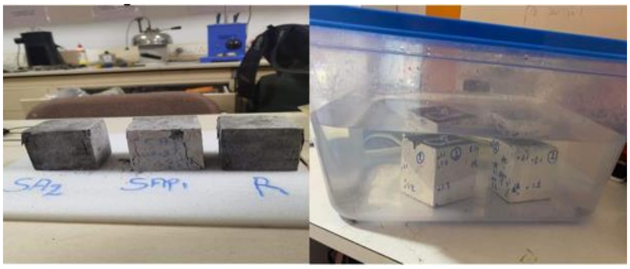


Figure 6. Promote the sealing process

DZM uses specialist software (image process) to select crack size measurements of the reference concrete and SAP concrete. In addition, the DZM system can help detect the self-sealing phenomenon on the concrete cracks' surface by taking images of the agents formed through the cracks as shown in Fig 8.

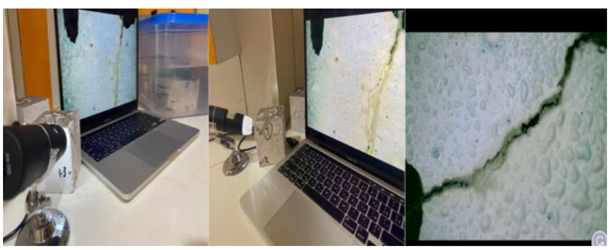


Figure 7. Evaluation sealing process by using a digital zoom microscope

3. Results and disccutions

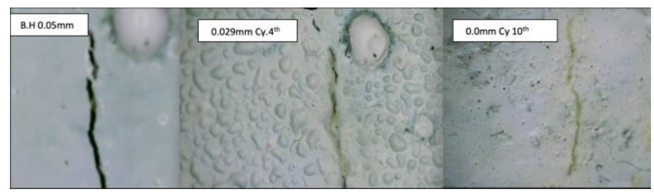
DZM system was carried out immediately after the phase of generating cracks in the samples at some particular dote on the cracks. Fig 9 displays images of the width of sealing cracks in the Mix1 SAP_1, Mix2 SAP_2 and Mix3 R for Cycles 0 (0 days), 4th (8 days), and 10th (20 days). Results as shown in Table 4, the average crack width in SAP_1 reduced from 0.05mm (actual width) to 0.029mm at Cy4th to 0.0mm at Cy10th completely sealing due to the precipitation of calcium carbonate ($CaCo_3$). The SAP_1 healed the crack up to 0.03mm and the percentage of partial sealing at Cy4th was 42%. For SAP_2 the average width of the cracks reduced from 0. 19mm (actual width) to 0.068 mm at Cy4th to 0.0mm at Cy10th through the processing of healing. The SAP_2 healed the crack up to 0.15mm and the percentage of partial sealing at Cy4th was 64%. For R. The average width of the cracks reduced from 0. 01mm (actual width) to 0.0073 mm at Cy4th to 0.005 mm at Cy10th through the processing of healing. The R. healed the crack less than 0.03mm and the percentage of partial sealing at Cy4th was 27%. It can absorb the crystal formation generated in SAP concrete and reference concrete but with different efficiency.

Table 4. Self-sealing capacity in various wet-dry cycles

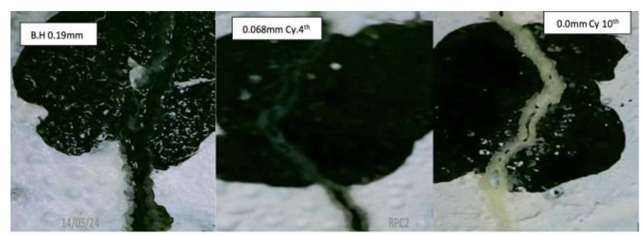
Items	Width of sealing cracks (mm)			Observations
	Cy0th	Cy4th	Cy10th	Partial sealing at Cy4th
R	0. 01	0.0073	0.005	27%
SAP_1	0.05	0.0290	0.000	42%
SAP_2	0. 19	0.0680	0.000	64%

Cy0th (0 days): Cy4th (8 days): Cy10th (20 days)

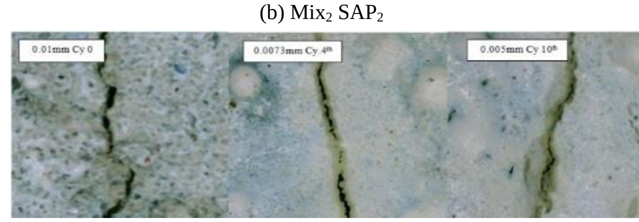
In conventional concrete, self-sealing can occur under abroach hydration of unhydrated cement particles but within a limited range of crack width. As a result, SAP_2 shows better self-sealing capacity compared to the rest of the mixtures because the particle size of SAP_2 is larger than that of SAP_1, and the amount of water uptake by these particles is more efficient to generating more sealing agents inside the crack to complete the hydration prosses within the concrete than smaller particle size SAP_1. The SAP_2 significantly improved the sealing ability of the cracks. Similar to most of the preceding research examined the influence of using different SAP particle sizes [25]. Mousavi et al. [26] reported that a large particle SAP size is more efficacious than a smaller particle SAP size in enhancing sealing efficiency. Gruyaert et al. [11] described a comparative study of self-synthesised SAP (400–600 and > 600 μm) size and with commercial SAP (acrylamide and acrylate cross-linked copolymers). To maintain the workability of the mortar mix, supplementary water was added to the mixes due to the swelling property of SAP; it was 20 g water per g of (SAP self-synthesized) and 17 g water per g of (SAP commercial). The results indicated that SAP with sizes of >600 μm were most effective in $CaCO_3$ precipitation and filling cracks for sealing efficacy.



(a) Mix_1 SAP_1



(b) Mix_2 SAP_2



(c) Mix3 R.C.

Figure 8. images of sealing cracks in the Mix1 SAP_1, Mix2 SAP_2 and Mix3 R.C for Cycles 0(0 days), 4^{th} (8 days), and 10^{th} (20 days)

4. Conclusions

This study outlined the use of a super-absorbent polymer (SAP) to accelerate the production of self-sealing concrete. The self-sealing technique aim to close the crack thus preventing water from penetrating into the concrete and to gain a more durable and sustainable material. The composition of the concrete, the chemistry of the sealing agent, and the number of wet-dry cycles through the sealing period are critical parameters for the efficiency of sealing cracks. Thus, from the experimental program the following concluding remarks were found:

- Many SAP parameters affect the sealing process such as particle size. Alarge particle's SAP_2 (> 600 μm) size is more effective than a smaller particle's SAP_1 (425–600 μm) size in enhancing sealing efficiency. In this investigation, the SAP_1 healed the crack up to 0.03mm and the percentage of partially sealing at Cy4th was 42% while the SAP_2 healed the crack up to 0.15mm and the percentage of partially sealing at Cy4th was 64%. Cy4th was taken as a point of comparison.

- In general, the addition of SAP terminates the hydration process in cracks generated in the hardened concrete. So, SAP_2(> 600 µm) is shown to have a higher capacity as a sealing agent inside cracks than SAP_1(425–600 µm) smaller particles because the presence of water through the crack is considered an essential factor in completing the chemical reaction. Also, the large particle size is more effective in completing the hydration process within the concrete.
- In conventional concrete self-sealing can occur when dehydrated cement particles are hydrated, within a certain fracture width range. Therefore, this type of concrete healed the crack less than 0.03mm and the percentage of partial sealing at Cy4th was 27%.
- One of the most difficult aspects of designing the SAP-based concrete mix is supplying the proper dosage of self-sealing agent to the specific area where cracking is most likely to develop.

Authors' contribution

All authors contributed equally to the preparation of this article.

Declaration of competing interest

The authors declare no conflicts of interest.

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Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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