



Durability of mortar and concrete containing pozzolans as a partial cement replacement in the marine environment: a review

Lanh Si Ho^{1,*}, Huong Thi Thanh Ngo¹, Hieu Trung Vu¹, Sang Thanh Nguyen², Vinh Ngoc Chau³, Viet Quoc Dang⁴

¹University of Transport Technology, Hanoi, Vietnam

²University of Transport and Communications, Hanoi, Viet Nam

³Department of Civil and Environmental Engineering, University of Michigan, Ann Arbor, MI 48109, USA

⁴Faculty of Bridge and Road Construction, Mientrung University of Civil Engineering (MUCE), Phu Yen Province, Viet Nam

Article info

Type of article:

Review paper

DOI:

<https://doi.org/10.58845/jstt.utt.2023.en.3.3.13-25>

*Corresponding author:

E-mail address:

lanhhs@utt.edu.vn

Received: 14/07/2023

Revised: 22/09/2023

Accepted: 25/09/2023

Abstract: This paper reviews the durability of concrete containing pozzolans in marine environments, which mainly focuses on resistance to chloride ion penetration and sulfate attack. The results of previous literature showed that the main ions of seawater are almost similar but the salinity varies with location. The mechanism of chemical reaction between seawater and concrete was carefully reviewed and explained. Besides, the main attacks that caused the degradation of concrete structures in marine conditions were summarized. The results of previous research indicated that using pozzolans (fly ash, slag, metakaolin, rice husk ash, etc.) as a partial cement replacement in concrete improved the durability of concrete such as resistance to chloride ion penetration and resistance to sulfate. The use of pozzolans not only improves the durability of concrete in the marine environment but also solves environmental impacts such as reducing CO₂ emissions and landfilling areas. It can be concluded that the utilization of pozzolans in concrete, particularly in marine concrete structures is a promising approach for sustainable development.

Keywords: Sulfate resistance, marine environment, chloride penetration, durability, pozzolans.

1. Introduction

The concrete structures constructed in marine conditions generally have a shorter service life in comparison with conventional concrete due to the deterioration caused by seawater attacks [1]. The corrosion of reinforcement caused by chloride-induced is known as a first deterioration issue [2,3]. To resist the corrosion of reinforcement, different types of reinforcement such as fiber reinforcement, stainless, and carbon steel have been used [4,5].

However, these reinforcements are still difficult to wide application due to the high cost of production. Indeed, the service life and durability of concrete are strongly governed by many factors such as the selection of material mixture, and quality control during the construction process on-site. The durability of concrete is known as the resistance against degradation, which is related to chloride penetration, sulfate attack, and other attacks. From those perspectives, it is needed to find alternative

solutions to improve the resistance to harmful attacks. One of a potential approach is to use supplementary cementitious materials (SCMs or pozzolans or by-products) to partially replace cement in mortar and concrete.

Over decades, many by-products such as fly ash (FA), slag, rice husk ash (RHA), and other agricultural ashes have been employed to replace cement in mortar and concrete at different levels [6–10]. It is known that these SCMs have an important role when suitable cement content is substituted by these waste materials. In general, SCMs in mortar and concrete mitigate the permeability of mortar and concrete because SCMs refine the pore system and make denser structures of concrete, which results in an improvement of resistance to corrosion, and aggressive attacks [11]. The durability of concrete is generally improved when SCMs are included, also it was reported that the inclusion of SCMs in mortar and concrete mitigates environmental impacts and reduces the cost of concrete production [12]. Another research indicated that the replacement of cement with 15% FA and 25% slag could significantly improve the mechanical

properties of concrete over a long-term period [13]. The incorporation of pulverized fuel ash with the OPC helped to improve mechanical properties and resistance to permeability and chloride ion penetration of concrete [14].

Based on the above information, this research will systematically examine and evaluate the durability of concrete containing pozzolan materials in previously published works. This study will mainly focus on the durability of concrete comprising pozzolans in terms of carbonation, chloride ion penetration, and sulfate attack. Besides, in this paper, the mechanism of chemical reaction between concrete and seawater will be carefully reviewed. Furthermore, the potential attacks of concrete in marine environments will also be assessed. This paper will contribute more to the understanding of the durability of concrete in marine environments under aggressive attacks (chloride and sulfate attacks). Finally, this paper will provide a systematic knowledge of the deterioration of concrete in the marine environment.

2. Component of seawater and main attacks

2.1. Main chemical components of seawater

Table 1. The content of major ions included in the seawater

Concentration (mg/l)	Cl ⁻	Na ⁺	Mg ²⁺	SO ₄ ²⁻	Ca ²⁺	K ⁺	CO ₃ ²⁻
UAE [16]	26000	15000	2300	3699	500	520	-
Red Sea [17]	22660	11350	1867	3051	531	1350	150
Barcelona [18]	20800	11640	1360	2820	490	420	-
Melbourne [19]	20700	11940	1430	3420	-	622	-
Yellow Sea [20]	19630	10780	1297	2700	408	388	140
Atlantic [21]	19000	10500	1350	2670	400	380	140
Yokosuka [22]	17087	9920	1167	2379	356	346	110
North Sea [23]	16550	12200	1110	2220	430	500	-
Black Sea [24]	9500	4900	640	1362	236	230	215
Baltic Sea [25]	3000	1800	240	1230	98	67	-
Vietnam Sea [26]	12201-16590	4048-7331	331-486	1412-1873	158-259	129-203	-
ASTM D1141-98 [15]	19845	11024	1314	2766	418	397	145

Note: “-“ means not applicable

The major ions of seawater from different locations are shown in **Table 1**. From Table 1, it can be observed that the salinity varies and strongly depends on the location of each area (i.e.

climate and geography conditions). For example, from Table 1, it can be observed that the salinity of the Baltic Sea is very low due to the temperature climate. Besides, in the case of Vietnam, it can be

observed that the salinity of seawater in the Vietnam region is much higher than that of the Baltic Sea due to the tropical climate. **Table 1** also shows the result of major ions in seawater specified by ASTM D1148, which can be used to prepare artificial seawater [15].

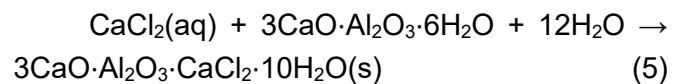
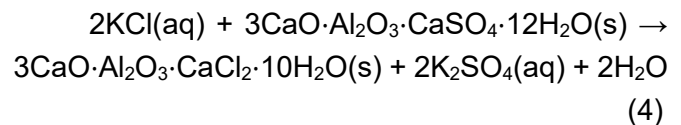
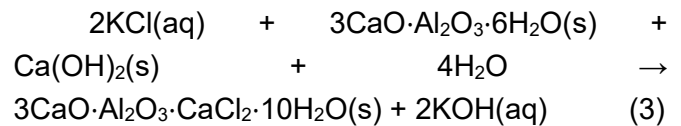
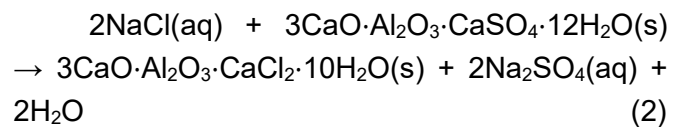
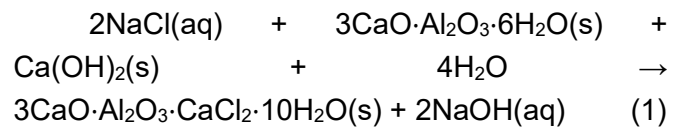
2.2. Chemical interaction mechanism of concrete in seawater

Concrete is a porous material that can absorb seawater when exposed to the marine environment. This means that various aggressive ions have the ability to penetrate the concrete pore structure and chemically interact with its hydration products. The durability of concrete in a marine environment is influenced by the interplay of alterations in its physical properties and chemical reactions [27]. It is widely acknowledged that the degradation of concrete structures in a marine setting is primarily governed by the permeability of the concrete. This permeability is often associated with various marine exposure conditions and chemical reactions. For a more comprehensive understanding of these interactions, readers can refer to a review article by Yi et al. [28]. To summarize these mechanisms, three main ions in seawater - chloride, magnesium, and sulfate - are of particular concern in this context.

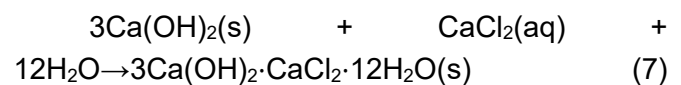
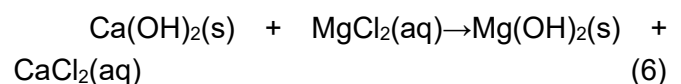
As demonstrated in **Table 1**, the chloride ions found in the seawater environment primarily originate from sodium chloride (NaCl), magnesium chloride (MgCl₂), calcium chloride (CaCl₂), and potassium chloride (KCl).

The chloride ion, which has the highest concentration among other main ions in seawater, tends to absorb in calcium silicate hydrate CSH or promote the formation of Friedel's salt (3CaO·Al₂O₃·CaCl₂·10H₂O) and Kuzel's salt (Ca₄Al₂(OH)₁₂Cl(SO₄)_{0.5}·5H₂O) via ion exchange between C₃A and monosulfaluminate and the free chloride ion, see Equations (1-5) [29]. This product is generated in the inner layer of concrete rather than the surface layer because the stability of Friedel salt depends on the pH as well as the presence of magnesium and sulfate ions [28,29].

Friedel's salt serves as a place for chloride to chemically bind in the cement matrix. The problem introduced by Friedel's salt in the concrete structure is that, when the pH of concrete decreases, the dissolution of Friedel's salt increases, releasing free-chloride ions into the pore solution [29]. In this scenario, if a critical concentration of free-chloride is released at the depth where reinforcing steels are located, corrosion of steels may occur.

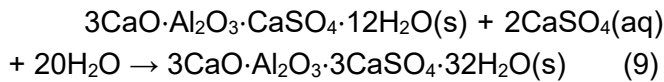
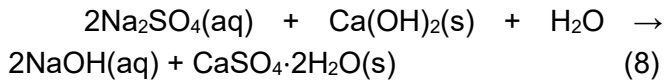


In the meanwhile, magnesium ion in seawater can react with Portlandite in concrete and precipitate as brucite (Mg(OH)₂) [30]. This reaction lowers the pH of the pore solution, giving rise to issues not only related to Friedel's salt dissolution, as mentioned earlier but also destabilizing the calcium-silicate-hydrate (C-S-H) phase [31]. The decalcified C-S-H can partially or completely transform into non-cementitious magnesium-silicate-hydrate (M-S-H) [32,33] which in turn reduces the strength of the concrete structure, see Equations (6-7).



Additionally, the sulfate ions from seawater environments are mainly from Na₂SO₄. The presence of sulfate in seawater can lead to the

formation of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) through a reaction with Portlandite or result in the formation of ettringite ($3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{CaSO}_4 \cdot 32\text{H}_2\text{O}$) by reacting with aluminate phases, see Eqs. (8-9). These chemical reactions have the potential to induce cracking in the concrete [34,35] because both gypsum and ettringite are expansive products.



2.3. Major deterioration of concrete structures in marine environment

The deterioration of concrete due to chemical

and physical attacks in the marine environment is governed and affected by the coupled effects of aggressive ions and environmental conditions. The mechanism of the deterioration is explained by salt crystallization, delayed formation of expansive products, abrasion, the decomposition of cement hydrates products, etc. [36,37]. In addition, the deterioration of concrete can be also affected by the location of the structures. As indicated in the previous study, microclimatic location could also impact the sulfate resistance of concrete. Because the microclimatic location is related to sunshine duration and wind, which strong influences the microclimatic conditions of sulfate [36].

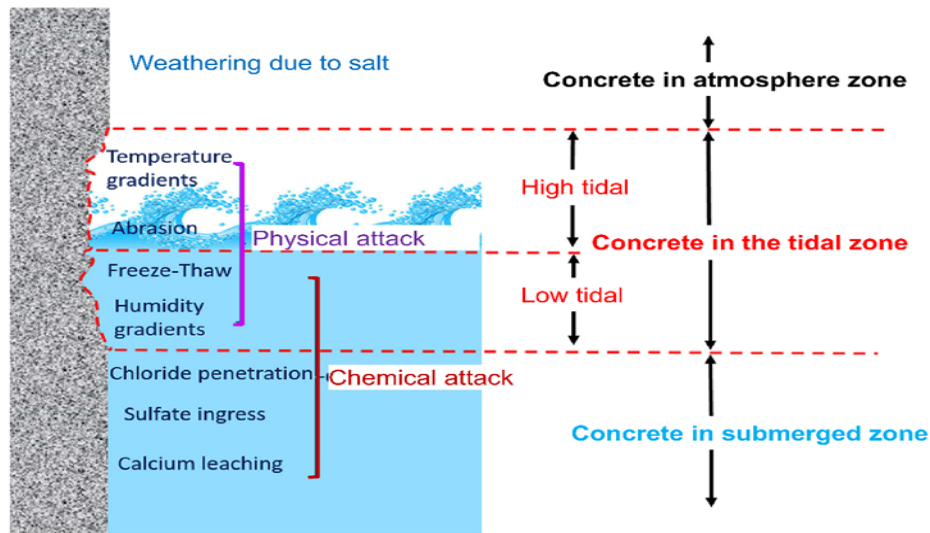


Fig. 1. The chemical and physical attacks of concrete in the marine environment modified from [36].

Fig. 1 shows the major attack of concrete in the marine environment. From Figure 1, it can be observed that the deterioration of the tidal zone is not only related to chemical attacks but also involves physical effects such as the wetting and drying process and the wave attack [36]. However, in the case of the atmosphere and submerged zone, the degradation is only affected by salt weathering and chemical attack. This is a fact that the voids and cracks of concrete can be filled by the products of chemical reactions (sulfate, carbonation, and magnesium ions), which can further reduce the penetration of aggressive substances. As a result, the concrete in the submerged zone has less deterioration [36].

In contrast, the products of those chemical reactions (sulfate, carbonation, and magnesium) can be washed out due to the continuous splash of waves, which leads to the inner part of concrete being exposed repeatedly to seawater. Consequently, the concrete will gradually cause the loss of concrete mass. Thus, it can be said that the attack of one factor can promote the attack from other factors, which significantly increases the deterioration degree of concrete structures. These results have been verified by many investigations of concrete structures in marine environments. For example, many authors have confirmed that the wharves structures in the splash zone are the most deteriorated region in terms of corrosion, spalling,

and delamination [38,39]. Another study on mortar samples indicated that specimens taken from the intertidal zone have crack zones, while the samples collected from the area above that zone have no degradation signals [40]. The local microclimatic condition could also affect the deterioration level of structures. The concrete girder and deck located on a higher temperature side showed a higher deterioration degree in comparison with the lower temperature side [39,41]. Because it is known that a higher temperature causes a greater diffusion coefficient as well as a faster chemical reaction.

3. Performance of mortar and concrete containing pozzolans

3.1. The results of chloride penetration

Fig. 2 shows the effect of slag replacement with different water/cement (w/c) ratios on the total charge passed results at 42 days [42]. It can be observed that the total charge passed increased with increasing of W/C ratio for both types of mixture. Besides, the replacement of OPC with 50% GGBS significantly reduced the total charge passed. This significant reduction is caused by the pozzolanic reaction between GGBS and $\text{Ca}(\text{OH})_2$, which resulted in a pore refinement [42]. In other words, the pore size of 50% GGBS mixture was smaller than that of OPC mixture, which led to the substantial reduction of the total charge passed. Thus, it can be said that the replacement of cement by ground granulated blast furnace slag (GGBS) strongly caused a reduction of chloride ion penetration. The longer curing period resulted in a smaller chloride migration coefficient [43]. It can be observed that the lowest chloride migration coefficient for the whole curing period was found for the mixture comprising metakaolin (MK) and GGBS. Besides, the mixtures containing 10% by-products (MK and GGBS) prove a substantial resistance to chloride penetration (**Fig. 3**). The significant decrease of chloride migration coefficient was explained by pore refinement due to the inclusion of GGBS and MK [43].

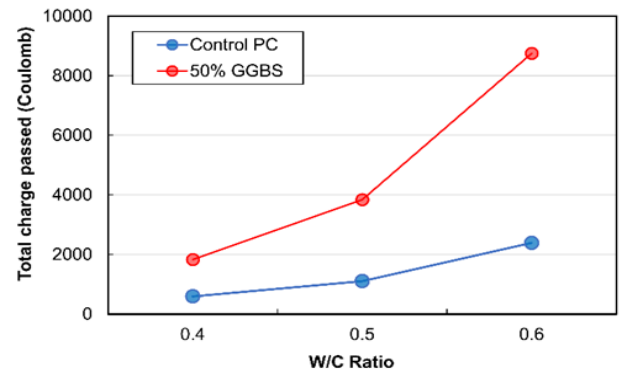


Fig. 2. Influence of GGBS replacement with various w/c ratios on total charge passed at 42 days of water curing [42]

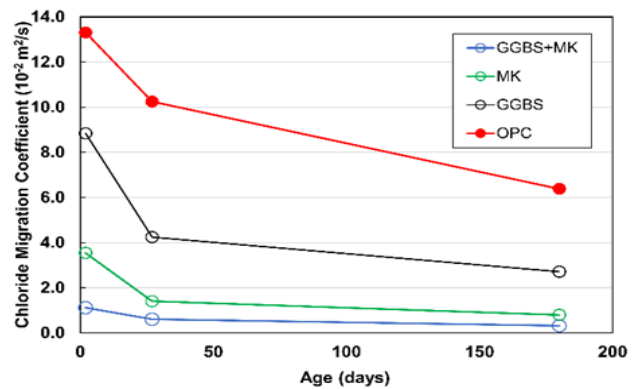


Fig. 3. Influences of GGBS and MK replacement on migration coefficient of chloride ions [43]

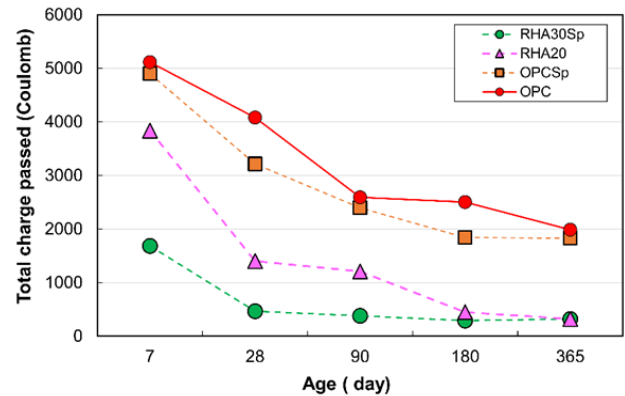


Fig. 4. Comparison of charge passed values (Coulomb) RHA concrete and OPC specimens with and without SP [44]

The charge-passed values of concrete specimens containing rice husk ash (RHA) for up to 365 days are presented in **Fig. 4** [44]. It was observed that the OPC concrete (both with superplasticizer (SP) and without SP) showed higher values of charge-passed compared to concrete containing RHA. From the figure, it can be

observed that the concrete specimens including RHA had a low or moderate chloride penetrability. The low chloride ion penetration observed in the concrete specimens comprising RHA was explained effect of pore refining. Furthermore, the addition of significant content of SP can also reduce the chloride ion penetration of concrete.

It was observed that the concrete containing silica fume (SF) has a lower value of chloride ion permeability compared with concrete OPC concrete and concrete containing fly ash (FA) (Figs. 5-6) [45]. The authors stated that concrete with 10% SF or 20% FA replaced with cement significantly improved the resistance of chloride ion penetration. At the age of 28 days, the concrete containing 20% FA showed a smaller value of chloride penetration in comparison with OPC concrete. At a later age (360 days), the effect of FA

replacement is more significant. This reduction of chloride penetration may be decreased with time and depends on many factors such as the amount of FA replacement, $\text{Ca}(\text{OH})_2$ content, etc. The results found in Figs. 5-6 show that the replacement of OPC by FA is less effective than that of SF. The concrete specimens comprising 10% SF produced a lower chloride penetration value compared to FA concrete specimens for all w/c ratios as well as all levels of FA replacement. The lower value of chloride penetration in the concrete specimens with 10% SF is due to the refining pore system caused by the SF effect. The resistance against chloride penetration of concrete containing FA was improved in the long-term curing period due to the effect of the pozzolanic reaction between FA particles and cement hydrate products.

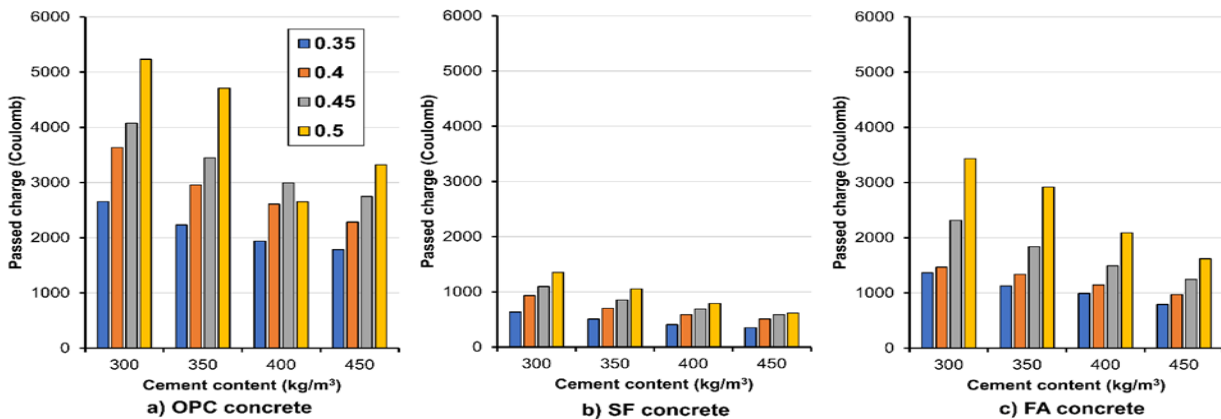


Fig. 5. Values of charge passed (Coulomb) for different concrete and various w/c ratios at 28 days of curing [45]

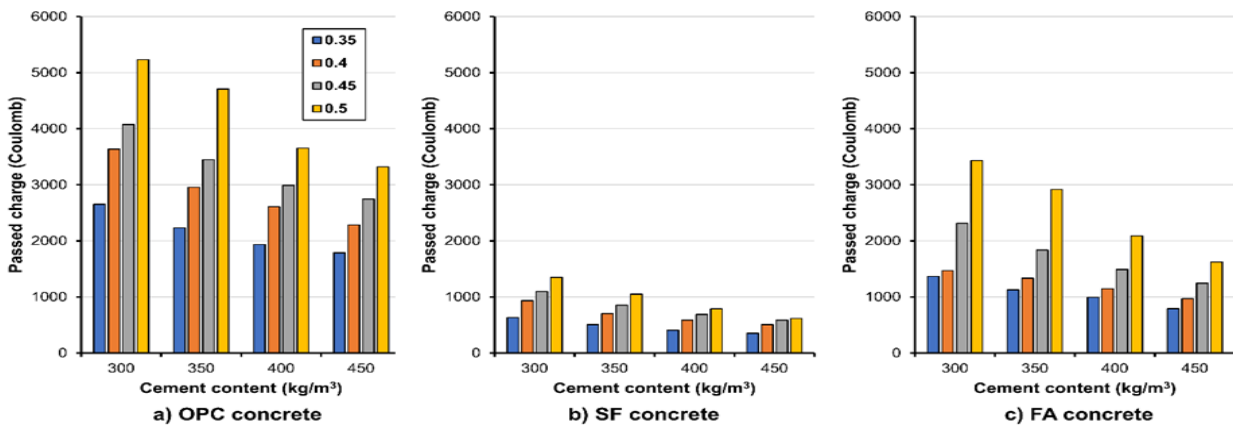


Fig. 6. Values of charge passed (Coulomb) for different concrete and various w/c ratios at 360 days of curing [45]

3.2. Results of sulfate resistance and strength loss

Concrete and mortar containing supplementary cementitious materials (SCMs) (i.e. pozzolans) showed a higher sulfate attack resistance in comparison with plain OPC concrete and mortar as indicated by many previous studies. It was suggested that concrete corrosion is generally expressed by expansion, spalling, and cracking [46]. The length change of the mortar specimens submerged in sulfate solutions is used to assess expansion, spalling, and cracking [46]. The effects of slag and FA length change of mortar samples with a fixed ratio w/c of 0.4 are shown in Fig. 7.

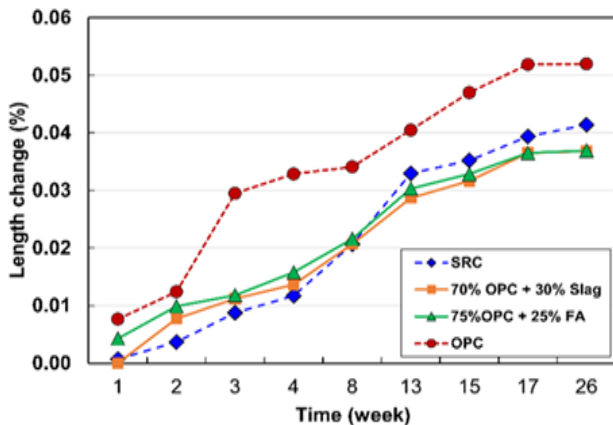


Fig. 7. Impact of SCMs on length change of mortar samples [48]

From Fig. 7, it can be observed that the specimens that used SCMs or sulfate resistance cement (SRC) reduced the length change of species compared with the OPC specimens. Furthermore, for a longer curing period, the specimens containing SCMs showed better sulfate resistance compared to OPC specimens. In another study, the cube mortar specimens with a size of 50 mm were made using a w/c ratio of 0.50. Then, the specimens were stored in Na_2SO_4 solutions with various concentrations for a duration of two years in order to detect deterioration due to physical change [47]. The authors stated that cement type, solution concentration, and immersion period strongly affected the deterioration level [47]. In which, the mixture of

cement type I containing 20% FA exhibited the best performance in terms of sulfate resistance. Another study conducted by Chatveera and Lerwattarak [48] employed black rice husk ash (BRHA) to replace OPC cement at different levels of 0, 10, 30, and 50% by mass of binder. Then, the resistance to sulfate attack was evaluated using expansion and strength loss indexes. The mortar specimens were prepared using two w/c ratios of 0.55 and 0.65. After that, the specimens were cured for 180 days in 5% MgSO_4 and 5% Na_2SO_4 solutions. The results showed that strength loss and expansion of specimens decreased with an increase in BRHA replacement levels. Particularly, the mixture with 30 and 50% BRHA showed a smaller value of expansion in comparison with the mixture using SRC. Interestingly, the expansions of the specimens submerged in Na_2SO_4 were higher than those of the specimens submerged in MgSO_4 . It was reported that the expansion and cracking on the surface were caused by the establishment of ettringite and gypsum, which favor for the decalcification of calcium components. This decalcification process leads to the decomposition of M-S-H and C-S-H, which are known as noncementitious materials, causing a loss of strength [49]. Furthermore, when the amount of BRHA increase, the pozzolanic reaction increase, which means that more $\text{Ca}(\text{OH})_2$ is consumed. Thus, to retain the system balance, the $\text{Ca}(\text{OH})_2$ is also generated from C-S-H for additional pozzolanic reaction with pozzolans. Consequently, this process generates a large number of unstable C-S-H, and the Ca^{2+} can be substituted by magnesium ions in C-S-H structures [49,50]. The substitution capacity of Mg^{2+} is larger than Ca^{2+} of Na^+ because the quantity of valence electrons of Na^+ is smaller than that of Mg^{2+} . As a result, C-S-H structures are prone to change to M-S-H structures, which are known with low bonding capacity, leading to a lower strength of concrete. In contrast, the strength loss was reduced with the increase of BRHA level when the specimens were subjected to Na_2SO_4 attack. However, the increase

in BRHA levels led to an increment of strength loss in comparison with OPC specimens when immersed in $MgSO_4$ solution (Figs. 8-11). As a result, it was concluded that BRHA can be used to replace OPC for enhancing resistance to Na_2SO_4 attack; but it may have a negative effect in terms of $MgSO_4$ attack [48].

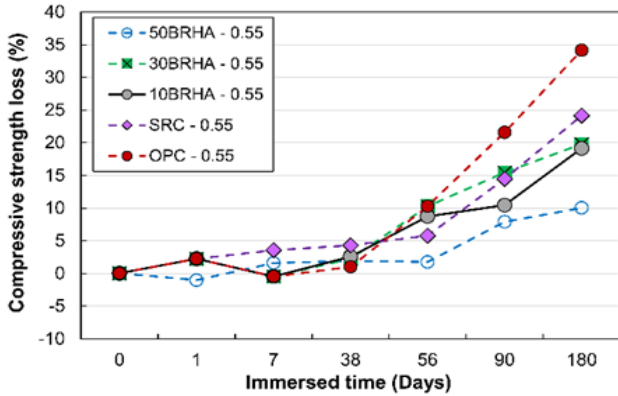


Fig. 8. Values of strength loss of specimens submerged in Na_2SO_4 solution (w/b of 0.55) [48]

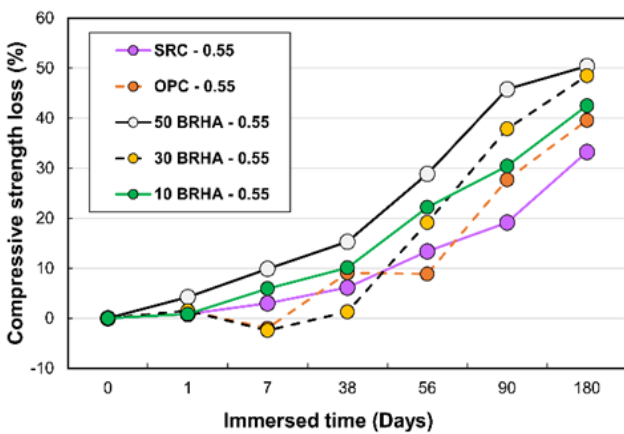


Fig. 9. Values of strength loss of specimens submerged in $MgSO_4$ solution (w/b of 0.55) [48]

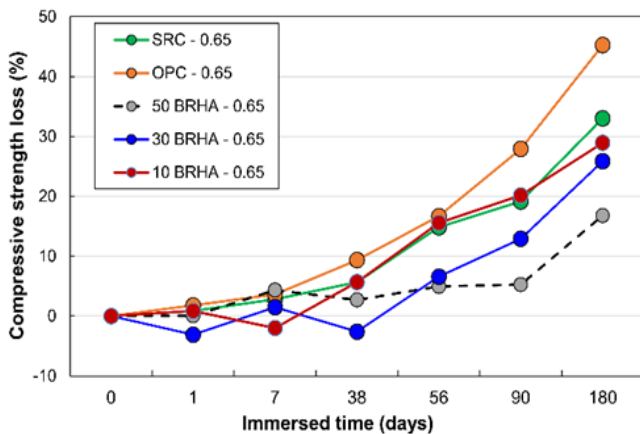


Fig. 10. Values of strength loss of specimens submerged in Na_2SO_4 solution (w/b of 0.65) [48]

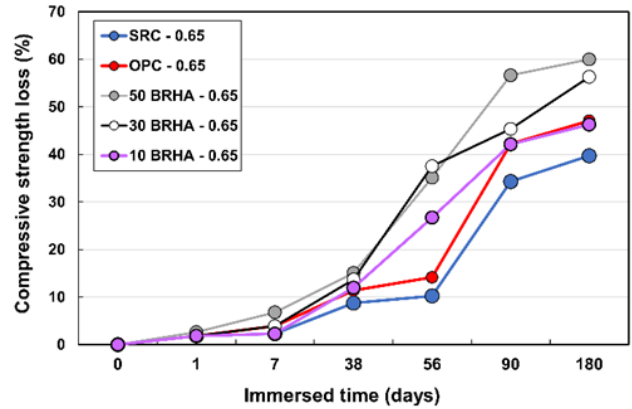


Fig. 11. Values of strength loss of specimens submerged in $MgSO_4$ solution (w/b of 0.65) [48].

It was reported that sulfate attack is expressed to the formation of ettringite and gypsum, which causes the expansion, spalling, and cracking (i.e. deterioration) in OPC concrete. Previous studies indicated that gypsum and ettringite were formed due to the reaction between sulfate and cement hydrate products [51,52]. The stiffness of the OPC paste was decreased and the water absorption of ettringite was increased when the paste was immersed in the sulfate solution [53]. A sulfate attack causes the expansion and cracking of the concrete, which leads to an increment of strength loss due to a reduction of cohesion among hydrated paste as well as among particles of aggregate [52]. Besides, several studies indicated that FA in concrete played an important role in enhancing sulfate resistance [54,55].

It was also reported that concrete partially containing ground granulated blast furnace slag (GGBS) has a higher sulfate resistance in comparison with OPC concrete because C_3A amount in concrete containing GGBS is smaller than that of OPC concrete [56]. The previous study used metakaolin (MK) and rice husk ash (RHA) as a partial OPC cement replacement and concluded that the sulfate resistance of these concrete is superior to that of conventional OPC concrete [57]. As a result, it can be stated that concrete containing SCMs significantly improved sulfate resistance, which will lead to an improvement of long-term mechanical properties as well as the durability of concrete.

3.3. Results of carbonation

Carbonation is one of the important factors, which directly affects the corrosion of reinforcement. Many factors affect the carbonation rate of concrete and mortar, including physical and chemical properties of binder, porosity, and permeability of cement hydrate matrix. The effect of by-product materials (ground limestone, fly ash, and slag) on the penetration of CO₂ of concrete

with a specified w/c ratio was investigated [58]. The authors reported that at the same strength grade, the carbonation rate of concrete with a 50% slag replacement level is similar to that of conventional OPC concrete as shown in **Table 2**. Furthermore, from **Table 2**, it can be also observed that the replacement of OPC by FA or slag up to 50% did not significantly affect the carbonation depth of concrete.

Table 2. Carbonation depth of concrete with and without SCMs inclusion at 28-day compressive strength of 40-45 MPa [58]

Component of binder (%)				Cement amount (kg/m ³)	w/c ratio	Compressive strength	Depth of carbonation (mm)		
PC	Limestone	FA	Slag				30 days	90 days	360 days
100	-	-	-	300	0.6	43.0	0.5	3.5	4.8
85	-	-	15	300	0.6	43.0	0.5	3.5	5.9
50	-	-	50	350	0.5	42.2	0.5	3.0	5.5
85	15	-	-	350	0.5	45.0	0.0	3.0	4.7
75	25	-	-	350	0.5	40.3	0.5	4.5	6.9
75	-	25	-	350	0.5	44.0	0.0	3.5	5.7

The carbonation depth of different concrete mixtures (including concrete containing MK and GGBS) is shown in **Fig. 12** [43]. From the figure, it can be observed that the inclusion of MK and GGBS led to a reduction of carbonation depth. The reduction of carbonation depth was explained by the pozzolanic reaction and filling function of MK and GGBS, which could reduce the CO₂ penetration to the pore system of concrete [43].

A study conducted by Yu et al. [59] indicated that the Portlandite amount of RHA paste (30% OPC replaced by RHA) was decreased from 3 days of curing, and Portlandite content was approximately zero after 90 days of curing. From the experimental results, they stated that concretes containing RHA have more C-S-H and less Ca(OH)₂ compared with OPC concrete, which may improve carbonation resistance. This result is consistent with the results obtained from other authors [60].

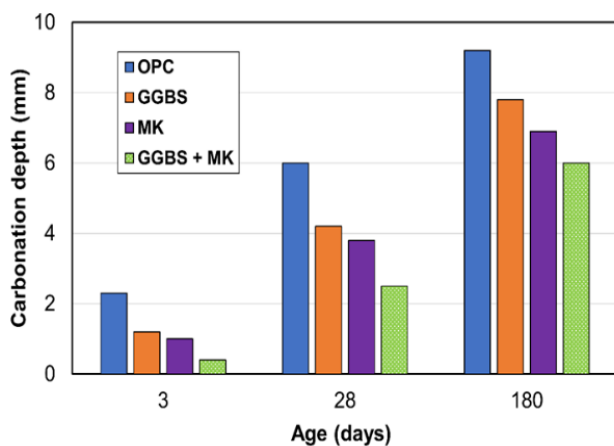


Fig. 12. Values of carbonation depth for different concrete specimens [43]

4. Conclusions and recommendations

On the basis of the above previous published studies, the following summaries can be derived as follows:

- The major ion content of seawater in different location all over the world are similar. However, there is a wide range of salinity, which varies from location to location. Thus, in order to predict accurately the long-term performance of concrete, the artificial seawater in the laboratory must have a similar salt concentration to real

seawater of the target marine condition and location.

- Many previous studies have been conducted to investigate the application of supplementary cementitious materials (SCMs) such as fly ash, metakaolin, slag, rice husk ash, etc., in concrete with different OPC replacement in terms of durability (chloride penetration, carbonation, and sulfate resistance).

- Previous studies indicated that concretes containing different SCMs levels have a higher durability (high resistance to chloride ion penetration, sulfate attack, and carbonation) in comparison with plain OPC concrete.

- The utilization of SCMs in concrete has two main advantages. Firstly, it reduces the material cost and secondly, it helps to reduce environmental impact (disposal problems and CO₂ emission).

- Finally, effective utilization of by-products as a cement replacement in mortar and concrete will be a crucial alternative solution to improve the durability of mortar and concrete in the future.

From the literature, there are some research gaps that need to be implemented in future studies as follows: durability of mortar and concrete using SCMs, cement, and fiber should be investigated. In addition, the durability of concrete comprising SCMs concerning different curing temperatures should also be examined in the future. Finally, the environmental impact assessment and cost-benefit analysis should be examined in future studies in order to understand the beneficial use of by-products for concrete in the marine environment.

Acknowledgment

This research is funded by the Ministry of Transport, Vietnam, under grant number DT2303.

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