Influence of supplementary cementitious materials on transport properties of concrete and interfacial transition zone

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Transport properties of concrete directly affect durability. A good comprehension of different transport properties and the role of supplementary cementitious materials (SCMs) will help in improving concrete quality. This article presents a brief review of the role of SCMs in concrete, transport mechanisms and their correlation with durability. The progress of research on transport properties like water penetration, sorption, electrical resistivity, chloride ingress, etc. with the partial replacement of different blenders is reviewed. The article also briefly examines the influence of SCMs on the interfacial transition zone (ITZ) and the link between ITZ and overall transport properties.

Keywords: Concrete, durability, ITZ, supplementary cementitious materials, transport properties.

MEASUREMENT of concrete transport properties helps evaluate ionic flow and durability. The entry of harmful substances into concrete through transport mechanisms can result in poor structural integrity[1]. Durability depends on transport properties, which impact the resistance of concrete to degradation. Many structural failures and damages are caused by chloride corrosion[2]. The need for more durable concrete is evident from case studies of structures losing integrity[3,4].

Transport mechanisms such as permeability, diffusion and electromigration require a medium for ion transportation. Measured transport properties include gas permeability, electrical resistivity and chloride ingress[1,5]. Ionic movement and transport mechanisms impact concrete deterioration, and a reduction in pore solution alkalinity also causes deterioration in concrete[6].

Concrete permeability can cause reinforcement corrosion from chloride and sulphates in water[5]. This can be prevented by cathodic protection using Zn- or Al-based alloys as an anode in marine areas[7]. Transport properties depend on water/binder (w/b) ratio, and reducing water content can reduce cement paste and therefore transport properties. Supplementary cementitious materials (SCMs) like ground granulated blast-furnace slag (GGBS) and fly ash (FA) improve durability and reduce cost when used as direct cement replacement or in blended Portland cement (BPC)[8,9].

SCMs impact concrete transport properties considerably, with each type (silica fume (SF), metakaolin (MK), slag (S), etc.) having a different effect. Water absorption of ordinary Portland cement (OPC) and BPC is similar, but other transport properties vary significantly depending on SCM type and replacement level, with BPC being less effective compared to OPC[10]. The threshold limits of transport properties such as WA, water penetration depth and rapid chloride permeability test (RCPT) are used in the Iranian national code on durability[11].

This article provides insight into how SCMs impact different transport properties and aids in selecting SCMs for durable concrete. It also examines the role of interfacial transition zone (ITZ) on concrete transport properties. ITZ, a thin region (~50 μm) between aggregate and cement paste, has unique microstructural features. This article highlights the effect of SCMs on ITZ and their influence on concrete transport properties.

Review methodology

A comprehensive review was conducted by searching relevant research articles through various sources such as Google Scholar, ResearchGate, Academia and publication house (and research society)-based search engines like Elsevier, Springer, Wiley, and Taylor and Francis (ASCE and RILEM). Keywords related to the topic, such as ‘SCMs’ and ‘chloride ingress’ were used. The search was continued until all relevant results were exhausted. Also, requests were made to the authors for access to their full publications. Next, a time filter (e.g. ‘since 2020’) was applied to find recent publications. This was repeated for different topics. The selected articles were examined and a conclusive review was provided along with a discussion of future research ideas. This review methodology was inspired by a similar methodology by Ravichandran *et al.*[12].

SCMs in concrete

SCMs are materials that improve concrete properties when used with Portland cement through hydraulic or pozzolanic

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Table 1. Effect of supplementary cementitious materials (SCMs) in concrete

SCMs	Properties	Remarks
Slag	Latent hydraulic	Improves concrete properties through hydraulic and pozzolanic reaction[15]
Limestone filler (LF)	Filler effect	Increases finer fraction without altering water demand[10]
Metakaolin (MK), silica fume (SF)	High reactivity	Accelerates cement hydration, generates additional calcium silicate hydrate, improves microstructure[15] and interfacial transition zone (ITZ)[18]
Ground granulated blast-furnace slag (GGBS), fly ash (FA)	Low reactivity	Refines pore size distribution, improves resistance to sulphate attack[19] and corrosion[15]
Nano-silica (NS)	Filler effect, high reactivity, high specific surface area	Enhances microstructure through filler and nucleation effect, and increases early strength[20,21]

Table 2. Design mix combinations used by various researchers

Reference	Binary combination	Ternary combination
Ahmadi *et al.*[4]	7.5% SF, 10% ZP and 20% FA	5% ZP – 5% SF and 10% ZP – 10% FA
Wu *et al.*[38]	5% LF, 10% LF, 35% GGBS and 70% GGBS	30% GGBS – 5% LF, 65% GGBS – 5% LF, 60% GGBS – 5% LF
Ghafoori *et al.*[39]		SCC mixtures: 10–20% LF (8 and 3 μm) with 20% FA

ZP, Zeolite powder. SCC, Self-compacting concrete.

activity, or both[13]. They hydrate to form cement-like products, replacing cement and promoting sustainability while reducing CO_2 (refs 14–16). SCMs also affect concrete transport properties based on particle size, physical and chemical action, pozzolanic reactivity, and nature of the hydration products[17] (Table 1, refs 18–21).

SCM combinations (two or more) yield synergistic benefits[22,23], such as combining SF and FA, which yields refined microstructure and improved durability[24].

Effect of physical properties and chemical composition of SCMs on transport properties

The extent of concrete microstructural improvement depends on SCM reactivity, which is influenced by their physico-chemical properties[25]. The reactivity can be modified through changes in surface area, thermal history or thermal activation[26]. Thus, shape, chemical composition and mineralogy determine SCM reactivity[26]. The physical properties and chemical composition of SCMs have been well documented in previous studies[27,28].

Influence of physical properties: The physical properties of SCMs, such as specific surface area, particle size distribution, shape and density affect their reaction kinetics. Higher surface area leads to faster dissolution but also increases water demand[29,30]. Smaller particle size results in more nucleation sites and accelerated hydration[31], as observed in studies based on nano-silica (NS)[32].

Influence of chemical properties: The reactivity of SCMs is determined by their crystalline and/or amorphous phases. Durdziński *et al.*[33] observed that different samples of FA have different glass phases, which have varying dissolution rates based on composition and structure[34], with reaction rate increasing with calcium content. Skibsted and Snelling[26] have provided a comprehensive review of the same.

Effect of SCMs on different transport properties

Electrical resistivity

Electrical resistivity (ER) measures the resistance of a material to ion flow in an electric field, which is influenced by microstructural properties like pore shape and size, and tortuosity[35]. Pore liquid determines conductivity at constant moisture content, assuming negligible conductivity of aggregates and hydrated products[10]. ER has a direct correlation with the rate of corrosion[36]. Thus, it provides a fair idea about corrosion resistance[37]. Experiments have shown that for different mixtures containing FA and slag, ER was inversely proportional to chloride diffusion coefficient, providing an indirect estimate of chloride penetration resistance[36].

The presence of SCMs enhances the concrete ER due to pozzolanic reaction, leading to refined pore structure, reduced ionic concentration of the pore solution and altered pore liquid chemical composition[36]. Late hydration products in the pores further affect the evolution of resistivity with time[10]. A combination of SCMs is beneficial for improving ER. Ahmadi *et al.*[4] conducted experiments using SCMs mentioned in Table 2 and reported that mixtures containing SF (binary and ternary) showed the highest increase in ER, with resistivity increasing by 77%, 62%, 30%, 78%, and 58% compared to the control mix (Table 2)[4,38,39]. Adding 5–15% SF also led to a significant improvement in ER[40]. Mixtures containing the slag also showed a significant increase in resistivity[10,36].

Sorptivity

Capillary sorption is defined as the rate of increase in weight per unit area due to the sorption of water by one face[10]. This phenomenon is measured by determining the slope of best fit line between the mass increase per unit area/water density ratio against the square root of elapsed time.

SCMs improve concrete microstructure and hence sorptivity. Zaccardi *et al.*[10] conducted experiments using slag and limestone filler (LF) and observed lower capillary sorption rate in mixtures with low paste content. Slag refines pore structure through pozzolanic reaction, while LF increases packing density and acts as a filler. On the contrary, BPC mixes with water/cement ratio of 0.6 and 0.45 showed higher capillary sorption due to the dependence on total capillary porosity, mainly contributed by the paste.

Aggregates are considered impermeable, leaving paste to contribute maximum porosity. Less paste reduces total porosity and sorptivity. According to Wu *et al.*[38], excessive replacement of cement with GGBS (70%), beyond the optimum level, results in refined pore structure but with increased pore volume.

Gas permeability and water penetration depth

Permeability is the property by virtue of which any porous material allows the flow of liquids or gases through it[41]. Most permeability measurement techniques are based on Darcy's law. Permeability depends mainly on pore size, pore connectivity, tortuosity, viscosity of the fluid and capillary pores[42]. Permeability is a function of pore size, while diffusion is independent of it[43].

The addition of SCMs improves concrete permeability significantly. MK being highly reactive, significantly improves porosity. Studies have reported a 40–50% reduction in gas permeability with 15–20% cement replacement by MK[1,44].

Binary or ternary mixtures can improve transport properties. Ahmadi *et al.*[4] found lower water penetration depth (WPD) values in all mixtures compared to control concrete, with a reduction of 10–45% (Table 2)[4,38,39]. Mixtures with zeolite powder (ZP) showed the greatest reduction in permeability, while FA was not effective due to poor pozzolanic activity.

Another factor affecting porosity is the w/b ratio. Porosity was experimentally validated to increase with w/b ratio[10]. Experiments were conducted with slag and/or LF at different w/b ratios starting at 0.4 and it was observed that slag refined pores with a nominal decrease in porosity. However, with an increase in w/b ratio (to 0.45), the results showed an opposite trend. Menéndez[45] reported that limited LF content in BPC can improve durability, while excess may cause deterioration. Studies have demonstrated improvement in concrete with combined use of LF and slag[46,47]. LF increases early strength by accelerating hydration, while slag improves later strength. Wu *et al.*[38] recommended the synergy effect of LF and GGBS for improved concrete. According to Ozbay *et al.*[48], GGBS reacts with calcium hydroxide to refine pore size and reduce permeability and forms additional calcium silicate hydrate to provide increased surface area for adsorption[49,50].

The particle size of SCMs also affects concrete transport properties. Ghafoori *et al.*[39] experimented on self-compacting concrete mixtures (Table 2)[4,38,39]. They observed a significant decrease in WPD with LF, but further reducing the particle size of LF below 8 μm and increasing the replacement level beyond 10% had an insignificant effect on WPD. This can be explained with the help of dilution, filler and nucleation effect of LF[51]. Being a good filler, LF also acts as a nucleation site and accelerates hydration resulting in disoriented CH structures which improve WPD up to a certain replacement level, beyond which it loses its significance due to the dominance of percolation effect of ITZ. Similar results were reported by Ramezanianpour *et al.*[52].

Water absorption

Water absorption (WA) is the ability of a material to intake water through capillary suction. It depends upon pore volume and interconnection of pores[53]. In partially saturated conditions, salt and water enter primarily through absorption[54].

SCMs improve concrete WA considerably, reducing the WA coefficient by 20–30% with 10–15% MK[55,56]. Binary and ternary mixtures can further reduce WA, as seen in experiments by Ahmadi *et al.*[4]. The mixtures showed a reduction of 40%, 25%, 21%, 30%, and 23% compared to control concrete, and none of them exceeded WA by 2% (Table 2)[4,38,39]. The presence of SF, ZP and FA reduced WA in order of their pozzolanic reactivity[4].

Studies have shown that the water/paste ratio and particle size can affect porosity; with a reduction in water/paste ratio at a constant water/cement ratio, the total porosity decreases[57]. Ghafoori *et al.*[39] conducted a WA test on SCC mixtures with the same water/cement ratio but different water/paste ratios (Table 2)[4,38,39]. It was reported that WA reduced significantly on using 3 and 8 μm. LF, with more replacement leading to less WA due to the filling effect, as confirmed by XRD[39]. The improvement was better for 3 than 8 μm (especially at the early stages) due to denser packing and the absence of further filling effect from the hydration product of FA.

Chloride ingress

Chloride-ion penetration (CP) concrete slowly through a multi-mechanistic phenomenon, with diffusion being the major one. Diffusion indicates ionic flow due to concentration gradient[9] and causes major deterioration in RCC bridges[58].

Table 3. Previous studies on ITZ

Reference	Study parameters	Remarks
Farran[75]	First study	Origin of ITZ
Watson and Oyeka[76]	Oil permeability tests on concrete and mortar, water/cement between 0.3 and 0.8	Concrete permeability ~100 times higher than cement paste
Houst *et al.*[77]	CO_2 and O_2 diffusivity measured at 0.67 water/cement with varying cement/sand	Diffusivity decreased with increasing sand content up to 50%; thereafter diffusivity increased (percolation effect of interconnected ITZs)
Carcasses *et al.*[78]	Permeability test with different sand content (water/cement ratio 0.35)	Sharp increase in permeability at 40% sand content
Asbridge *et al.*[79]	Comparison of chloride diffusivity at steady and non-steady states	Mortar showed increased diffusivity at 35% sand content, suggesting interconnected ITZs
Wu *et al.*[38]	ITZ study with LF and GGBS replacement	Linear relationship between chloride diffusion coefficient and pore volume was obtained
		Reduction in pore (>100 nm) volume indicated densified ITZ
Sun *et al.*[80]	Porosity	ITZ porosity about six times higher than porosity at 50 μm from aggregate
Zheng *et al.*[81]	Numerical model to determine chloride diffusivity	Diffusivity decreased with increasing volume fraction of aggregates, curing period, and maximum aggregate diameter, but increased with width of ITZ or water/cement ratio

The rate of CP in concrete depends on its binding capacity, which is influenced by the presence of SCMs[59,60] and C_3A content[61,62]. Chlorides react with concrete, forming physical and chemical bonds which reduce diffusion rate, leading to inaccurate readings if measured before reaching a steady state. RCPT and rapid chloride migration test (RCMT) were used to measure CP. RCPT measures the movement of all ions and is affected by pore solution and structure, while RCMT measures only the depth of CP and depends solely on pore structure. The results of RCPT may be affected by the presence of conducting fibres and highly ionic, corrosion-inhibiting admixtures[63], while RCMT is not affected by pore solution conductivity[57].

SCMs reduce CP in concrete due to improved microstructure with age and the progress of hydration[64]. For example, a 10% replacement of MK in concrete has been shown to reduce CP resistance by 88% (ref. 65), while a 15% SF addition improved CP resistance twofold[40].

More than one SCM is often examined for improving CP. Ahmadi *et al.*[4] found that binary SF mixtures and ternary mixtures performed better than other mixtures and control samples, with the ternary mixture showing a 33–49% reduction in CP compared to the control mix with a refined porous matrix and increased tortuosity (Table 2)[4,38,39].

The size of particles in a concrete mixture affects CP. Ghafoori *et al.*[39] conducted RCPT and RCMT on different mixtures using LF (Table 2)[4,38,39]. They concluded that a reduction in particle size leads to reduction in passage of charge[39]. However, the effect of particle size becomes insignificant beyond an optimum replacement level due to the refinement of pore structure.

The bulk coefficient of diffusion also measures CP. Studies have shown reductions up to 70% with 10–15% MK replacements, leading to improvements in concrete microstructure[1,55,66,67]. However, casting and curing temperatures can also affect pore structure, resulting in a greater diffusion coefficient[68].

Effect of ITZ on transport properties

ITZ is an inhomogeneous region with a distinguished microstructure which determines the strength of concrete. It is a source of microcracks and the entry of harmful species[69,70]. The strength difference between ITZ and cement paste matrix determines the tendency for microcrack propagation in the zone[71,72]. This highlights the need to study the influence of SCMs on ITZ.

SCMs are fillers that improve the ITZ microstructure through pozzolanic reaction products. Geopolymer concrete has a stronger ITZ than normal concrete[73]. Using SCMs like SF can reduce ITZ thickness by 36% (ref. 74). SCMs can also be combined to achieve a synergy effect, as seen in a study by Wu *et al.*[38] that examined the impact of LF and GGBS on transport properties in ITZ. Returning to objective of reviewing past studies on ITZ, a summary is presented in Table 3 (refs 75–81).

Link between ITZ and overall concrete transport properties

Studies on ITZ have contradictory observations, with three different viewpoints. The first viewpoint by Wong *et al.*[69] claims that transport properties such as permeability, sorptivity and diffusivity decrease with an increase in volume fraction of aggregates (VFA); but this was found to be due to increased microcracking and decreased tortuosity, and not just ITZ porosity. Transport properties are influenced by multiple factors, including microstructure, volume fraction of bulk and microcracks, and not just ITZ. This has been validated against a numerical model[81]. Several studies support this conclusion[82–84].

Wu *et al.*[85] observed a different trend, which represents this second viewpoint. Several factors that may impact the transport properties of bulk and ITZ were studied, including (i) reduction in permeability with increasing VFA, (ii) porous ITZ with a dense paste matrix and (iii) increased permeability due to interconnected ITZ. Results showed that permeability in concrete decreased with an increase in VFA up to 35% (due to the dominance of dilution and tortuosity effects). Beyond 35% VFA, an increasing trend was observed, as the ITZ effect dominated over dilution and tortuosity effects. This was validated through chlorine migration coefficient measurements, which showed that the ratio of the chlorine migration coefficient of ITZ to that of the matrix increased with increasing VFA above 35%. The results also confirmed that ITZ volume and influence on transport properties depend on VFA and the assumed ITZ thickness and that the influence of ITZ depends on the degree of interconnection (DOI). At a threshold VFA of 35%, DOI was between 0 and 1, indicating the initiation of percolation. As inter-aggregate distance reduced with increasing VFA, DOI approached 1, leading to a sharp increase in the ratio of chlorine migration coefficient of ITZ to that of the matrix, indicating complete percolation of ITZ. Similar results have been reported in previous studies[86,87].

A third viewpoint reported ambiguous observations by Maghsoodi and Ramezanianpour[88]. It was initially observed that transport properties (gas transport and electrical conduction) decreased with increasing aggregate fraction up to 0.35. Beyond 0.35, the trend reversed, with a continued decrease in transport properties. This trend cannot be explained by current theories on ITZ percolation effect and requires further research for a proper explanation.

Discussion and way forward

Incorporation of SCMs in concrete has been proven to be a cost-effective approach while also improving the strength and durability of the material. Although the role of SCMs in enhancing transport properties has been explored, the influence of ITZ on overall concrete properties remains contradictory and needs further research. To better understand the impact of SCMs on transport properties, tests focusing on (i) quantifying the effect of SCMs on the size of ITZ, and (ii) development of effective cohesion between SCM particles and hydration products should be conducted[89].

Additionally, a correlation between effective cohesion and the long-term durability of concrete should be established. Developing a model to predict the effect of SCMs on transport properties, similar to that explored in previous studies on compressive strength[90,91], can also be a valuable future direction of research. Furthermore, analysing of the combined effect of multiple transport mechanisms in the presence of SCMs would provide a deeper insight into the role of SCMs on transport properties. The interplay between carbonation and SCMs on chloride permeability is an example of a well-researched topic in this field, as reviewed by Abdulhussein and Kopecskó[92].

Conclusion

Salient observations from this study are summarized below.

(i) The extent of improvement of transport properties with SCM depends on its type, chemical composition, particle size, pozzolanic reactivity and replacement level.
(ii) SCMs can serve as good fillers (LF) or form pozzolanic products (SF, MK, FA, GGBS) and refine the microstructure of bulk and ITZ.
(iii) The combined effect of two or more SCMs is more beneficial, leading to a synergic effect.

The contradictory results regarding the influence of ITZ on overall transport properties highlight the need for further research.

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