

A Review of Sustainable Supplementary Cementitious Materials as an Alternative to All-Portland Cement Mortar and Concrete

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Abstract: Supplementary cementitious materials (SCMs) have been widely used all over the world in ready-mixed concrete due to their economic and environmental benefits; hence, they have drawn much attention in recent years. Whether deriving from industrial waste, agro-waste or by-products, supplementary cementitious materials can be mixed with blended cement to enhance concrete strength. Supplementary cementitious materials may contain fly ash (FA), silica fume (SF), ground granulated blast furnace slag (GGBFS), rice husk ash (RHA), metakaolin (MK) and palm oil fuel ash (POFA), to name a few. The utilization of these materials in concrete can partially reduce the consumption of Portland cement, which, in turn, can lessen construction costs, providing materials suppliers, contractors and engineers with substantial advantages. Furthermore, despite the drawbacks of their binary blends, the combination of supplementary cementitious materials can lead to many advantages, such as optimized strength, workability and durability. Unfortunately, these advances have not been fully taken into consideration in state specifications. Hence, by adopting a review approach, this study aimed to provide new insights into the effect of the incorporation supplementary cementitious materials on the properties of mortar and concrete.

Key words: Supplementary Cementitious Materials, Cement, Concrete, Blended Cement, Fresh Properties, Hardened Properties, High performance concrete.

INTRODUCTION

Concrete, the most widely used construction material, essentially consists of embedded particles of aggregates in a cement paste matrix. This paste is composed of a mixture of hydration products, un-reacted cement, water and pores. Technologically simple and easy to make, concrete has been considered the first widely used construction material throughout the world. It has been reported that Portland cement consumption increased drastically from 2 million metric tons to 1.3 billion metric tons over a little more than a century (1880-1990). As shown in Fig. 1, this upward trend, as stated by CEMBUREAU, started near the end of 20th century (Aitcin, 2000). Therefore, it is hardly surprising to see that the process of making cement is among the third largest CO₂ producers in the world. According to (Malhotra, 2002), during this process, for every ton of cement produced, more than half of all CO₂ emissions are released into the air. This dramatic increase in CO₂ emissions from cement production has been visualized by (Muga *et al.*, 2005) and is shown in Fig. 2. Undeniably, cement production depends on many other factors. More often than not, the production of cement is an extremely energy intensive production process because the energy and the consumption of other natural resources per ton are estimated to be approximately 4 GJ and 1.6 ton, respectively (Muga *et al.*, 2005; Malhotra, 1993). The CO₂ emission is evaluated based on the compressive strength of concrete through the combination and separation of different types of concrete already used in construction sites in Korea. Given the relationships between life cycle characteristics and CO₂ use, the emission of CO₂ can be assessed for environmentally friendly concrete production (Park *et al.*, 2012). To combat the aforementioned problems, cement can be replaced by supplementary materials. These materials can be categorized as natural or artificial. The former can be found in natural pozzolans and volcanic tuffs, and the latter can be obtained from fly ashes, condensed silica fume and metallurgical slags (Mehta, 1989; RILEM 73-SBC Committee, 1988; Sersale, 1983; ACI Committee 226, 1987; Malhotra, 1987). Most of these supplementary cementing materials are by-products; thus, their inclusion not only serves as an invaluable means to preserve environmental resources but also enhances concrete construction properties, including its sustainability (Aitcin, 1998; Malhotra and Mehta, 1996; Mehta and Monteiro, 2006).

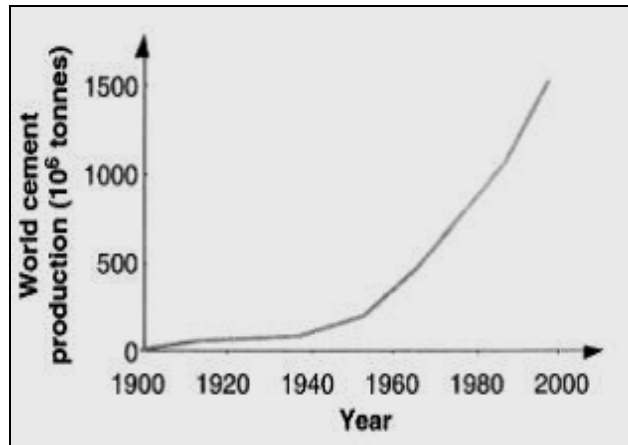


Fig. 1: Global production of cement according to CEMBUREAU (Aitcin, 2000).

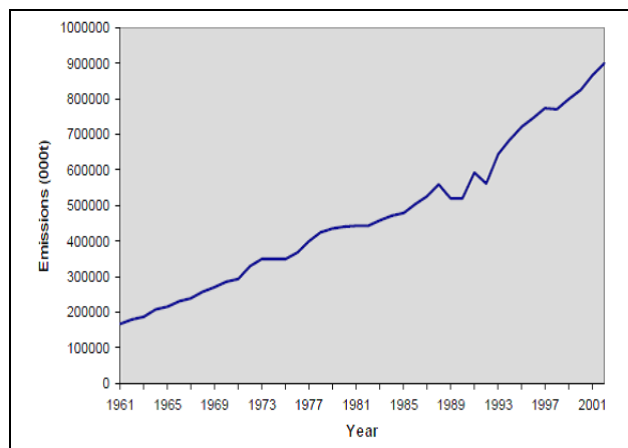


Fig. 2: Carbon dioxide emissions due to the world production of cement (Muga *et al.*, 2005)

Furthermore, due to their filler effect and pozzolanic reaction characteristics in improving concrete performance, these materials have been increasingly used in high-strength and high-performance concrete (known as HSC and HPC, respectively) production. To increase the durability, the stiffness, and the mechanical properties of HSC, it is essential for the water/binder ratio to be decreased and the binder content to be increased. Therefore, super-plasticizer (SP) must be used to replace the water ratio with silica fume, coal fly ash or natural pozzolan. Due to the combination of these superior characteristics, HSC has been used extensively as a better alternative for off-shore structures, near-shore and high-rise buildings, bridges, and nuclear containment compared to its normal-strength counterpart. On the other hand, to increase the freshness and hardness of HPC, either cement alone or a mixture of cement and mineral components (silica fume, blast furnace slag and fly ash, to name a few) are used in ternary systems, which ultimately result in more economical and ecologically friendly high-performance concrete. With such properties, to date, normal-strength concrete has been replaced with multi component products that have one or more admixtures in addition to water, cement, and fine and coarse aggregates. However, each one of these components could have a direct share in the end-product cost and its service behavior. Despite their vital role in producing durable and strong concrete, these cementitious materials may decrease the early strength of concrete, particularly if the cement replacement rate is high (Naik and Singh, 1998; Mehta, 1994). Nevertheless, this decrease is preventable if the concrete is cured at an elevated temperature (Maltais and Marchand, 1997). Furthermore, if the water to cement ratio is decreased, cracking can be minimized (Naik and Ramme, 1987; Malhotra, 1992). High fineness additives and cement (Mittal and Kumar, 1996; Osback and Smith, 1981), activation (Davidovits, 1994; Poon *et al.*, 2001a), and cement replacement with more than one additive (Naik and Ramme, 1987; Malhotra, 1992) are the other means to compensate for this loss.

Following the discussion on improving concrete quality and its performance, ternary blended cements that consist of, for example, fly ash, Portland cement, and/or ground granulated blast-furnace slag have recently been considered better alternatives than traditional binary blended cements. Adding fly ash to slag cement can not only enhance feasibility but also decrease water bleeding in slag cement concrete. Due to poor hydration heat in

Japan, this approach is widely used for mass concrete construction (Uchikawa and Okamura, 1993). A significant amount of research has also been devoted to the use of silica fume in binary and ternary cement to increase the concrete's strength while decreasing its permeability, which is easily achievable because silica fume is extremely fine and its pozzolanicity is remarkably high. However, its high surface area can negatively affect its feasibility (Bagel, 1998; Khan *et al.*, 2000). Likewise, agricultural waste is another economically reasonable ingredient that has recently drawn much attention due to population growth, social advancement, and industrial and technological developments. Undeniably, the utilization of agricultural waste in both aggregates for concrete construction and fiber-reinforced concrete has a considerable impact on the environment. For example, fly ash as a cement replacement is available in significant quantity and has economic and engineering advantages. Similarly, RHA and POFA can be combined with pulverized fuel ash and slag to be used in multi-blended cement. In particular, pulverized fuel ash has unique characteristics that reduce water bleeding, thus enhancing the feasibility of the use of lower early-strength concretes (Khan *et al.*, 2000; Ravindra, 1986). In contrast, slag is an active additive (Wan *et al.*, 2004) that is predominantly used to repair material (Sobolev and Yeginobali, 2005) because of its high sulfate and acid resistance and its low heat hydration (Binici and Aksogan, 2006).

Supplementary Cementitious Materials:

Generally, concrete is a versatile material that consists mainly of sand, water, gravel and Portland cement, which is considered the main cementitious ingredient in concrete. To date, supplementary cementitious materials have been widely used (60%) in ready-mixed concrete due to their natural nature (PCA, 2000). As an example, North America pioneered the use of these materials in the 1970s. As shown in Fig. 3, these by-product materials, such as calcined shale, silica fume, calcined clay, ground granulated blast furnace slag, and fly ash, can be mixed with blended cement to enhance concrete strength (Kosmatka *et al.*, 2003). Due to their vast availability, these supplementary cementitious materials can either be used independently or in a combination of two or even three (ternary mixtures) by concrete producers. Nevertheless, not all of these natural materials function directly in concrete. Indeed, they should meet certain established standards to attain the desired effect on concrete. To exhibit cementitious properties, some of these materials, such as slag, can be used individually. Nevertheless, some pozzolans or siliceous materials cannot chemically react with water (i.e., exhibit pozzolanic or hydraulic activity) unless they are mixed with Portland cement. Under such circumstances, pozzolans can form calcium silicate hydrate and other cementitious admixtures. Pozzolanic activity occurs when a mixture of aluminous siliceous materials and calcium hydroxide create cementitious properties. In fact, lime more readily reacts with amorphous silica than with crystalline materials (Habeeb and Fayyadh, 2009). The classification and specifications of such supplementary materials are summarized in Table 1.



Fig. 3: Some of Supplementary cementitious materials. From the left to the right, class C fly ash, metakaolin (calcined clay), silica fume, class F fly ash, ground granulated blast furnace slag, and calcined shale (Kosmatka *et al.*, 2003)

Table1: Classification and specifications of such supplementary cementitious materials

Ground granulated iron blast-furnace slags—ASTM C 989 (AASHTO M 302)	
Grade 80	Slags with a low activity index
Grade 100	Slags with a moderate activity index
Grade 120	Slags with a high activity index
Fly ash and natural pozzolans—ASTM C 618 (AASHTO M 295)	
Class N	
Raw or calcined natural pozzolans including:	Diatomaceous earths
	Opalinecherts and shales
	Tuffs and volcanic ashes or pumicites
	Calcined clays, including metakaolin, and shales
Class F	Fly ash with pozzolanic properties
Class C	Fly ash with pozzolanic and cementitious properties
Silica fume—ASTM C 1240	

- ASTM C 989, Specification for Ground Granulated Blast-Furnace Slag for Use in Concrete and Mortars.
- ASTM C 618, Specification for Coal Fly Ash and Raw or Calcined Natural Pozzolan for Use in Concrete.
- ASTM C 1240, Specification for Silica Fume Used in Cementitious Mixture.

The use of supplementary cementitious materials in concrete is advantageous in many ways. Their use improves and increases 1) rheological properties, making concrete easier to pump, place and finish; 2) the strength of concrete; and 3) the resistance to chloride ions and sulfate attack. Supplementary cementitious materials are also advantageous in other ways: reducing 1) the permeability to water and other fluids, 2) the corrosion rate of embedded steel, 3) the risk of delayed ettringite formation, and 4) deleterious expansion due to the alkali-silica reaction.

Types of Supplementary Cementitious Materials:

Rice Husk Ash (RHA):

The coating of the seeds or grains (husk) of rice plants can absorb significant amounts of silica (approximately 85%) from the soil during its growth period (Smith and Kamwanja, 1986; Siddique, 2008). The weight of silica extracted from rice husks or rice hulls is approximately 30% of the gross weight of a rice kernel. This amount is also responsible for 80% and 20% of the organic and inorganic substances in the plant, respectively. Rice husks are also economically advantageous in agricultural and manufacturing processes because of the tons of rice produced by rice paddies all over the world (Rice market monitor, 2009). Rice husks are by-products that are responsible for 20% of the RHA weight after the husks have been burned (Anwar *et al.*, 2001). This ash (Fig. 4 (Van Dong *et al.*, 2008), with its high specific surface area and non-crystalline features, can result in highly pozzolanic reactions (Tashima *et al.*, 2004); hence, this ash is considered a better alternative for Portland cement in fillers for high-performance concrete (Smith and Kamwanja, 1986; Zhang *et al.*, 1996; Hasparyk *et al.*, 2000; Sakr, 2006; Sata *et al.*, 2007).

One of the most salient causes of the highly pozzolanic reaction is due to the fineness of RHA after grinding. According to (ASTM C 618-94, 1994), these rice husk ashes should comply with the current standards, such as ASTM C 618-84. Table 2 (below) shows the physical and chemical features of rice husk ashes (Mehta, 1992; Zhang and Mohan, 1996; Bui *et al.*, 2005). According to this table, like silica fume, silica accounts for more than 80 per cent of all substances in the rice plant. Rice husk ash is designated as a class F pozzolan because the amount of CaO in the rice husk ash is less than 10 per cent. Furthermore, with such large size particles, it is possible for RHA particles to promote high surface area because of their cellular properties. This unique characteristic cannot be observed in silica fume, for example. Fig. 5 (Rukzon *et al.*, 2009) shows the average particle shape of the rice husk ash. Moreover, it has been found that, although other supplementary materials (e.g., slag and fly ash) are all equally responsible for concrete strength development, rice husk ash has greater pozzolanic reactivity, which leads to earlier pozzolanic reactions at a controlled temperature (Zhang *et al.*, 1996; Malhotra, 1993; Ismail and Waliuddin, 1996; Nehdi *et al.*, 2003). This feature is why rice husk ash is used as an additive in high-performance concrete.



Fig. 4: Rice husk ash powder (Van Dong *et al.*, 2008)

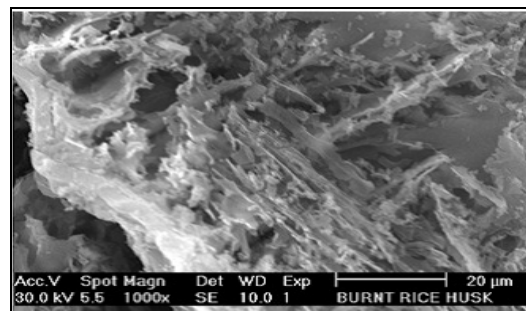


Fig. 5: Scanning electron microscope of RHA particle (Rukzon *et al.*, 2009)

Table 2: Chemical and physical features of rice husk ashes

Chemical properties	Composition %	Mehta, 1992	Zhang and Mohan, 1996	Bui <i>et al.</i> , 2005
Silicon dioxide (SiO ₂)		87.2	87.3	86.98
Aluminium oxide (Al ₂ O ₃)		0.15	0.1	0.84
Ferric oxide (Fe ₂ O ₃)		0.16	0.16	0.73
Calcium oxide (CaO)		0.55	0.55	1.40
Magnesium oxide (MgO)		0.35	0.3	0.57
Sulphur oxide (SO ₃)		0.24	0.24	0.11
Sodium oxide (Na ₂ O)		1.12	1.12	2.46
Potassium oxide (K ₂ O)		3.68	3.68	----
Loss on ignition (LOI)		8.55	8.5	5.14
Physical properties		Mehta, 1992	Zhang and Mohan, 1996	Bui <i>et al.</i> , 2005
Specific gravity (g/cm ³)		2.06	2.06	2.10
Mean particle size (µm)		----	----	7.4
Fineness: passing 45µm (%)		99	99	----

Fly Ash (FA) or Pulverized-Fuel Ash (PFA):

Fly ash (FA) or pulverized-fuel ash (PFA) is a pozzolanic material that is used in cement-based materials to enhance long-term strength, workability, resistance to sulfate attack and durability in concrete. When harvested from electric power generation plants, FA can exhibit high pozzolanic activity because it is formed from the burning of pulverized coal (Haque and Kayali, 1998). This industrial non-hazardous waste results from the burning off of volatile matter and carbon in coal due to ignition in the furnace, which then helps the mineral impurities in the coal (e.g., feldspar and clay) to fuse in suspension and float out with the exhaust gases. This process is followed by the cooling and solidification of the fused material into what it is called fly ash (Fig. 6 (Kosmatka *et al.*, 2003). However, there is still one more step necessary for the fly ash to be collected from the exhaust gases. This step can be performed by bag filters or electrostatic precipitators, depending upon the collecting system. A typical FA particle was visualized by (Kosmatka *et al.*, 2003) and is shown here in Fig. 7. For more information about the physical and chemical characteristics of FA, refer to Table 3 (Poon *et al.*, 2000; Wu *et al.*, 2002). The fly ash particles are fine-grained particles that are normally sized between less than 1 µm to over 100 µm. However, it is worth mentioning that the diameter of the average fly ash particle is smaller than 20 µm in size, and only 10-30% of all fly ash particles have a size of more than 45 µm. The surface area of fly ash, on the other hand, typically varies from 300 to 500 m²/kg, where 200 m²/kg is the lowest surface area and 700 m²/kg is the highest surface area. The density of fly ash also ranges from 540 to 860 kg/m³. The maximum bulk density reported for FA under close-packed storage or vibration ranges from 1120 to 1500 kg/m³. In general, the specific gravity of FA ranges from 1.9 to 2.8. Depending upon its chemical and mineral constituents, the color may vary from tan to dark gray.

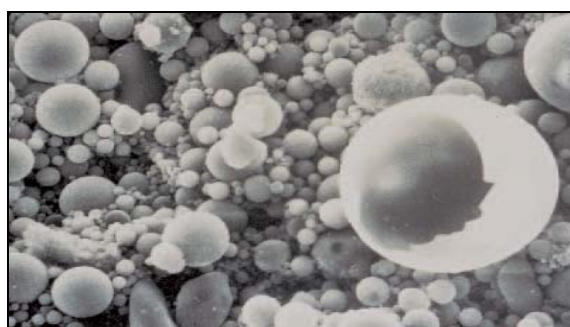


Fig. 6: Scanning electron microscope micrograph of fly ash particles (Kosmatka *et al.*, 2003)



Fig. 7: Fly ash powder (Kosmatka *et al.*, 2003)

Table 3: Chemical and physical properties of some fly ashes.

Chemical properties	Fly Ash, Class F (Haque and Kayali, 1998)	Fly Ash, Class C (Poon <i>et al.</i> , 2000)	Requirements of ASTM C 618-84	
Composition %			F	C
Silicon dioxide (SiO ₂)	56.8	35.0		
Aluminium oxide (Al ₂ O ₃)	28.2	18.5		
Ferric oxide (Fe ₂ O ₃)	5.3	5.4		
SiO ₂ + Al ₂ O ₃ + Fe ₂ O ₃	90.3	58.9	70.0	50.0
Calcium oxide (CaO)	3.0	27.7		
Magnesium oxide (MgO)	5.2	5.5		
Sulphur oxide (SO ₃)	0.7	2.4	5.0	5.0
Loss on ignition (LOI)	3.9	0.2	6.0	6.0

Physical properties	Fly Ash, Class F (Haque and Kayali, 1998)	Fly Ash, Class C (Poon <i>et al.</i> , 2000)	Requirements of ASTM C 618-84	
			F	C
Specific gravity	2.31	2.69	---	---
Fineness: Retained on 45µm (%)	6.3	5.2	≤ 34	≤ 34

Ground Granulated Blast Furnace Slag (GGBFS):

Ground Granulated Blast Furnace Slag (GGBFS) is a by-product material produced by the blast-furnaces used to make iron. Operating at a temperature of approximately 1600°C, a mixture of iron ore, coke and limestone in the furnace can produce two products, namely, molten iron and molten slag. Due to its light weight, molten slag floats on top of the molten iron. Chemically similar to Portland cement, molten slag is composed mostly of silicon dioxide (30% - 40%) and CaO (40%). Silicates and alumina, the main ingredients in molten slag, are then cooled down by high-pressure water jets (Higgins, 2007). Ultimately, this rapidly water-quench process results in glassy, granulate particles. These particles are further processed by drying and grinding to the required size to make a fine, glassy powder known as granulated blast furnace slag (Hooton, 2000). The results can be different and depend very much upon the speed of cooling of the slag melts. In the case of slow cooling, for example, the resulting material is composed of a stable solid with Ca-Al-Mg silicates. However, granular glassy material that holds latent hydraulic properties is formed during rapid cooling (between 900° and 800°C). This process results in non-crystalline slag. The water in the granulated slag can eventually be dried in a rotating ball mill. As shown in Fig. 8, ground granulated slag sized less than 45 µm has a surface area of approximately 400 to 600 m²/kg. Fig. 9 shows scanning electron microscopy (SEM) images of ground slag, which has rough edges and sharp angles. Germany was the first country that produced granulated blast furnace slag (Malhotra, 1996), which has also been used in North America for public purpose concrete. Since its initial production, ground granulated blast furnace slag has been widely used as a constituent of blended cement or a mineral admixture in cementitious materials (Shi and Qian, 2000; Demirboga, 2003).

In concrete, the GGBFS replacement rate may vary from 35 to 65 per cent, which ultimately results in a reduction of approximately 0.5 ton of CO₂ per half replacement of each ton of Portland cement. Some components of slag can have more than 70 per cent cementitious properties. As shown in Table 1, according to ASTM C 989 (AASHTO M 302), slag can be classified into three strength grades, namely, Grade 80, 100, or 120, depending upon its increasing level of reactivity (ASTM C 989-93, 1993). Regarding the chemical properties, the author of (Sohaib *et al.*, 2001) believes that the nature and type of the iron ore, the limestone flux composition and the coke consumption are the key factors in determining the variety of slag. Table 4 shows the physical properties of GGBFS along with its chemical composition. When it is compared with Portland cement, numerous studies show that the early strengths in concrete containing GGBFS are lower due to its initial reaction rate. Hence, when slag Portland cement is blended with water, the hydration mechanism is activated in the Portland cement component, which is then followed by the immediate reaction of slag, resulting in the release of calcium and aluminum ions. This process is completed by the formation of a C-S-H gel through the reaction of slag with alkali hydroxide and Ca (OH)₂ (Neville, 2005). The calcium compound plays a critical role

in slag hydration because it can accelerate the development of cementitious properties. In addition to $\text{Ca}(\text{OH})_2$, gypsum is another slag hydration activator that is released by C_3S and C_2S hydration.



Fig. 8: Ground granulated blast-furnace slag powder (Kosmatka *et al.*, 2003)

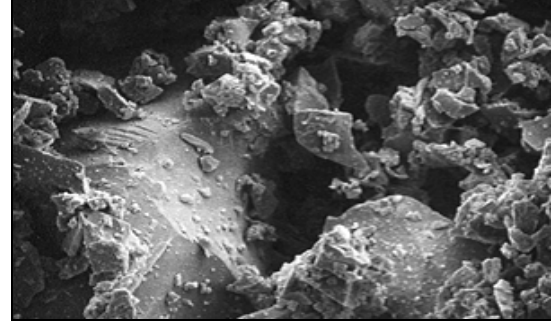


Fig. 9: Scanning electron microscope micrograph of slag particles (Kosmatka *et al.*, 2003)

Table 4: Chemical and physical properties of GGBFS from two different sources of slag (Leung *et al.*, 2009).

Chemical properties	BS 6699, 1992	GGBS –CRC*	GGBS –SG**
Composition %	Requirement		
Pure Silica content (SiO_2)	Not specified	32.6	31.8
Aluminium oxide (Al_2O_3)	Not specified	14.8	14.5
Ferric oxide (Fe_2O_3)	Not specified	0.5	0.4
Calcium oxide (CaO)	Not specified	36.0	37.6
Magnesium oxide (MgO)	< 14%	10.3	9.4
Sulphur oxide (SO_3)	< 2.5%	0.2	0.1
Sodium oxide (Na_2O)	Not specified	0.4	0.3
Potassium oxide (K_2O)	Not specified	0.6	0.5
Loss on ignition (LOI)	< 3.0 %	1.1	0.6
Physical properties	BS 6699:1992	GGBS –CRC*	GGBS –SG**
	Requirement		
specific surface (Fineness) cm^2/g	Not less than 2750	4490	4550
Density (kg/m^3)	Not specified	2880	2910
Soundness (expansion)mm	Not specified	0.0	0.5

* CRC: Dong Run Pai.

** SG:Guangdong Shao Gang.

Metakaolin (MK):

Similar to the other supplementary materials discussed so far, metakaolin (MK) is a pozzolanic material, but, unlike the other materials, it is not a by-product because it is made under carefully controlled conditions (Justice *et al.*, 2005). By heating kaolinitic clay, one the richest natural clay minerals, MK is obtained. For the kaolinite to break down and produce an amorphous material for pozzolanic and latent hydraulic reactivity, a temperature between 650 and 900°C is necessary. It is only within this temperature range that the calcium silicate hydrate (CSH) in cement paste can be produced as the result of the reaction of metakaolin with $\text{Ca}(\text{OH})_2$. Indeed, within the interfacial transition zone (ITZ), which is between the paste fractions and aggregate, this reaction is very crucial because it can enhance the strength in metakaolin concrete (Justice *et al.*, 2005; Poon *et al.*, 2001b). According to (Wild and Khatib, 1997; Bentz and Garboczi, 1991), it is in this region that a high concentration of aligned $\text{Ca}(\text{OH})_2$ crystals can result in increased porosity and lower strength. By reacting to the $\text{Ca}(\text{OH})_2$ produced by cement hydration, metakaolin can dandify the structure of the hydrated cement paste. Compared with silica fume systems, it has been reported that metakaolin systems have a higher initial reactivity due to their higher rates of pozzolanic reaction and $\text{Ca}(\text{OH})_2$ consumption (Poon *et al.*, 2001b). Moreover, metakaolin incorporation may be the cause of the earlier and faster reaction with $\text{Ca}(\text{OH})_2$ (Justice *et al.*, 2005; Wild and Khatib, 1997). Fig. 10 and 11 show a typical metakaolin and a SEM image of its particles, respectively. Metakaolin is used as an additive to concrete (approximately 10% of the cement mass) when very high strength and very low permeability are needed in special applications (Kosmatka *et al.*, 2003). Regarding the physical properties of metakaolin, Table 5 shows that the majority of the particles are less than 16 μm in size, with 3 μm as the smallest size ([http:// www.metakaolin.com](http://www.metakaolin.com)) Table 6 shows the typical chemical properties of metakaolin (Ambroise *et al.*, 1994). The uses of metakaolin for various types of concrete are listed below:

- 1) Glass fiber-reinforced concrete
- 2) Fiber cement and ferrocement products
- 3) High-strength, high-performance, and lightweight concrete
- 4) Precast concrete for architectural, civil, industrial, and structural purposes
- 5) Pool plasters, repair material, mortars and stuccos.



Fig. 10: Metakaolin powder
([http:// www.metakaolin.com.](http://www.metakaolin.com))

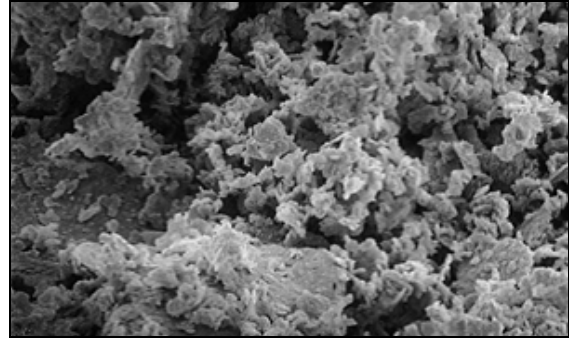


Fig. 11: SEM of metakaolin particles

Table 5: Physical properties of metakaolin ([http:// www.metakaolin.com](http://www.metakaolin.com))

Property	Value
Specific gravity	2.60
Bulk density (g/cm ³)	0.3 to 0.4
Colour	Off-white
Physical form	Powder

Table 6: Chemical composition of metakaolin (Ambrose *et al.*, 1994)

Composition	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	SO ₃	Na ₂ O	L.O.I
By mass %	51.52	40.18	1.23	2.0	0.12	0.53	0.0	0.08	2.01

Silica Fume (SF):

Silica fume (SF) is being used increasingly as a supplementary cementing material for concrete elements and is formed from the smelting of condensed silica fume and volatilized silica in submerged-arc electric furnaces. SF is also known as silica dust, condensed silica fume, micro silica or volatilized silica and is usually a grey or premium white color (Fig. 12). In the ferro-silicon metal industry, this process is usually performed at a temperature of 2000°C, which leads to the reduction of high-purity quartz to silicon. This process, in turn, produces SiO₂ vapors that, in the process of condensing and oxidizing, change to non-crystalline silica. By-product materials that result from the production of silicon metal and ferrosilicon alloys are approximately 75% silicon and contain 85-95% non-crystalline silica. When this amount is reduced to 50% silicon in the production of ferrosilicon alloys, the silica content is reduced and the by-product is less pozzolanic. As shown in Table 7, the type of alloy that is produced depends directly on the SiO₂ content of the silica fume (ACI Committee 234, 1995). In its non-crystalline form, condensed silica fume is basically silicon dioxide. Similar to fly ash, SF has a very fine spherical shape, as shown in Fig. 13. SF can be sold in the form of powder or liquid; however, there is more accessibility to its latter form. Silica fume is usually used at 5% to 10% (by mass) of the total cementitious material (Kosmatka *et al.*, 2003). Similar to RHA, silica fume should also meet ASTM C 1240. The majority of SF particles are very small and less than 1 μm in diameter. The physical properties and chemical composition of silica fume are summarized in Tables 8 and 9, respectively.



Fig. 12: Silica fume powder (Kosmatka *et al.*, 2003)

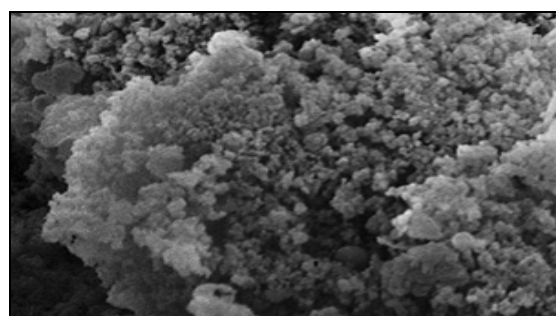


Fig. 13: Scanning electron microscope silica fume particles (Kosmatka *et al.*, 2003)

Table7: Content of silicon dioxide of silica fume produced from different alloy sources (ACI Committee 234, 1995)

Alloy type	SiO ₂ content in SF
50% ferrosilicon	61–84%
75% ferrosilicon	84–91%
Silicon metal	87–98%

Table 8: Typical physical properties of silica fume (Terence, 2005)

Property	Value
Particle size (typical)	< 1µm
Bulk density as-produced	130–430 kg/m ³
slurry	1320–1440 kg/m ³
densified	480–720 kg/m ³
Specific gravity	2.22
Surface area (BET)	13.000–30.000 m ² /kg

Table 9: Chemical composition of silica fumes samples

Oxides %	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	SO ₃	Na ₂ O	L.O.I	
(Sandvik and Gjorv, 1992	92.1	0.5	1.4		0.5	0.3	0.7	-	0.3	2.8
(Titherington and Hooton, 2004	96.7	0.23	0.07		0.31	0.04	0.56	0.17	0.15	2.27
(Yazici, 2008).	96.3	0.89	1.97		0.49	0.96	1.31	0.33	0.42	-

Palm Oil Fuel Ash (POFA):

Palm oil fuel ash (POFA) is another recycled construction material that is generated from palm oil fruit and used to strengthen concrete. POFA can be produced from the burning of palm oil shells and husks in palm oil mills. The result, according to (Sata *et al.*, 2004), is 5% POFA that not only has high pozzolanic properties but also can be used as a supplemental material for cement. The color of palm oil fuel ash changes from gray to dark gray when the percentage of unburned carbon increases, which is shown in Fig.14. However, it is worth mentioning that the quality of this by-product depends significantly on the mill boiler system. For example, if, during the combustion process, the boiler is well-maintained, the resulting fine ash color will be whitish grey. This ash is usually produced at the bottom of the flue tower where it is trapped in a combustion chamber (Zahairi, 1990; SumadiSalihuddin, 1993). Malaysia has been recognized as a leading country in the palm oil industry since the 1960s. In palm oil production, Malaysia ranked first for approximately 10 years (1990-2002) by producing 11,880,000 tons per year (Vijayaraghavan *et al.*, 2007). Since then, POFA has been recognized globally as a great contribution to building construction materials (Zahairi, 1990; SumadiSalihuddin, 1993; Abdul-Awal *et al.* 1997a). Table 10 lists the chemical and physical properties of POFA according to different studies. As shown in Table 10, POFA is mainly composed of silicon dioxide (greater than half) and, therefore, is considered its prime ingredient. It is also important to note that the ASTM C618 general requirement for class N pozzolan is that the ash contains at least 70% SiO₂, Al₂O₃, and Fe₂O₃. According to chemical composition, by and large, most ashes are similar; however, they are considered much finer than ordinary Portland cement. In fact, it has been reported that most POFAs generally meet the standards and can be classified as class C pozzolan. To meet other ASTM C618-84 standards, further grinding is compulsory to reach to the desired fineness. Regarding the physical properties of POFA, Fig. 15 shows that the particles are usually angular, irregular and porous.



Fig. 14: Palm oil fuel ash (Borhan *et al.*, 2010)

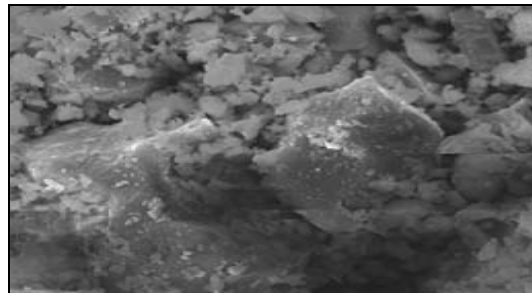


Fig. 15: SEM of ground POFA (Safiuddin *et al.*, 2011)

Table 10: Chemical and physical properties of POFA

Chemical properties, Composition %	Abdul- Awal, and Hussin, 1997 b	Jaturapitakkul <i>et al.</i> , 2007	Chindaprasirt <i>et al.</i> , 2007	Tangchirapat <i>et al.</i> , 2009
Silicon dioxide (SiO ₂)	43.60	57.70	57.80	65.30
Aluminium oxide (Al ₂ O ₃)	11.50	4.50	4.60	2.50
Ferric oxide (Fe ₂ O ₃)	4.70	3.30	3.30	1.90
Calcium oxide (CaO)	8.40	6.50	6.60	6.40
Magnesium oxide (MgO)	4.80	4.20	4.20	3.00
Sulphur oxide (SO ₃)	2.80	0.20	0.30	0.40
Sodium oxide (Na ₂ O)	0.39	0.50	0.50	0.30
Potassium oxide (K ₂ O)	3.50	8.20	8.30	5.70
Loss on ignition (L.O.I.)	18.00	10.50	10.10	10.00
Physical properties,	Abdul- Awal, and Hussin, 1997 b	Jaturapitakkul <i>et al.</i> , 2007	Chindaprasirt <i>et al.</i> , 2007	Tangchirapat <i>et al.</i> , 2009
Specific gravity (g/cm ³)	2.22	2.43	2.43	2.33
Mean particle size (µm)	---	7.40	8.00	10.10
Fineness: passing 45µm(%)	519	m ² /kg (Blaine) 99.0	99.0	98.50

Blended Cement (BC):

Blended cement is a result of blending well-defined proportions of two or more constituent materials with Portland cement to produce better performance (Shondeep and Bonen, 1994). This blended cement concrete mixture can include Portland cement clinker in addition to any of the aforementioned natural pozzolans, such as silica fume and fly ash. It has been reported that a combination of two or more additives exhibits better performance in improving concrete and mortar properties than a single additive (Bagel, 1998; Khan *et al.*, 2000; Sobolev and Yeginobali, 2005; Isaia *et al.*, 2003; Siddique, 2011; Jones *et al.*, 1997). These supplementary materials have also gained a reputation for their environmental benefits because their usage can decrease the carbon dioxide emissions into the atmosphere (Malhotra, 1998). The utilization of these agricultural and industrial waste materials in concrete has also been proven to be satisfactorily beneficial in terms of density, resistivity, durability and strength. However, the long-term strength of some of these by-products may differ because it appears early for some and towards the end for others (Toutanji *et al.*, 2004). These additives are widely used in the U.S. and Europe, and hence, there are more specifications, which are shown in Table 12. To date, various studies have shown that these materials can also be used in precast products and the production of concrete pipes (Sagoe and Mak, 1994).

Table12: Blended cements according to American and European specifications.

Specifications	Name	Portland cement content	Blended minerals
ASTM C595	Granulated blast furnace slag	30–75%	Portland blast furnace slag
	Slag-modified Portland cement	>75%	Granulated blast furnace slag
	Portland pozzolan cement	60–85%	Pozzolan
	Pozzolan modified Portland cement	>85%	Pozzolan
	Slag cement	<30%	Granulated blast furnace slag
EN 197	CEM I Portland cement	95–100%	Minor addition constituents
	CEM II(a) Portland	65–94%	Blast furnace slag, silica fume, composite cementpozzolans (natural or calcined), fly ash, burnt shale, limestone
	CEM III(b) blast furnace cement	5–64%	Blast furnace slag
	CEM IV(c) pozzolanic cement	45–89%	Silica fume, pozzolans, fly ash
	CEM V(d) composite cement	20–64%	Blast furnace slag, pozzolans, fly ash

a Includes sub classification depending on type of blended mineral.

b Includes sub classification depending on content of slag: 36–65%, 66–80%, 81–95%.

c Includes sub classification depending on content of pozzolans (silica fume + pozzolans + fly ash): 11–35%, 36–55%.

d Includes sub classification depending on content of blending minerals (blast furnace slag + pozzolans + fly ash): 36–60%, 62–80%.

Binary Blended Cement (BBC):

Mixing any single cementitious material additive and ordinary Portland cement results in binary blended cement. To improve the performance of traditional cement, this approach was adopted by reducing the Portland cement share in the concrete and replacing it with only one supplementary cementitious material. Similar to any other admixtures, binary blended cement can have advantages and disadvantages. For instance, contrasting influences on the workability and early strength in concrete are among its drawbacks. However, at later ages, binary blended cements can have incredible strength and durability due to the extra hydrated blended system of clinkers and pozzolans.

Multi Blended Cement (MBC):

Although the use of BBC can be dated back many years ago, the use of multi-blended cement with more than two or three supplementary cementitious materials is still new. One reason is the complex nature of the materials that are utilized in the concrete; hence, adequate information is necessary to create a sustainable product. Despite this disadvantage, the advantages of using MBC outweigh those of BBC. A large number of studies have been conducted to analyze the MBC system, and they have come to the conclusion that, due to their effects on strength and durability, their application is more beneficial in structures in hostile environments (Khan *et al.*, 2000). Using more than one waste pozzolan product has also proven to be advantageous technically, economically and environmentally. However, due to their different properties, these waste minerals might show various reactions when they are mixed with water (Toutanji *et al.*, 2004).

The Effect of Supplementary Cementitious Materials on the Fresh Properties of Concrete:

Generally, water is one of the main components of a typical concrete. However, the proportion of the mixing water depends very much on the distribution of the aggregates, their packing effect, and the voids in a solid material. Because, for example, the particle size is not perfectly distributed in a typical concrete mixture,

more water is necessary to maintain a certain workability. For satisfactory consistency in a typical concrete mixture, more water is also necessary for the plasticity of cement paste because of the existence of the electric charge on the surface of the Portland cement that forms the floc and traps the dirt in the water, which increases the slump. To evaluate the fresh properties in the concrete, however, the reliable slump test can be performed. To prevent imperfections, extremely fine supplementary cementitious materials, such as metakaolin and silica fume, are used to increase the water consumption and reduce the slump (Neville, 2005; Habeeb and Mahmud, 2009). However, not all supplementary materials increase the water consumption. For example, ground granulated blast furnace slag and fly ash not only decrease the water demand but are capable of improving the properties of fresh concrete at the same time (Safiuddin and Zain, 2006).

These advantages were shown in the study performed by (Megat Johari *et al.*, 2001); by replacing only 10% of silica fume, there was a significant improvement in the workability of HSC. Currently, it seems that only using superplasticizer in addition to a low water ratio can result in such concrete. Similar to normal concrete, ground granulated blast furnace slag and fly ash can also enhance the workability of HSC. However, it has been reported that, if better effects are desired, higher replacement levels are necessary, which was proven in the study conducted by (Gesoglu *et al.*, 2009). In their study, they could enhance the filling and passing ability of self-compacting concretes by incorporating mineral admixtures that in turn increased the L-box H2/H1 ratio. In their study, when this effect occurred, the T50 slump flow time also increased gradually, particularly in the case of SF. However, only a mixture of PC+FA+Slag could meet the standard criteria of the EFNARC committee in terms of the V-funnel flow time (EFNARC, 2002). By comparing blended cements and a control mixture, (Wu *et al.*, 2002) reported that the water required to produce the slump in the former is 10% less than that required in the latter. However, the water reduction effect was not really significant when blended cements were compared with each other. This finding is somewhat in line with the previously mentioned study that reported that when fly ash was used as a supplementary cementitious material with a 40% rate of replacement, there was an increase in the water reduction effect. The same results can also be obtained by combining fly ash and super plasticizers in HPC. This approach has been supported by (Yin *et al.*, 2002) because the results of their study showed that the incorporation of these two materials improved the workability and the performance of the concrete.

Effect of Supplementary Cementitious Materials on the Hardened Properties of Concrete:

Studies on cement-based composites show that supplementary cementitious materials reduce the porosity of concrete. It has also been well documented that more homogenous hydration products can be produced from either binary or multi-blended cements, which, in fact, can be performed by adding supplementary cementitious materials to fill the existing voids in the cement, hence creating strength and durability. One of the main differences between ordinary Portland cement pastes and blended cement is that, in the former, the pores are continuous while, in the latter, the pores are discontinuous after approximately 28 days of curing. Indeed, as moist curing continues, there is greater hydration in cement, and the result is a concrete with fewer pores and less permeability. Due to hydration, adding pulverized-fuel ash to concrete can not only reduce the water content and dense packing but also increase the hydration and pozzolan reactions, which eventually decrease the permeability of concrete. In the hydration process, the leaching of water-soluble calcium hydroxide can create preformed voids in the hardened concrete. However, the leaching of $\text{Ca}(\text{OH})_2$ can be reduced by adding calcium hydroxide during the pozzolanic reaction. In this reaction, voids can be blocked by C-S-H and added to the density of the concrete, which in turn reduces the permeability. Pozzolanic cement-based materials, such as metakaolin, rice husk ash and silica fume, play a pivotal role in either early- or late-age strength enhancement in concrete (Malhotra, 1993; Safiuddin and Zain, 2006). Nevertheless, for other cement-based materials, such as fly ash and ground granulated blast furnace slag, this strength enhancement does not occur at early ages (Safiuddin and Zain, 2006). When compared with their binary blends, the combination of the aforementioned cementitious materials, fly ash and ground granulated blast furnace slag, happen to have many advantages such as 1) greater compressive strength at all ages (Tan and Pu, 1998), 2) greater microstructure and hydration rate and 3) better resistance to sulfate attack (Li and Zhao, 2003). These results are in line with the findings of (Jianyong and Pei, 1997), which showed that a mixture of FA and GGBFS increases workability by strengthening the splitting tension and rupture.

(Park *et al.*, 2012) concluded that the compressive strength of concrete produced during the winter showed increase of approximately 5% in emissions of CO_2 compared with concrete produced in the season. In addition, they showed that the amount of CO_2 emitted for concrete containing SCM was reduced by as much as 47% compared with concrete without SCM. The reason for this result is due to the replacement of cement and admixtures that have significant amount of carbon dioxide with materials such as fly ash or granulated blast furnace slag, which have a low amount of CO_2 . (Amjad and Salihuddin, 1999) compared two blended cement concrete mixtures of OPC/PFA/SF and OPC/PFA/SF/slag in terms of porosity, strength and oxygen permeability and compared them to OPC. They concluded that the strength values of both OPC/PFA/SF and OPC/PFA/SF/slag were 60 MPa. Nevertheless, OPC achieved higher early strength than either blended cement.

At seven days, this value was more or less the same for all mixes. At 28 days, however, the blended mixture without slag achieved the highest strength value. To be more specific, the strength value of OPC/PFA/SF was 13% higher than that of OPC/PFA/SF/slag and 23% higher than that of OPC. This superiority continued for OPC/PFA/SF at all ages up to 364 days. The higher strength of both blended mixtures was due to the filling of their voids, hence giving the concrete a more dense structure. (Amjad and Salihuddin, 1999) reported a significant decrease in the porosities of two blended cements. Compared to OPC concrete, the porosity was 2.9 times lower at 63 days and 3.8 times lower at 182 days. After 182 and 365 days of hydration, a lower permeability (40% and 50%, respectively) was observed for both blended cements when compared with OPC. Finally, they found similarities in the ways both systems function in terms of seawater curing when exposed to a tidal zone as opposed to OPC.

In another similar approach, (Khan *et al.*, 2000) compared the use of binary and ternary blended cements based on OPC, PFA and SF to develop high-performance mortar. Replacing only 8-12% of SF resulted in the best possible performance and strength and the lowest permeability and porosity for all levels of PFA. Nevertheless, using a binary cement, such as PFA alone, did not lead to any significant differences in terms of the permeability or porosity of mortar. Furthermore, PFA is reported to have an early-age strength development. However, (Khan *et al.*, 2000) reported that if SF is added to a low level of PFA, the early-age strength development can be improved. Indeed, adding more than 35% of PFA (with or without SF) does not have the same type of results compared to OPC. By testing the strength gain, (Menendez *et al.*, 2003) performed another comparison study between Portland cement, limestone and ground granulated blast furnace slag. In the binary combination of ground granulated blast furnace slag and Portland cement, the early-age strength was low, but it improved at later ages. This trend, however, was exactly the opposite for a binary combination of Portland cement and limestone. Finally, high early-age and later-age strengths were observed in the ternary combination. (Li and Ding, 2003) investigated the mechanical and physical properties of PC in terms of super plasticizers used in concrete by comparing a binary cement of PC/MK and a ternary blended cement of PC/MK/slag. Their findings indicate that the proportion of 20-30% ultra-fine slag and 10% MK and PC can have a significant influence on the fluidity of ternary blended cements, which in turn, can improve the compressive strength of the cements at 28 days. By studying the heat reduction in blended cement concrete mixtures of silica fume, fly ash and slag, (Zhang *et al.*, 2002) reported that these minerals can decrease the 3-day hydration heat. Similar to double- and triple-adding approaches, they can also delay the initial time of hydration temperature in HPC.

A comparison of OPC/SF, OPC/PFA, and OPC/slag blends and a combination of SF, slag and PFA was investigated by (Toutanji *et al.*, 2004) in terms of the strength and durability of concrete. In comparison with other combinations, the results show that if the shares for SF, slag and PFA are 10%, 25% and 15%, respectively, better strength and resistance to wet-dry and freeze-thaw can be achieved. For better binder quality and less Portland cement consumption, however, some other researchers suggest adding more than two cementitious materials. (Nehdi *et al.*, 2004) investigated the durability of a quaternary blended cement of GGBFS, SF, RHA and FA (class F). A comparison of self-consolidating cement composed entirely of ordinary Portland cement and one composed of quaternary blended cement shows that the chloride ion penetrability can be remarkably lower in the latter. Arriving at a similar conclusion in terms of strength, (Khatib and Hibbert, 2005) reported that using ternary blends of PC, MK and GGBFS can compensate for the drawbacks that exist in each binary blend. (Anwar, 2006) investigated the tensile and compressive strength and the dynamic elastic and static Young's moduli of a cement with a combination of fly ash and silica fume. The results show that these two supplementary cementitious materials can overcome the shortcomings of one another because silica fume can compensate for the reduction of the early-age strength caused by fly ash.

(Sahmaran *et al.*, 2007) compared the sulfate resistances OPC, SRPC and ternary blended cements. Their results indicated that the expansion of sulfate in ternary blended cements was lower than that in OPC. Similarly, ternary blended cements scored higher than SRPC in terms of performance at room temperature. Somewhat similar to SRPC, ternary blends can have good performances in cycles of hot and cold temperatures. In mortars consisting of OPC, RHA and FA, (Chindapasirt and Rukzon, 2008) investigated the corrosion resistance, porosity and strength. The findings show that ternary blended cements have improved strength at later ages only if the replacement level is low. Although RHA was more influential than FA, both could enhance the corrosion resistance. To reduce the consumption of Portland cement and also increase the quality of blended mortars, they also incorporated palm oil fly ash, which, according to (Chindapasirt and Rukzon, 2008) increases the resistance to chloride penetration. As an effective pozzolan material, RHA ranked first, followed by POFA and FA. For better resistance to chloride penetration and strength, an equal portion of RHA and FA or POFA and FA is suggested by (Chindapasirt and Rukzon, 2008). (Chindapasirt *et al.*, 2008) also conducted another study on blended Portland cement mortar containing POFA, RHA and FA. In that study, they wanted to observe the extent to which carbon dioxide affects the penetration of chloride and its ion diffusion coefficient in the aforementioned blended mortar. It can be inferred from their findings that, in chloride ingress situations, this mixture could improve the performance of blended Portland cement mortar. Similar to their previous study, among the three materials considered, RHA was considered the most effective pozzolan, followed by POFA and

FA. The amount of calcium hydroxide and the pH level of the mortar were also reduced due to the application of pozzolan. Mortar is vulnerable to chloride attack and decreases the resistance to chloride penetration, while carbon dioxide increases it. Finally, (Chindaprasirt *et al.*, 2008) reported that the mixture of these blended pozzolans can result in the reduction of resistance to chloride penetration in mortar after exposure to carbon dioxide. In another study, conducted by (Penga *et al.*, 2009), the proportions of fly ash, slag and silica fume were increased to achieve better compressive strength. These proportions were reported as 10% for FA, 17% for slag and 15% SF.

(Guneyisi *et al.*, 2010) investigated the shrinkage of high-performance concrete with a quaternary blended binder containing silica fume, fly ash, slag and metakaolin. Due to the accumulative effects of the multi binder on high-performance concrete, the compressive strength was significantly reduced. A high portion of fly ash and silica fume is reported to be the key factor affecting the drying shrinkage. Compared to control concrete, specimens of metakaolin and silica fume have a greater compressive strength. The permeability, porosity, compressive strength and resistance to chemical agents of multi-blended mortars containing fly ash, silica fume, rice husk ash and POFA were investigated by (Borhan *et al.*, 2010). The results indicate that, although the final-age strength of both the control mortars and the multi-blended concrete were approximately the same, at early ages, this strength was 20% lower for the multi-blended concrete. Nevertheless, the multi-blended concrete outperformed the control mortars in terms of low permeability. Furthermore, when exposed to chemical attack, the multi-blended cement mortars showed a greater resistance and higher durability.

Conclusions:

As the title of this paper indicates, a review approach was adopted to provide new insights into the influence of the incorporation of supplementary cementitious materials on the properties of mortar and concrete. This paper also aimed to provide several valuable insights on improving the quality of concretes and reduce their environmental impact. These points are summarized below.

1) The supplementary cementitious materials examined thus far, SF, FA, RHA, and MK, are alike in that they have been proven to be invaluable materials to enhance the performance of concrete. Each one of these materials can positively influence the concrete performance in terms of the early-age properties, late hardening, drying shrinkage, or compressive and tensile strengths.

2) The utilization of supplementary cementitious materials in either cement or concrete can compensate for environmental, technical and economic issues caused by cement production. Most of these supplementary cementing materials are by-products, and their inclusion serves as an invaluable means to protect environmental resources, which may result in more viable constructions in the future.

3) Because these supplementary cementitious materials are sourced differently, their effects on cement or concrete also vary. Hence, different combinations and quantities can have various effects on concrete. Therefore, the best way to guarantee the most favorable results is by using mix designs and trial batching. Furthermore, favorable results can also be achieved by using information from other projects.

4) The use of supplementary cementitious materials in concrete is generally limited by state specifications that are not actually performance-based. Thus, this study recommends updating these specifications for better performance-based results. Guidelines will also be beneficial if they follow the trends for various combinations of supplementary cementitious materials.

5) More broadly, it is recommended that further research be undertaken on more industrial and agro-waste minerals and by-products as supplementary cementitious materials. The more these materials are added to concrete, the less cement is used, which in turn, can lead to the environmental benefits of lower emission of CO₂.

6) In a nutshell, there is still a paucity of research in terms of crack formation, brittle behavior, corrosion time initiation, shrinkage, passivity and carbon dioxide absorption. More information on these issues would help to create a greater quality concrete while using a high volume of supplementary cementitious material.

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