

A REVIEW OF THE PAST, PRESENT AND FUTURE OF CONCRETE ADMIXTURES

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ABSTRACT

Purpose: Sustainable environment could be achieved with constructions using concrete that generates little heat. One way to achieve this is by using water reducing admixtures. More so, there is coming a time in some parts of Nigeria where normal strength concrete will be less required due to unavailable land space and high-rise buildings will require high or ultra-high strength concrete, produced with the aid of water reducing admixtures. Therefore, this paper reviews the past, present and future of concrete admixtures (water reducing admixtures) and the need to develop effective eco-friendly (natural) admixtures.

Keywords: Admixture; Water Reducers; Superplasticizer; Compatibility; CNSL.

1. INTRODUCTION

Concrete is a composite material consisting of a mixture of cement, sand, granite/gravel, water and/ or admixture. It has a historical background similar to concrete admixture. It has a historical background similar to concrete admixture. The history of concrete has always hinged on producing concrete with significant strength and to overcome some difficult situation such as hot or cold weather placements, pumping requirements, early-strength requirement or very low water-cement ratio specifications (Oladiran *et. al.*, 2012), during use of the concrete.

For instance, according to Radic, *et. al.*, (2008), around 10,000 BC in Israel, there was a reaction between limestone and oil shale during a spontaneous combustion to form a natural deposit of cement compounds. Similarly, evidence of burning gypsum was found at Cata Huyuk, Asia around 9000BC. In addition, in Egypt, about 3,000BC, meteorite, the oldest findings of iron was discovered. Evidence showed that ancient Egyptian used mud mixed with straw to bind dried bricks. Ancient Egyptians also used gypsum mortars and mortars of lime in the construction of the pyramids (Radic, *et. al.*, 2008). Greeks, Cretans and Cypriots used lime mortars which were much harder than the Romans' mortar. The Chinese Great wall was built using cementitious materials, the Babylonians and Assyrian used bitumen as binding agents during road construction (Burn, 2005).

Studies such as Dodson (1990); Rixom & Mailvaganam (1999); Ramachandran (2002); Nevile & Brooks (2010) and Aiicin & Flatt (2016) believed that the earliest recorded use of modern concrete was during Roman periods spanning between 300BC to 476AD a range of more than 700years. The Romans used pozzolana cement from

Pozzuoli, Italy, near Mt. Vesuvius to build the Appian Way, Romans bath, the coliseum, Pantheon and the Pan du Gard aqueduct in southern France. They used lime volcanic ash (a pozzolana materials) and admixtures such as Animal fat, milk, eggs and blood to improve the properties of concrete.

2. LITERATURE REVIEW

2.1. History of Cement

During the first industrial revolution, between 1760 to sometimes around 1840 in Europe and United States, Portland cement was discovered. In Europe, the hydraulic lime and volcanic ash first developed by John Smeaton in 1765 when he erected the Eddystone, Lighthouse, off the coast of Plymouth, Devon, England (Dickinson, 1939). Also, in the United States, cement known as natural cement/ Rosendale natural cement was in used (Aiicin & Eberhardt, 2016).

Thereafter, at about 1800 in France and England, a material obtained by burning modules of clayey limestone was used. The invention of Portland cement is attributed to Joseph Aspdin of Leeds, Yorkshire, England, who in 1824 took out a patent for a material that was produced from a synthesis of clay with limestone. It was called Portland cement because the products resemble a Portland stone obtained in Portland (Courtland, 2011). The adoption of Portland cement spread to all countries like wide fire and by the end of 20th century, it was used world over. China and India became the leading manufacturers as at 21st century (Nick & Kenton, 2013).

2.2. History of Admixture

According to Aiicin & Eberhardt (2016), a faulty bearing of a grinding mill had been releasing some heaving oil which resulted in the discovering of the beneficial effect of air entraining admixture (Mindess *et.al.*, 2003). The discovery originated from the use of American Naturally occurring materials called Rosendale Natural cement. American cement had been in used since 1818 after natural cement rock was discovered by Convass White in Fayetteville, New York. This cement was produced from Marl limestone or argillaceous limestone (a limestone containing clay) which were burnt between 800 and 1100°C before the wide spread use of Portland cement across the world nearly put the Rosendale Natural cement out of use because it hardened too fast (Eckel & Burchard, 1913). Due to freezing and thawing cycle in the New York state, it was observed that salt scaling does occurred on the concrete produced using Portland cement. This led to the degrading of the concrete overtime (Jackson, 1944), whereas structures produced using Rosendale cement remained unchanged with adequate strength.

An engineer named Bertrand H. Wait, started experimenting on blends of Portland cement in 1933. He was able to make concretes with a scaling resistance against freezing and thawing using salt solution that was 12 times greater than that of concrete made from pure Portland cement (Holbrook, 1941).

According to Aiicin & Eberhardt (2016), the reason for the greater resistance of these blended cements against freezing and thawing was not clear. However, it could have been a consequence of the natural cement itself or of the fact that one out of the two Rosendale cements that were used in the New York contains a small amount of beef tallow as a grinding aid. At the end of the 1930s, the Portland Cement Association initiated intensive studies on the effect of introducing small amounts of tallow, fish oil, and stearate resins as air-entraining agents.

The second story of admixture discovery according to Dodson (1990), an Engineer from the Department of Transportation (DOT) wanted to mark the middle of the first three concrete lanes in the United State to avoid an accident on the highway. The DOT Engineer instructed the contractor to do the marking; however, the Engineer was not impressed with the black carbon used. The Engineer therefore instructed the dispersion of the black carbon. After some time, the Engineer discovered that the chemicals (polysulfonate salt) used for the dispersion, had improved the strength and durability of the portion of the road where the chemicals were applied.

2.3. History of Water Reducing Admixture

The earliest known use of water-reducing admixture involved the use of small amount of organic materials to increase the fluidity of cement in 1932 (Rixom & Mailvaganam, 1999). The organic materials adopted were polymerized naphthalene formaldehyde sulfonate salts. This was followed by the use of Lignosulfate in the 1930s to early 1940s. The Lignosulfates formed the basis of almost all the available water reducing admixture until 1950s when the Hydroxycarboxylic acid salts were developed.

High Range Water Reducing Admixture also known as superplasticizers were first developed in Japan by Kenichi Hattori in 1964. The superplasticizers produced contained beta naphthalene sulfonates and modified lignosolphonate. In the same year, the Melamine formaldehyde condensate in Germany (Jerath & Yamane, 1987) was developed. After a decade, the use of superplasticizer reached the American continent in 1974 (Sidney, 2011). In the year 1987, the latest generation was introduced to North America market called Poly carboxylate ether (Jayasree *et. al.*, 2011).

2.4. Water Reducing Admixture

The water-reducing admixtures allow a reduction in the water cement ratio at a given workability without significantly affecting the setting characteristics of the concrete. In practice, this effect can be utilized in three ways:

- i. By the addition of admixture with a reduction in the water-cement ratio, a concrete having the same workability as the control concrete can be obtained with unconfined compressive strengths at all ages which exceed those of the control.
- ii. If the admixture is added directly to a concrete as part of the gauging water with no other changes to the mix proportions, a concrete possessing similar strength development characteristics is obtained, yet having a greater workability than the control concrete.
- iii. A concrete with similar workability and strength development characteristics can be obtained at lower cement than a control concrete without adversely affecting the durability or engineering properties of concrete (Rixom & Mailvaganam, 1999).

Water reducers consist of Ca, Na, or NH₄ salts of lignosulfonic acid, Na, NH₄ or triethanolamine salts hydroxycarboxylic acid, and carbohydrates. Lignosulfonates containing (OH), (COOH) and (SO₃H) groups are more widely used than others. Hydroxycarboxylic acids such as citric acid, tartaric acid, salicylic acid, heptonic acid, saccharic acid and gluconic acid based admixtures contain (OH) and (COOH) groups. Gluconic acid-based admixtures are used extensively. Carbohydrates include glucose, sucrose or hydroxylated polymers obtained by partial hydrolysis of saccharides (Collepardi, 1994).

The role of water reducers (normal, accelerating or retarding) in terms of their effect on the hydration of cement is similar to that of accelerators and retarders (Ramachandran, 2002). The water reducers used in 1930s when the lignosulphate was discovered still remain in use till today, although, several others have been discovered. They include acrylates, methacrylate, polymers, styrene copolymer, ethylene, pentene, digested product of cellulose, sulfite yeast mash based products (waste product from fish oil processing) corn cobs, straw and sunflower treated with sulfuric acid (Flatt and Schober, 2012).

In 20th century, according to Ramachandran (2002), the water reducing admixture in used are prepared from hydrolysis of polysaccharide or oxalic acid, dicyclo-pentadiene derived acrylic resins condensation product with Na-carbazol disulfonate + calcium chloride, maleic anhydride styrene copolymer and alkoxylated alcohol or phenols. Many patents on water reducer in the 21st century have been produced from synthesized lignosulphate salts combined with other earlier discoveries.

2.5. High Range Water Reducing Admixture

Superplasticizers belong to a class of water reducers chemically different from the normal water reducers and capable of reducing water contents by about 30%. The admixtures belonging to this class are variously known as superplasticizers, superfluidizers, superfluidifiers, super water reducers, or high range water reducers (Ramachandran, 2002). Superplasticizers are used for the following;

- i. To produce concrete having very low water cement ratio to attain high early and ultimate strengths at much reduced permeability to moisture and salts.
- ii. To produce concrete having very high flowability where the admixture is added to the mix with no alteration in water-cement ratio to produce slumps more than 180mm (Rixom & Mailvaganam, 1999).

The superplasticizers are broadly classified into four generations: sulfonated melamine-formaldehyde condensate (SMF); sulfonated naphthalene-formaldehyde condensate (SNF); modified lignosulfonates (MLS); Polycarboxylic ether (PCE); and others including sulfonic acid esters, polyacrylates, polystyrene sulfonates, etc. Blends of different superplasticizers have also been developed (Jayasree *et. al.*, 2011).

Examples include, Naphthalene sulfuric acid-isocyanuric acid condensates, micropellets containing unsaturated dicarboxylic and anhydride, copolymer of styrene and maleic acid graft polymerized with a conventional lignin or Naphthalene Sulfonic acid/ Naphthalene sulfonic acid copolymer and condensate of melamine formaldehyde condensate (Ramachandran, 2002).

From the late 20th century to the end of 20th century, the superplasticizer developments have focused on producing formulation to control slump loss (a problem of compatibility), increase in fluidity, and strength of concrete. The formulation includes; azo type superplasticizers, indene carboxylic acid copolymer product from isobutylenes maleic anhydride copolymer and laurylamine. Other formulations produced up to date are synthetic (Ramachandran 2002; Aiicin & Flatt, 2016).

2.6. The Current Trend in Concrete Admixture

Compatibility Issues

Since the early 1960s when superplasticizer was first introduced (Evangeline & Neelamegam, 2015), there has been a problem of incompatibility between cement and the admixture. It can either be cement and superplasticizer incompatibility or incompatibility between superplasticizer and other admixtures or incompatibility between

superplasticizer and supplementary cementitious materials and cement (Tiji & Liji, 2016).

According to Tiji & Liji, (2016), incompatibility is a term used to describe the adverse effect on performance when a specific combination of cement and superplasticizer is used. It could be due to the effect of the chemical structure of superplasticizer, admixture type and dosage, cement composition and fineness, the effect of calcium aluminates in cement, role of calcium sulphates, and role of alkali (Jayasree *et.al.*, 2011). The factors affecting the compatibility between cement and superplasticizer are discussed below:

- i. **Effect of Chemical Structure of Superplasticizer:** The chemical structure of the superplasticizer affects the ability of the superplasticizer to blend with cement. For instance, the type and dosage, degree of polymerization, degree of sulphonation, the position of functional group in the benzene ring, the molecular weight distribution of the polymer, the addition rates and the time of addition of superplasticizer affect its interaction with cement (Tiji & Liji, 2016).
- ii. **Admixture Type and Dosage:** There are different types of superplasticizers, each having its chemical composition and structure. The reaction of lignosulphonate will be different from sulphonated melamine and as well different from sulphonated naphthalene formaldehyde, and polycarboxylate ether (Jayasree, 2011). Aiicin (1998) and Agullo *et.al.*, (1999) stated that for all superplasticizers, the rate of increase of fluidity of the paste or workability of concrete decreases as the dosage increases until there is no significant increase in the fluidity. This dosage point is often refers to as saturation point and it is unpredictable when there is a presence of pozzolana or other admixture type (Aiicin & Flatt, 2016)
- iii. **Time of Addition of Superplasticizer:** According to Uchikawa *et. al.*, (1992) and Aiad (2003), the delayed addition of sulphonated melamine formaldehyde and sulphonated Naphthalene helps in retaining its fluidity. The rate of adsorption is reduced when hydration had taking place compared to during or before hydration.
- iv. **Cement Composition and Finess:** Most superplasticizer carries an anionic charge which easily reacts with the celite and ferites compound in cement. Therefore, the higher the celite and ferrite content the better the rate of adsorption of cement. Also, the finer the cement, the higher the specific surface area and consequently the water demanded, given that workability is expected to be higher (Jayasree *et.al.*, 2011).
- v. **The Role of Calcium Sulphate:** In the early stages of cement hydration, the reaction that dominates is those of the Alite (C_3S) compound with the water to produce calcium hydroxide and Calcium Silicate Hydrate and of Celite (C_3A) with gypsum to produce ettringite (Ramachandran, 2002). It is at this stage that the interaction of the superplasticizer occurs. Superplasticizers with Sulphonate compete with calcium sulphonate released from gypsum for reaction with aluminate. When the solubility of the calcium sulphates is low, the superplasticizer molecules tends to get adsorbed first on the aluminate, thus preventing the normal setting reaction involving the formation of ettringite. In other to prevent this, the solubility of sulphates is important (Jayasree *et.al.*, 2011).
- vi. **Role of Alkalis:** Alkali in cement is essential for accelerating alite hydration. However, excess alkali could have adverse effects, one of them being the alkali aggregate reaction. Cement with high alkali content causes a workability problem

in concrete. The alkali also contributes to the low rheology of superplasticizer and cement (Jayasree *et.al.*, 2011).

The Need for Natural Admixture to Solve Incompatibility

Superplasticizers and cement incompatibility has always been an issue from the time the last generation of superplasticizer has been discovered. Several authors have focused on using chemical superplasticizers (MLS, SNF, SMF and PCE based superplasticizers) to understudy and address incompatibility (Banfill, 1979; Roy & Asaga, 1980; Bjornstrom & Chandra, 2003; Sindhu *et. al.*, 2017). However, little attention has been put to using superplasticizer from natural plant.

Table 1: Previous studies on materials used in addressing incompatibility

S/N	Authors Name	Title of Paper	Materials Used	Remarks
1.	Roy & Asaga (1980)	Rheological Properties of Cement Pastes, Effects of Time on Viscometric Properties of Mixes Containing superplasticizers.	Two Cement and SNF and SMF superplasticizer	Portland Cement and Chemical Admixture.
2.	Masood & Agarwal (1994)	Effect of Various Superplasticizers on Rheological Properties of Cement Paste & Mortar.	Cement, Seven Superplasticizer & CNSL Super Plasticizer.	Portland Cement, Chemical and Natural Admixture.
3.	Chiara <i>et.al.</i> , (2001)	The Influence of Mineral Admixture on the Rheology of Cement.	SNF, Natural Admixture (Fly ash, Metakaolin & Silica fume) & Cement.	Chemical Admixture, Pozzolana and Portland Cement.
4.	Aiad & Heikal (2003)	Effect of Superplasticizer on the Rheological Properties of Blended Cement Paste Containing Silica Fume.	SNF & PCE. Superplasticizer and Blended Cement of Silica Fume.	Chemical Admixture and Blended Cement.
5.	Bjornstrom & Chandra (2003)	Effect of Superplasticizer on the Rheological Properties of Cements.	MLS, SNF, SMF & PCE Superplasticizer with Cement.	Chemical Admixture and Portland Cement.
6.	Gad <i>et. al.</i> , (2005)	Rheological Properties of Different Cement Pastes made with Different Admixture.	PC, SRC & Blended Cement of High Slag& Fly ash and MLS, SNF & PCE.	Blended Cement, PC,SRC& Chemical Admixture.
7.	Olga <i>et. al.</i> , (2012)	Compatibilty Between Superplasticizer Admixture & Cement with Mineral Additions.	Blended Cement of Limestone, Fly Ash& Silica fume, MLS SMF, SNF & PCE.	Blended Cement, and Chemical Admixture.
8.	Shah <i>et. al.</i> , (2013)	Effect of HRWR on the Properties and Strength Development Characteristics of Fresh and Hardened concrete.	PC & 2PCE.	Portland Cement, and Chemical Admixture.
9.	Tiji & Liji (2016)	Compatibility Studies of an Admixture with Different Cement Brand of Varying Chemical Composition for SCC.	Different Cement Type & PCE	Portland Cement, and Chemical Admixture.
10.	Sindhu <i>et. al.</i> , (2017)	Studies on Rheological Properties of Superplasticizer on Portland Pozzolana Cement Paste.	PPC (Fly ash) and MLS, SMF, SNF and PCE.	PPC and Chemical Admixture

Masood & Agarwal (1994) compared the rheology of different superplasticizer including the superplasticizer developed from natural plant extract (CNSL). Although, according to Flatt & Houst (2001), Ramachandran (2002) and Marchon *et. al.* (2016), rheology of superplasticizer and cement cannot fully explain the chemistry of relationship between superplasticizer and cement, there is need to study the adsorption and zeta potential together with their performance for full understanding.

Recently, attention has been focused on the benefit of natural additions in form of pozzolana could do in addressing incompatibility (Chiara *et.al.*, 2001; Aiad & Heikal, 2003; Gad *et.al.*, 2005; Olga *et.al.*, 2012 and Sindhu *et. al.*, 2017). This has led authors into incorporating natural additives (such as fly ash, limestones and silica fume) to cement and superplasticizer, and their rheology, adsorption and zeta potential determined. The results obtained were promising.

However, there is need to study using superplasticizer produced from natural plant and the rheology, adsorption and zeta potential determine accordingly. A research by the author, on the use of naturally developed superplasticizer to address cement superplasticizer incompatibility is underway. Table 1 shows the materials used as admixture and the type of cement used to study cement superplasticizers incompatibility. Only in one case was a natural admixture used. In understanding compatibility, Flatt and Houst (2001) proposes three methods; rheology, adsorption, and zeta potential. The methods used by the ten authors in relations to the proposed are discussed in the Table 2. Only two authors were able to use the three methods.

Table 2: Previous studies on the methods used in addressing incompatibility

S/N	Authors Name	Title of Paper	Methods Used	Remarks
1.	Roy & Asaga (1980)	Rheological Properties of Cement Pastes, Effects of Time on Viscometric Properties of Mixes Containing superplasticizers.	Coaxial Cylinder Viscometer	Rheology Test
2.	Masood & Agarwal (1994)	Effect of Various Superplasticizers on Rheological Properties of Cement Paste & Mortar.	Brookfield DV-II Model Viscometer	Rheology Test
3.	Chiara <i>et.al.</i> , (2001)	The Influence of Mineral Admixture on the Rheology of Cement.	Marsh Cone Apparatus & Minislump.	Rheology Test
4.	Aiad & Heikal (2003)	Effect of Superplasticizer on the Rheological Properties of Blended Cement Paste Containing Silica Fume.	Rheotest Cell.	Rheology Test
5.	Bjornstrom & Chandra (2003)	Effect of Superplasticizer on the Rheological Properties of Cements.	Rheology, Adsorption & Zeta potential.	Rheology, Adsorption & Zeta potential.
6.	Gad <i>et. al.</i> , (2005)	Rheological Properties of Different Cement Pastes made with Different Admixture.	Rotating Coaxial Cylinder Viscometer.	Rheology Test.
7.	Olga <i>et. al.</i> , (2012)	Compatibility Between Superplasticizer Admixture & Cement with Mineral Additions.	Haake Rheowin Pro RV1 Rotational Viscometer, Total Organic Carbon & Smoluchowski Approximation.	Rheology, Adsorption & Zeta Potential
8.	Shah <i>et. al.</i> , (2013)	Effect of HRWR on the Properties and Strength Development Characteristics of Fresh and Hardened concrete.	Nil	No test
9.	Tiji & Liji (2016)	Compatibility Studies of an Admixture with Different Cement Brand of Varying Chemical Composition for SCC.	Marsh Cone Apparatus	Rheology Test
10.	Sindhu <i>et. al.</i> , (2017)	Studies on Rheological Properties of Superplasticizer on Portland Pozzolana Cement Paste.	Coaxial Cylinder Viscometer (Brookfield DV-II) & Marsh Cone	Rheology Test

2.7. The Future of Concrete Admixture

There is limited resource in virtually all countries around the world and it is a challenge that we have to face this century. Construction consumes about 40% of natural resources, and sustainable construction therefore represents a major societal concern (Aïtcin & Mindess, 2011). Clinker consumption could be made sustainable if effectively used.

However, the quantities of concrete required daily are extremely high. This means that concrete performance must be fully exploited. This infers significant increases in the number and dosage of admixtures used.

In contrast, the use of locally available resources would significantly contribute to this development. Its development will aid the understanding that valuably complements the know-how of enlightened formulators and practitioners.

3. CONCLUSION

This paper has shown the past, present and the future of admixture (that is; water reducers) and concrete in general. It has shown that the discovery of cement has led to insatiable desire to improve it. Admixture has been useful in this regard, which has led to discoveries of different type of admixtures and in specific, different generation of superplasticizer. More so, the amount of cement production needs to be reduced, therefore requiring maximum performance whenever concrete is produced. However, this is achievable with maximum and effective use of admixture.

Compatibility has posed a challenge to the maximum and effective use of admixture (superplasticizer). It calls for a new approach to addressing issues of compatibility. One of such new approaches is to encourage researchers into the development of admixture from naturally occurring materials. Sustainable environment is the future and it has begun.

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