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Recent advances in understanding the role of supplementary cementitious materials in concrete

Maria C.G. Juenger [a,*], Rafat Siddique [b]

[a] The University of Texas at Austin, 301 E. Dean Keeton St., Austin, TX 78712, USA
[b] Thapar University, P.O. Box 32, 147004 Patiala, India

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ABSTRACT

Supplementary cementitious materials (SCMs) are commonly used in concrete mixtures as a replacement of a portion of clinker in cement or as a replacement of a portion of cement in concrete. This practice is favorable to the industry, generally resulting in concrete with lower cost, lower environmental impact, higher long-term strength, and improved long-term durability. SCMs have been used in Portland cement concrete for decades and many of their effects are well-understood. Most recent research on SCMs has focused on a few areas: exploring new materials, increasing replacement amounts, developing better test methods, treating or modifying materials, and using additives (e.g. limestone or nanosilica) to improve performance. The advances in knowledge provided by research in these areas are reviewed in this paper, emphasizing the impact of the research on the field.

1. Introduction

Supplementary cementitious materials (SCMs), including fly ash, ground granulated blast furnace slag, silica fume, calcined clays and natural pozzolans, are commonly blended with clinker to make portland cement or used as a replacement for a portion of portland cement in concrete. The practice of using SCMs is increasing, with the world average percent clinker in cement having decreased from 85% in 2003 to 77% in 2010, and it is projected to further decrease to 71% in the future [1]. In the U.S., SCMs are usually added to concrete rather than blended with clinker, and currently more than 60% of ready-mixed concrete uses SCMs [2].

While fly ash and ground-granulated blast furnace slag represent the majority of SCMs used, there is a shift to embrace other materials, which is driven by many factors, including supply-and-demand concerns. In 2011, 3.6 billion tons of cement were produced worldwide [3], and this is projected to rise to 5.8 billion tons by 2050 [4]. A way to meet this rising demand is to continue increasing the use of SCMs in concrete. It is understood that only part of this demand can be met through the use of fly ash and slag, since the annual global productions of these materials are approximately 1 billion tons and 360 million tons, respectively [5,6]. Therefore, the focus of much of the recent research on SCMs has been on the exploration of alternative SCMs and their performance in concrete. While itemizing newly discovered alternative SCMs is not the goal of this review paper, research on these materials is discussed when findings are applicable to a wider range of SCMs.

This paper summarizes the advances achieved in the past four years in our understanding of SCM use in concrete. One of the primary reasons for SCM use is to reduce the environmental impact of concrete, and recent publications on this topic are reviewed first. Identifying appropriate new materials, maximizing their use, and improving their performance can best be achieved through appropriate material characterization and tests for pozzolanicity, which are reviewed next. Correspondingly, there have been significant advances in the pre-treatment of SCMs for improved reactivity or additives to improve mixture performance, particularly at the nanoscale. The interactions of SCMs with Portland cement is addressed in terms of the impact on early hydration, fresh state properties, mechanical properties, and long-term durability. Lastly, the role of SCMs in ultra-high performance concrete, is reviewed, focusing on the impact of these materials on long-term properties.

2. The role of SCMs in sustainable concrete production

While the use of SCMs in concrete in relatively small amounts (5–20% replacement of clinker) is often driven by economics and improvements in the long-term mechanical properties and durability of concrete, the impetus to replace an increasing percentage of clinker with SCMs often comes from pressure on the industry to reduce CO_2 emissions from concrete production. Often these high volume clinker replacements result in losses in performance at early ages, driving research into balancing sustainability and performance and finding means of performance prediction.

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* Corresponding author. Tel.: +1 512 232 3593.
E-mail addresses: mjuenger@mail.utexas.edu (M.C.G. Juenger), rsiddique@thapar.edu (R. Siddique).

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Since the manufacture of Portland cement is dramatically more energy-intensive and greenhouse gas-emitting than the production of other concrete components, a driver in the production of sustainable concrete is to minimize cement content. Most of the information on the environmental impacts of concrete components comes from life cycle analyses (LCA), which have been extended to include SCMs and admixtures to quantitatively compare them to Portland cement. However, when evaluating SCMs, careful attention needs to be paid to the classification of the materials for the purpose of allocation. For example, Van den Heede and De Belie [7] explained that fly ash and blast furnace slag are often considered to be "avoided waste," with no allocation of the power used for their production, making them attractive concrete components from an environmental perspective. A potential EU re-classification of these materials as "useful by-products" would demand allocation of a portion of the power and pollution used in their production, resulting in a much higher environmental load, exceeding that of cement. In this situation, one could adopt an economic allocation approach in lieu of a typical mass allocation in order to consider by-product SCM use as sustainable [7]. Petek Gursel et al. [8] also recently reviewed the methodologies used in LCAs and further pinpointed inconsistencies in assumptions and analyses and how these lead to errors in accounting. They also argued that LCAs must account for the drying, grinding, and preparation of SCMs, as this energy input is not negligible.

Assuming that increased SCM content and decreased cement content make sustainable concrete, there have been several recent studies presenting optimization methods for this strategy. For example, Hooton and Bickley [9] highlighted SCM use coupled with aggregate gradation optimization, admixtures, and fillers, enabling the reduction of the Portland cement content for a typical bridge deck concrete mixture from 12 vol.% to 3 vol.% (Fig. 1). Optimization of particle size distributions of powders in concrete, including cement, SCMs, and fillers, can be used to maximize SCM content with minimal negative impact on early age properties. For example Bentz et al. [10] used this method to design blended cements with 35 vol.% fly ash having identical early and later age mechanical properties to a straight cement mixture. Zhang et al. [11] used a gap-graded powder, consisting of fillers, SCMs, and Portland cement, with the materials ground and classified to fit the desired particle size distribution. They recommended that the most efficient use of materials for optimal early and late properties, economics, and environmental impact is to have Portland cement in the 8–24 μm fraction, with fine, reactive SCMs and fillers occupying the finer fraction and coarse, less reactive SCMs occupying the coarser fraction.

One of the challenges that arises from increasing the volume of SCM replacement is the prediction of mechanical properties and durability in these systems. To this end, Gruyaert et al. [12] explored the application of an "equivalent performance" metric and "k-value concept," both of which were found to be challenging to apply to a wide range of performance metrics. This is of particular concern when attempting to replace Portland cement with high volumes of SCMs, as some standards and legislation use these concepts to limit the maximum replacement by SCMs [7]. With all of these strategies, one should not forget that the most foolproof way to design for sustainability is to design for long-term durability, as argued by Hooton and Bickley [9], not only with respect to material choices, but with respect to construction processes; one should not neglect the construction processes that lead to improved durability.

3. Material characterization

There is an increasing variety of SCMs being investigated, with much of the recently published literature on SCMs focusing on trial testing of new potential SCMs from waste-streams and natural sources. Some of the challenges with the introduction of so many new materials are finding methods to appropriately characterize them and recognizing the limitations of some existing methods that were developed for other materials [13]. Some of the characteristics of interest when evaluating SCMs are their physical properties, including particle size distribution and specific surface area, and their chemical properties, including oxide composition, phase composition, and amorphous content. All of these affect pozzolanic reactivity, while some also affect interaction with cement hydration and water demand.

Particle size distribution is typically measured by laser diffraction techniques, which represent an advancement over sieving and other standardized methods, but which should be approached cautiously with SCMs because of challenges relating to the agglomeration, refractive index determination, etc. [14]. Surface area is typically measured by nitrogen sorption using the Brunauer–Emmett–Teller (BET) model for analysis, which is more reliable for SCMs than the air-permeability tests standardized for Portland cement [14]. Nitrogen sorption can also be used to assess more than the specific surface area, when additional models such as the Barrett–Joyner–Hallenda (BJH) models and t-plots are used. The BJH model was used by Quercia et al. [15] to determine the pore size distribution in a nanosilica using t-plots to differentiate between external and internal surface area. This type of information is useful when trying to understand the role of internal versus external surface area on workability and reactivity. Further, the t-plot analysis can be used to examine the shape and type of pores [15], providing extensive characterization for porous SCMs.

Oxide composition is important for pozzolanicity, with high silica and alumina content generally agreed to contribute to the pozzolanic reaction. However, recent work by Walker & Pavía [16] suggests that the amorphous content outweighs the silica content as a predictor of long-term pozzolanic activity, which is not surprising since crystalline

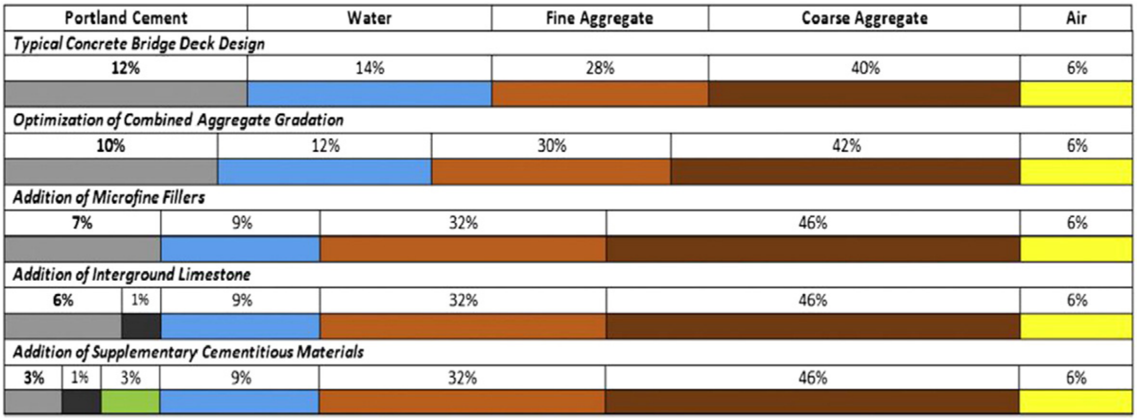

Fig. 1. Illustration of potential reductions in Portland cement content through various methods, including SCM use [9].