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A unified approach for mix design of concrete containing supplementary cementitious materials based on reactivity moduli

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ABSTRACT

A growing awareness of the environmental impact of concrete production and usage has resulted in an expansion in the use of supplementary cementitious materials (SCMs) in ordinary Portland cement (OPC) based concrete. Research has produced a significant number of mix designs based on a large range of SCMs at different replacement ratios. Significantly, the results of each study are not always transferable, as unlike OPC which is generally highly controlled chemically, SCMs of a given type are generally characterised by a broad chemical makeup. In this study, the fundamental chemical and mineralogical compositions of wide-range of cementitious materials are inspected and the critical elements (i.e. oxides) are identified. The hydraulic activity of individual and blended binders are assessed and quantified based on the oxides identified leading to the quantification of a reactivity index. It is then shown how the reactivity index can be used as the basis of mix design for unitary and blended binders containing a wide range of SCM types and replacement ratios.

1. Introduction

The current annual usage of concrete is estimated to be 20 billion tons worldwide (Bai et al., 2018), and is expected to rise as emerging economies rapidly develop. For example, it has been estimated that the construction of the infrastructure needs of fast-growing cities in developing countries could release 226 gigatonnes of carbon dioxide by 2050, which would make it difficult to remain within the 'carbon budget' set as part of the Paris Climate Agreement of 800 gigatonnes of total CO_2 emissions after 2017 (Bai et al., 2018).

The production of concrete requires the use of a binder, which has historically most commonly been Ordinary Portland Cement (OPC). OPC production is a heavy industrial process, based upon extensive mining, pyro-processing and grinding operations (Fairbairn et al., 2010; Tosti et al., 2018), all of which have a significant impact of the environment. For example, it has been reported that the cement industry is responsible for up to 3% of annual global energy demand and up to 9% of annual global CO_2 emissions (Crossin, 2015; Miller, 2018; Fan and Miller, 2018; Monteiro et al., 2017).

Society's growing environmental awareness and the associated need to reduce CO_2 emissions from the construction industry has been the impetus for a significant number of studies focused on limiting the impact of cement and concrete industries. These studies are generally focused on the conservation of resources (e.g. by limiting mining of limestone and coal sources (Fairbairn et al., 2010; Crossin, 2015)) or minimising the environmental impacts of concrete manufacture by investigating full or partial replacement options for OPC (Dadsetan and Bai, 2017; Long et al., 2015; Aprianti et al., 2015).

A commonly investigated approach for reducing OPC consumption is by the partial replacement with supplementary cementitious materials (SCMs). Supplementary cementitious materials are commonly industrial (coal ash, slag, silica fume) and agricultural (rice husk ash, palm oil fuel ash) wastes. From these studies it has been established (Dadsetan and Bai, 2017; Long et al., 2015; Aprianti et al., 2015) that the partial replacement of OPC with SCMs is a practical and economical means for improving the suitability of concrete while also utilising industrial waste.

In addition to directly reducing the environmental impact of concrete via the replacement of OPC, the incorporation of SCMs has also been shown to improve the mechanical (Uysal and Sumer, 2011; Le and Ludwig, 2016; Zhao et al., 2015) and durability (De Belie et al., 2017; Lothenbach et al., 2011) properties of concrete.

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The use of SCMs as a binder, therefore not only directly reduces the initial CO_2 emissions of concrete manufacture, it can also provide a means for reducing the volume of concrete required in construction and extending the service life of a structure, which is typically based on durability criteria (Siemes and Edvardsen, 1999).

The most commonly adopted method to develop a mix design for concrete containing SCMs is to conduct multiple trial tests, in which the binder and mix constituents are adjusted to obtain empirical trends, for example see the well-known Taguchi's method (Ozbay et al., 2009; Türkmen et al., 2008). This approach has been widely and successfully applied to OPC, which as shown in the ternary diagram in Fig. 1 (which shows the chemical composition of a range of binders), has only a narrow variation in chemical composition. Conversely, as also seen in Fig. 1, even for broad classes of SCMs (e.g. Class-F fly ash) a wide variation in chemical composition occurs. As a result it can be difficult to generalise the results of individual experimental campaigns.

To improve the generality of mix design procedures, in the paper, a unified approach for incorporating SCMs into concrete is developed. This approach is based on the definition and application of a reactivity index to allow for consideration of the chemical composition of a given binder.

Specifically, this research aims to:

1) Through an investigation of the fundamental chemical and mineralogical compositions of each individual binder, identify the factors that govern the hydration and pozzolanic reactions in concrete with blended OPC and SCM binders.
2) Quantify the reactivity of individual and blended binders through the use of existing reactivity moduli that are commonly applied to hydraulic and pozzolanic reactions.
3) Define a new reactivity index based on the parameters known to strongly influence the strength development of a binder (water to binder ratio and binder reactivity moduli).
4) Based on a large database of existing test results, develop a model to predict the mechanical properties of concrete with blended binders based on the binder reactivity index and mix proportions.

To achieve these objectives, in the remainder of the paper the reactivity of cementitious materials is firstly investigated and the major oxides governing the reactivity of OPC, slags and inorganic ashes are described. The concept of a reactivity moduli that may be

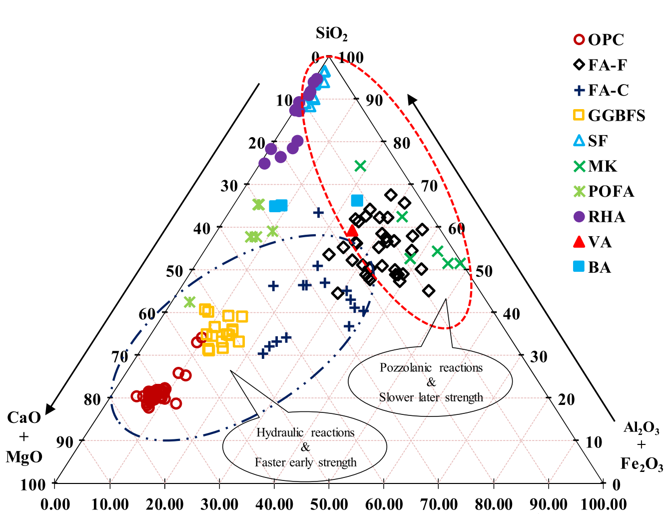

Fig. 1. Ternary diagram of cementitious materials.

used to predict the hydraulic and pozzolanic activity is then presented. An experimental database of 800 mix design containing partial OPC replacement with SCMs is then compiles and analysed based on the reactivity of the binders and the mix designs previously investigated.

To develop an improved mix design procedure, based on the analysis of the test database a new reactivity index is defined based on the combination of the reactivity moduli and the water to binder ratio. Having defined the influence of the binder in quantifying the overall compressive strength, the influence of aggregate is then investigated, along with the change in compressive strength over time and the connection between the compressive strength and the remaining major mechanical properties (tensile strength and elastic modulus). Finally it is shown how the mix design approach can be applied in practice through a series of examples in which it is applied to predict the performance of mix designs not included in the previous statistical analyses.

It is expected that the outcome of this work will:

1) Reduce the need to conduct multiple-trial tests in order to reliably develop mix designs for concrete containing SCMs.
2) Improve the transferability of the findings of existing and future studies on SCM incorporation by providing simple means for expressing mix designs in terms of the chemical composition of the binder.
3) Promote and standardize the usage of industrial by-products and agricultural wastes in cleaner concrete products.

2. Reactivity of cementitious materials

2.1. Oxides governing reactivity

In this section the major hydraulic and pozzolanic reaction mechanisms are reviewed such that a rational means for incorporating the chemical composition of the binder materials into the mix design procedure can be established.

For each major class of cementitious material (OPC, Slag and Inorganic Ash) the major phases influencing reactivity are summarized as follows and in Table 1:

- OPC: the four major phases associated with the hydraulic reactions in ordinary Portland cement clinker are Alite, Belite, Aluminate, and Ferrite. (Wild et al., 1995; Taylor, 1997).
- Slags: have a similar mineralogical composition to OPC and predominately contain oxides of CaO, SiO_2, Al_2O_3, and Fe_2O_3 that give GGBFS its self-cementing properties. Owing to its high MgO and Al_2O_3 content, the additional mineralogical composition in slag materials are merwinite ($Ca_3Mg(SiO_4)_2$), akermanite ($Ca_2Mg(Si_2O_7)$), gehlenite ($Ca_2Al(AlSi)O_7$) and rankinite ($Ca_3Si_2O_7$), and these phases are associated with both the hydraulic and pozzolanic activity (Darquennes et al., 2013; Behim et al., 2013).
- Inorganic Ashes: contain significantly lower proportions of CaO than either OPC or slags, which in general limits their self-cementing properties. An abundance of SiO_2 and Al_2O_3 in some ashes indicate the potential to support pozzolanic activity (Xie and Ozbakkaloglu, 2015; Sakai et al., 2005).

2.2. Reactivity moduli of cementitious materials

Knowing the oxides that govern the reactivity of a given binder material, the reactivity of any given cementitious material can be quantified using the concept of a reactivity moduli. These moduli,