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Synthesis and characterization of alkali aluminosilicate hydraulic cement that meets standard requirements for general use

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HIGHLIGHTS

- ASTM C1157 represents a shift away from specifications that dictate composition restrictions.
- Development of a new cement that meets standard requirement for general use.
- The new sustainable hydraulic cement was thoroughly characterized.

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ABSTRACT

A sustainable hydraulic cement was developed based primarily upon the alkali aluminosilicate chemistry. This cement employed largely coal fly ash, granulated ground blast furnace slag and natural feldspar as sources of aluminosilicates, with small concentrations of calcium oxide, sodium hydroxide and sodium silicate used as sources of alkalis and soluble silica. In addition, sodium tetraborate was incorporated into the cement formulation for set retardation. The dry raw materials were transformed into a hydraulic cement via input of mechanical energy using a ball mill. Comprehensive experimental evaluation of the resultant hydraulic cement confirmed that it meets standard requirements for general use in concrete construction.

1. Introduction

Portland cement has been the predominant inorganic binder for production of concrete and other construction applications for several decades. Important improvements have been made in the efficiency of cement manufacturing, which has had quantitative effects on the energy use and carbon emissions associated with production of cement. In spite of these improvements, cement manufacturing is still a major source of anthropogenic CO_2 emissions and energy consumption worldwide [1].

The chemistry and the prevalent manufacturing features of Portland cement have remained essentially unchanged for several decades [2]. The cement industry has made significant investments in existing manufacturing plants, and has developed thorough know-how of the Portland cement chemistry, processing and performance. In recent decades, we have observed development of a chemical admixtures industry that is largely centered around the Portland cement chemistry, and has brought about significant improvements in concrete performance[3,4]. These circumstances together with some highly desired features of Portland cement have created a strong inertia against fundamental changes in the chemistry and manufacturing process of cement. Portland cement benefits from the use of raw materials that are available abundantly across diverse geographic areas. In addition, the existing approach to manufacturing of Portland cement yields an end product that reliably and cost-effectively meets the performance requirements relevant to the use of Portland cement in concrete construction and other applications.

The demands for improvement of the sustainability and some performance attributes of Portland cement have been on the rise. The significant energy content and carbon footprint of Portland cement, and the position of Portland cement concrete as the most widely used material of construction have prompted efforts to develop alternative hydraulic cements with significantly improved

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sustainability [5,6]. Efforts towards development of alternative hydraulic cements are also driven by the growing need to enhance the service life and life-cycle economy of the concrete-based infrastructure [7].

In response to need for hydraulic cements that offer qualitative advantages over Portland cement in terms of sustainability and durability, the cement industry has developed performance-based ASTM standards that do not impose chemical constraints on hydraulic cements [8]. This initiative, however, has not yet led to breakthrough developments, and the performance-based standards have not been adopted and specified widely by agencies that own and manage infrastructure systems [9]. It should be noted that a number of alternative hydraulic cement chemistries are available commercially for niche applications. Examples include rapid-hardening hydraulic cements based on calcium aluminate, calcium sulfoaluminate and magnesium phosphate chemistries [10]. The performance and economics of these specialty hydraulic cements, however, do not allow their broad use in normal concrete construction.

Efforts have been undertaken in recent years towards development of "one-part" alkali aluminosilicate cements that, similar to Portland cement, undergo hydration reactions upon addition of water. These hydraulic cements are produced by heating of a blend of aluminosilicate precursors (albite, kaolin, etc.) and alkalis [11–13]. The resulting hydraulic cements produced, upon hydration, inorganic binders with viable strength levels. This is a new field of development, and some key aspects of alkali aluminosilicate-based hydraulic cements require further improvement. For example, some of these hydraulic cements lack adequate stability in the presence of moisture. Efforts have been made to use lower-cost sources of alkalis for production of one-part alkali aluminosilicate cement. These hydraulic cements undergo hydration reactions in a more alkaline environment than Portland cement. Red mud (or red sludge) is a potential source of low-cost alkalis. It is a highly alkaline (with high concentrations of NaOH and $NaAlO_2$) byproduct of aluminum production, which comprises clay, silt, sand, Fe_2O_3 and Al_2O_3 [14]. Red mud is generated at an annual rate of about 77 million tons; it is a hazardous waste with serious disposal problem in the mining industry. Since alkali aluminosilicate hydrates provide significant hazardous waste immobilization qualities, red mud has been considered as a source of alkalis in development of hydraulic cements. This was accomplished via thermal processing of red mud and rice husk ash; the resultant hydraulic cement, upon hydration, produced moderate levels of compressive strength [14]. Simple blending of raw materials has also been used as a means of producing a one part hydraulic cement based on alkali aluminosilicate chemistry [15]. This approach, however, retains the caustic nature of the alkaline raw materials, and can suddenly release excess heat upon addition of water to the cement. The affinity of alkaline constituents for water could also limit their shelf life in air. Experimental results indicated that curing at elevated temperatures was required to yield viable levels of early-age compressive strength. At later ages, a drop in compressive strength was observed. More recent investigations have produced one-part formulations by blending either coal fly ash and sodium silicate or rice hull ash and sodium aluminate to achieve viable compressive strengths with room-temperature curing [16,17]. The caustic nature of cement, limited shelf life, and sudden release of excess heat upon addition of water would still be the drawbacks of these cements. Simple mixing of dry raw materials without chemically integrating the alkalis into the aluminosilicate structure (which also enhances the activity of the aluminosilicate precursor) is not a viable approach to production of hydraulic cements based on alkali aluminosilicate chemistry.

Efforts to develop one-part hydraulic cements based on the alkali aluminosilicate chemistry have been largely focused on the mechanical properties produced upon hydration. The dimensional and chemical stability, weathering and moisture resistance, microstructure and other properties of the hydration products of these cements have not been investigated. More efforts are needed to understand and improve the chemical composition of these cements, the effectiveness and efficiency of transforming the blends of raw materials into a hydraulic cement, and to thoroughly characterize these cements in order to qualify them based on performance-based standards developed for hydraulic cements which use general use in concrete production. The primary performance-based standard used in this development work was ASTM C1157 (Standard Performance Specification for Hydraulic Cement) [18].

ASTM C1157 allows for unrestricted use of raw materials and processing methods to produce innovative hydraulic cements [18]. C1157 also contains optional requirements that are not available under other specifications. The ASTM C1157 requirements are largely based on the performance limits of ASTM C150 (Standard Specification for Portland Cement) [19] and ASTM C595 (Standard Specification for Blended Hydraulic Cements) [20], the traditional cement specifications that contain a combination of prescriptive and performance limits. In ASTM C1157, cements are classified into six types according to their intended use: GU for general construction, HE for high early strength, MS for moderate sulfate resistance, HS for high sulfate resistance, MH for moderate heat generation, and LH for low heat generation [18]. The focus of this investigation is on development of a new hydraulic cement that meets the general use (GU) hydraulic cement requirements.

ASTM C1157 represents a shift away from prescriptive specifications that dictate composition restrictions. Instead, the emphasis is on the ability of cement to perform. For the tricalcium aluminate (C_3A) content of Type II or V cement is prescriptively limited to control sulfate resistance. In ASTM C1157, assurance of sulfate resistance of Type MS or HS cement is determined by testing (ASTM C1012) mortar bars made with the cement [21]. Laboratory tests, and not chemical analyses, are used as predictors for various aspects of performance. Some key performance specifications considered in this project were: (i) compressive strength development with time; (ii) heat of hydration, and initial and final set times; (iii) expansion tendencies due to alkali-silica reaction; (iv) change in length correlating with autoclave soundness; and (v) expansion of mortar bar immersed in lime-saturated water. Scanning electron microscopy, thermogravimetry, calorimetry and X-ray diffraction techniques were employed to gain more insight into the hydration mechanisms and the resultant microstructure of the new hydraulic cements.

The end product of this development effort is a hydraulic cement that meets standard requirements and is also compatible with the mainstream mix design and construction practices used with normal Portland cement concrete. These features, combined with sustainability and some important performance advantages of the new hydraulic cement, facilitate its transition to concrete construction markets.

2. Materials and methods

2.1. Materials and their characteristics

The aluminosilicate precursors used in this process were coal fly ash, ground granulated blast furnace slag, and albite. The (dry) alkalis used as raw materials were sodium hydroxide, sodium silicate, and calcium oxide. Sodium tetra-borate (Borax) was also added to the cement formulation to retard its set time [22]. The weight ratios of raw materials used for production of an alkali aluminosilicate-based hydraulic cement are presented in Table 1. A predecessor for this formulation was devised to balance the Si, Al, Na and Ca molar ratios to enable formation of viable alkali aluminosilicate hydrates. The base formulation was then refined via