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Alkaline activation of synthetic aluminosilicate glass

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Abstract

The alkali activation of aluminosilicates is a procedure for obtaining new binders with many of the same properties as traditional portland cement. The type and characteristics of the products formed, however, and consequently their mechanical performance, are conditioned by a number of parameters. The present study explored the potential reactivity of synthetic alkaline silicoaluminates alkali-activated at high temperatures by analysing parameters such as the chemical composition of the starting material and the vitreous phase content. When alkali-activated, the glass produced in the laboratory developed high mechanical strength, while the performance of the resulting binders was conditioned by the SiO_2/Al_2O_3 ratio in the starting materials. The optimal values were observed to lie between 3 and 4.

Keywords: D. Glass; Alkaline activation; Geopolymer; Mechanical strength; N–A–S–H gel

1. Introduction

The alkaline activation of aluminosilicates is an effective alternative to the use of portland cement [1,2]. To date, the aluminosilicate materials used for this purpose have consisted primarily of industrial by-products such as fly ash. While this practice is beneficial both environmentally and economically, the uneven supply of these prime materials and the variations in their characteristics may pose problems. A pressing need has thus arisen for an abundant supply of aluminosilicate-based materials available anywhere on the planet and convertible (via less intense heating than required to manufacture portland clinker) into reactive vitreous materials apt for alkaline activation and the concomitant generation of new aluminosilicate binders.

In the context of a quest for sustainability, lower costs and prime material availability, common clay and feldspars (materials with very little calcium that would consequently not need to be decarbonated) may apt for alkaline activation after thermal treatment and conversion to glass [3–6]. Their use, in addition to enlarging on the range of materials that can be alkali-activated, would reduce the exploitation of the limestone normally used in cement manufacture in favour of other globally abundant mineral resources, thereby lowering the environmental impact of cement manufacture.

In the alkaline activation of aluminosilicates, many parameters affect the reaction products formed and consequently the quality of the binder. These parameters include the composition of the starting materials (e.g., the SiO_2/Al_2O_3 ratio or the reactive silica content), the type of alkali activator and the reaction conditions [7–16]. By controlling these parameters, optimal conditions can be designed to produce higher amounts of N–A–S–H gel ($xNa_2O \cdot yAl_2O_3 \cdot zSiO_2 \cdot nH_2O$), the substance that affords these systems their mechanical strength and durability.

Many studies can be found in the literature that support the existence of a direct relationship between the mechanical performance of activated systems and the characteristics of prime materials, and more specifically their SiO_2/Al_2O_3 ratio [17–22]. The variability of these materials, however, and therefore the wide differences in their chemical composition depending on their origin, along with the lack of uniformity in the way the SiO_2/Al_2O_3 ratio is measured (by weight, molar or atomic ratio, or in % of oxides), constitute a sizeable obstacle to understanding the relationship between the two.

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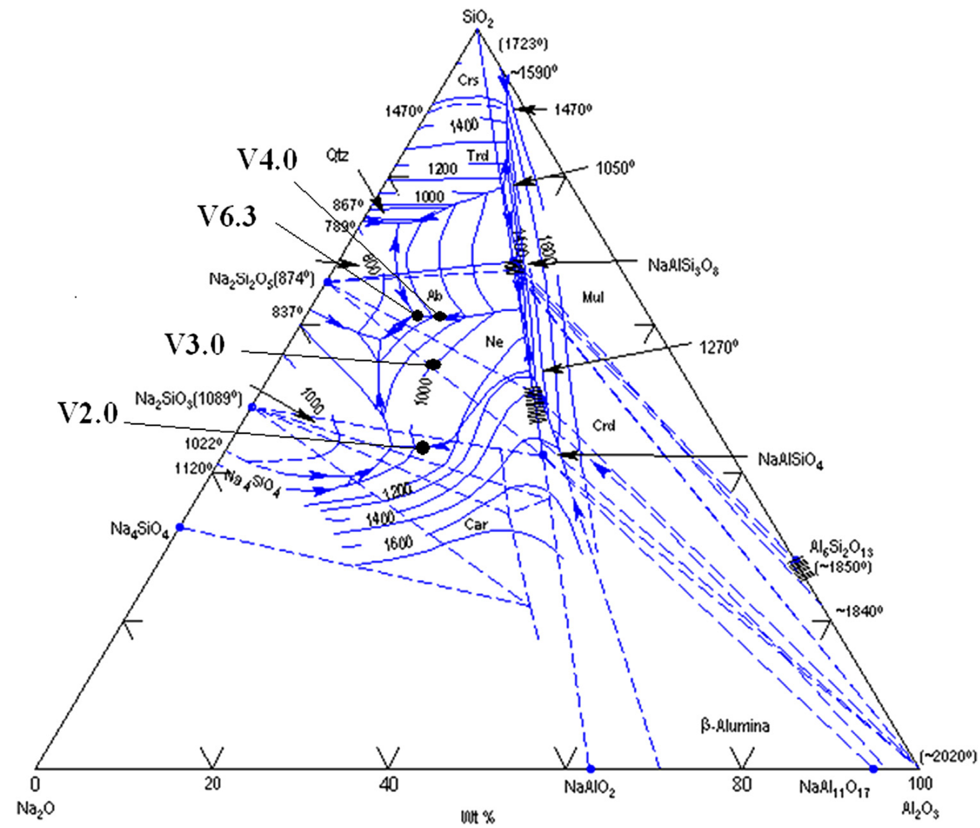

Fig. 1. Phase equilibrium diagram for the $Na_2O–Al_2O_3$ system[25] showing the primary crystalline phase fields, isotherms and binary and ternary invariant point temperatures.

The present study focused on the effect of raw material starting composition (SiO_2/Al_2O_3 ratio) on the mechanical strength of the product. An in-depth understanding of the most reactive compositions in the $Na_2O–SiO_2–Al_2O_3$ ternary diagram could go a long way to facilitating this task. Glass with the desired composition, obtained by heating to high temperatures, would provide a readily controlled raw material that would not be affected by the compositional variability that constrains the use of traditional supplementary cementitious materials (SCMs). The present article addresses the problem holistically, from preparation of the glass to characterisation of the activated product.

The aim of the research was to prepare alkaline aluminosilicates glasses (via high temperature melting and quenching) potentially apt for alkali activation to produce good binders, and to study the activation products formed.

The choice of the most favourable compositions for preparing the alkaline aluminosilicate glass (with laboratory reagents) was based on the information found in the literature on the SCMs, such as fly ash and metakaolin, best suited for the manufacture of alkaline cements (normally with SiO_2/Al_2O_3 ratios of 2–4) [17,23–26]. Further information was drawn from the phase diagram for the $Na_2O–SiO_2–Al_2O_3$ system (see Fig. 1) (in which the compositions commonly regarded in the literature on alkali-activated aluminosilicates as the most reactive were moved to a position close to the invariant points (eutectics, peritectics) with the lowest melting points) [27,28].

2. Experimental

2.1. Glass synthesis

The point of departure for glass preparation was the study of four strategic compositions in the $Na_2O–SiO_2–Al_2O_3$ ternary system, all within or very close to the nepheline ($NaAlSiO_4$) primary crystallisation field (Fig. 1). The composition with a SiO_2/Al_2O_3 ratio of 2 (sample V2.0) was nearly a eutectic ($Na_2O \cdot SiO_2$–nepheline–carnegieite) with a 915 °C melting point (located on the 1000 °C isotherm). The other three compositions were located in the $Na_2Si_2O_5–NaAlSi_3O_8–NaAlSiO_4$ solid state compatibility triangle, in which 732 °C marked the invariant point. Compositions with SiO_2/Al_2O_3 ratios of 3 (V3.0) were located on the 1000 °C isotherm, the ones with a ratio of 4 (V4.0) on the 800 °C isotherm and composition 6.3 (V6.3) was the ternary sub-system eutectic. The highest SiO_2/Al_2O_3 ratio (6.3) was selected to determine the minimum amount of aluminium required in the source aluminosilicate to yield potentially reactive materials. The compositional characteristics of the glass are shown in Table 1.

The glass was synthesised by melting laboratory reagents at high temperatures. The reagents used to generate the compositions selected (see Fig. 1) were mixed in a turbula, compacted into pellets, placed in ceramic (inert for these materials) crucibles and heated for 2 h in a Termolab electric laboratory