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## The role of particle size on the performance of pumice as a supplementary cementitious material



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#### ABSTRACT

A critical area overlooked in previous research on pumice is understanding how its physical characteristics influence its behavior as a supplementary cementitious material (SCM). This study investigated three pumices with different particle size distributions to observe whether these porous materials exhibit enhanced nucleation and growth of hydration products, in the same way as non-porous materials, and whether the rate of pozzolanic reaction can be changed through particle size. The effect of particle size on compressive strength, rheology and resistance to alkali silica reaction (ASR) was also evaluated. Results showed that reducing particle size increased the rates of cement hydration, pozzolanic reaction, and compressive strength gain, while also increasing mixture viscosity. Interestingly, particle size did not impact the yield stress of the mixture or the resistance to ASR. These new findings give insight about how the particle size of pumice can be used to overcome drawbacks reported in previous literature.

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#### 1. Introduction

The push towards more economical, sustainable and durable concrete mixtures has made supplementary cementitious materials (SCMs) very important in the concrete industry. However, at a time of increasing SCM demand, global changes in the power generation industry have led to regional supply shortages of fly ash, one of the most widely used SCMs. Consequently, there is increased interest in the cement and concrete community to evaluate other materials that can serve as suitable SCMs to overcome this scarcity. Pumice, a natural pozzolan, is one such promising material that has seen renewed interest in being used as an SCM [1-4]. Pumice is formed from highly silicic volcanic lava that tends to hold more dissolved gases than other types of lava due to its viscosity. As the lava rapidly cools and hardens into a glassy structure, the dissolved gases form pores or vesicles that result in low density volcanic rock [5]. The high silica content and amorphous structure are what give pumice its pozzolanic properties and make it a candidate for use as an SCM in concrete. In fact, pumice was used in concrete throughout history dating back to the ancient Greek and Roman civilizations [3]. Even as recent as the 20th century, finely ground pumice was used as an SCM in mass concrete structures, before the price and availability of fly ash made it more economical than pumice. Some notable examples of US structures with pumice are the Los Angeles aqueduct in 1912, the Friant Dam in 1942, the Altus Dam in 1945 and the Glen Canyon Dam in 1964 [3,6].

With the rising demand for SCMs, pumice is experiencing a renaissance in concrete research. Recent studies have shown finely ground pumice to meet most of the requirements listed in ASTM C 618 for natural pozzolans [4,7] and to lower calcium hydroxide content in cementitious mixtures, which is an indication of pozzolanic reaction [4,8,9]. Researchers have also evaluated the performance of pumice mixtures in terms of compressive strength, durability, and mixture workability [1–4,8,9]. However, a critical area of research that has been overlooked in previous literature is understanding how the characteristics of the pumice itself, like particle size and composition, influence its behavior in cementitious mixtures. For example, previous literature has indicated that replacing cement with finely ground pumice decreased the compressive strength of mortar and concrete specimens [1,4,7,9], but there has not been any investigation on how these effects on strength relate back to characteristics of the pumice itself, and whether those characteristics can be modified to improve the compressive strength of pumice blended mixtures. Similarly, for durability, although studies have found pumice to be effective at

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controlling alkali silica reaction (ASR), results vary on the minimum percentage needed to mitigate expansions [2,7]. Since pumice is a natural pozzolan whose properties can vary depending upon its sourcing, these differences in reported results are not unusual. However, it is important to understand how these properties of pumice change its effectiveness in mitigating ASR so that minimum replacement dosages can be predicted for new sources. Evaluating the effects of pumice characteristics on mixture workability is also important, as fresh state properties ultimately affect hardened concrete properties. While there are some studies that have qualitatively measured workability of pumice concrete mixtures using the slump test [4,8,9], a more comprehensive analysis is needed to evaluate how the viscosity and yield stress of pumice blended mixtures change with characteristics of the pumice itself.

The main objective of the study presented here was to address the shortcomings in understanding how physical properties of pumices influence their behavior in cementitious systems. We used three pumices with similar compositions but differing particle size distributions to evaluate the role of particle size in the performance of pumice as an SCM. In this way we could hold all material variables constant other than pumice particle size, so as to eliminate the overwhelming effects of parameters such as composition on pozzolanicity, particularly amorphous content and soluble silica and alumina contents [10]. The influence of pumice particle size was examined for its impact on the following: cement hydration kinetics, pozzolanic reactivity, compressive strength, resistance to ASR, and rheology. Based on previous research on other SCMs and filler materials [11,12], we hypothesized that in the early stages of hydration, when the process is dominated by the availability of nucleation and growth sites, pumice with a fine particle size distribution can increase the rate of cement hydration by providing an increased surface area for the nucleation and growth of hydration products. While this is simply referred to as "filler effect" in previous literature [11,12], in this paper it will be referred to as "size filler effect" to differentiate it from "space filler effect," which is the growth of additional hydration products due to increased space per grain of cement when SCM or filler materials are used [11,12]. Much of the work examining size filler effects has been done with nonporous materials, and it is of interest to determine if the same principles apply for porous SCMs like pumice. Isothermal calorimetry was used to observe how pumice particle size can change cement hydration kinetics through size filler effects. Thermal gravimetric analysis (TGA) was used to measure calcium hydroxide consumption in hydrated pastes to see whether pumice particle size affected the rate of pozzolanic reaction. Compressive strengths of pumice-containing mortars were measured from 1 day to 1 year, to observe how the changes to early cement hydration kinetics and pozzolanic reaction rate correlate to the rate of strength gain. Based on previous research on fly ash and limestone [13,14], we hypothesized that the low early strengths of the pumice mixtures reported in literature [1,4,7,9] were most likely due to the dilutionary effect of replacing hydraulic cement with a slower reacting pozzolanic material like pumice. However, we expected that this dilutionary effect could be overcome, at least partially, to achieve higher early age strengths by using a finer pumice pozzolan that can provide increased surface area to enhance nucleation and growth of hydration products during the early stages of hydration. Further, other than a paper by Mielenz et al. [15], most studies report compressive strength data for pumice-containing mixtures only until 28 days. Therefore, we measured the strength of pumice-containing mortars for one year to provide insight into the role of particle size on the rate of strength gain at later ages and to evaluate the amount of time it takes to offset the dilutionary effect of replacing cement with pumice. ASR resistance of pumice-containing mortar bars was evaluated to see if particle size changed the effectiveness of the pumice pozzolans at mitigating expansions due to ASR. Finally, the effect of pumice particle size on mixture workability was measured in terms of yield stress and plastic viscosity of pastes. Pumicecontaining mixtures were compared against mixtures with an inert quartz filler and control samples without SCMs or filler materials.

#### 2. Materials and methods

The three pumice powders used in this paper were sourced from USA and are referred to as Pumice-N, Pumice-D and Pumice-S. Pumice-N and Pumice-D are commercially available SCMs, while Pumice-S was originally a fine aggregate that was ground in the laboratory using a Bico Inc. UA V-Belt Drive Pulverizer and sieved through a No. 200 sieve (75 µm opening) for use as an SCM. All three pumices were tested to determine if they could be classified as Class N pozzolans according to the guidelines stated in ASTM C 618 [16]. The results, presented in Table 1, show that the physical and chemical characteristics of the pumices fulfilled all of the requirements for Class N pozzolans. X-ray diffraction (XRD) tests, conducted using a Siemens D-500 X-ray Diffractometer with Cu-Ka radiation and a dwell time of 4 s, showed the pumices to be mostly amorphous (Fig. 1). X-ray florescence (XRF) analyses of the pumices were conducted using a Bruker S4 Explorer according to the procedures of ASTM D 4326 [17]. The results, presented in Table 2, confirmed the oxide compositions of the pumices to be almost identical to each other. It was assumed, therefore, that the amorphous content and soluble silica and alumina contents of the pumices were nearly identical, so that the only difference between them was the gradation. An inert quartz filler, purchased from Old Hickory Clay Company, was also used in this study. The cement used in all mixtures was an ASTM C 150 [18] Type I portland cement sourced from USA. The XRF oxide composition of the cement, found using the same procedure as the pumices, is also presented in Table 2.

The particle size distributions of the pumices and quartz filler were analyzed using a Horiba Partica LA 950-V2 Laser Scattering Particle Size Distribution Analyzer and are presented in Fig. 2. Table 3 lists the d<sub>10</sub>, d<sub>50</sub>, and d<sub>90</sub> values of the particle size distributions, which respectively refer to the particle size below which 10%, 50% and 90% of the sample lies. From the particle size distribution results, Pumice-N, with a median particle size  $(d_{50})$  of approximately 3 µm, was observed to have the finest particle size distribution among all the SCMs/filler materials tested. Pumice-D was slightly coarser than the quartz, with a d<sub>50</sub> value of approximately 13  $\mu\text{m}.$  Finally, Pumice-S was observed to have an even coarser distribution than Pumice-D, with its d<sub>50</sub> value being approximately 20 µm. Since Pumice-S was originally a fine aggregate that was ground in the laboratory, it was expected that it would have a coarser distribution than both Pumice-N and Pumice-D, which are commercially marketed as SCMs. The surface areas of the pumices and guartz filler were measured using an ASAP 2020 Micromeritics Surface Analyzer with nitrogen gas and are presented in Table 3. Due to the porous nature of the pumice pozzolan [5] that gives rise to internal surface area, Pumice-D had a much higher surface area than the quartz, despite having a similar particle size distribution.

The control sample was a mixture that had 100% portland cement (OPC) as its binding material. The other mixtures had 20% of the cement content by weight replaced with either pumice or the quartz filler. The isothermal calorimetry and thermal gravimetric tests used pastes with a water to cementitious (and/or filler) material ratio (w/cm) of 0.45 and 50 g of cementitious (and/or filler) material. Prior to each test, the pastes were mixed by hand for 2 min. For the calorimetry tests, approximately 10 g of paste was

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