# ABSTRACT

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Title: Influence of Chemical and Physical Properties of Poorly-Ordered Silica on Reactivity and Rheology of Cementitious Materials
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Silica fume is a widely used pozzolan in the concrete industry that has been shown to have numerous benefits for concrete including improved mechanical properties, refined pore structure, and densification of the interfacial transition zone between paste and aggregates. Traditionally, silica fume is used as a 5% to 10% replacement of cement; however, newer classes of higher strength concretes use silica fume contents of 30% or greater. At these high silica fume contents, many detrimental effects, such as poor workability and inconsistent strength development, become much more prominent.

In order to understand the fundamental reasons why high silica fume contents can have these detrimental effects on concrete mixtures, eight commercially available silica fumes were characterized for their physical and chemical properties. These included traditional properties such as density, particle size, and surface area. A non-traditional property, absorption capacity, was also determined. These properties or raw material characteristics were then related to the hydration and rheological behavior of pastes and concrete mixtures. Other tests were performed including isothermal calorimetry, which showed that each silica fume reacted differently than other silica fumes when exposed to the same reactive environment. Traditional hydration models for ordinary portland

cement were expanded to include the effects that silica fumes have on water consumption, volumes of hydration products, and final degree of hydration.

As a result of this research, it was determined necessary to account for the volume and surface area of unhydrated cement and unreacted silica fume particles in water-starved mixture proportions. An adjustment factor was developed to more accurately apply the results from hydration modeling. By combining the results from hydration modeling with the surface area adjustments, an analytical model was developed to determine the thickness of paste (hydration products and capillary water) that surrounds all of the inert and unreacted particles in the system. This model, denoted as the "Paste Thickness Model," was shown to be a strong predictor of compressive strength results. The results of this research suggest that increasing the paste thickness decreases the expected compressive strength of concretes at ages or states of hydration.

The rheological behavior of cement pastes containing silica fume was studied using a rotational rheometer. The Herschel-Bulkley model was fit to the rheological data to characterize the rheological behavior. A multilinear model was developed to relate the specific surface area of the silica fume, water content, and silica fume content to the Herschel-Bulkley rate index. The Herschel-Bulkley rate index is practically related to the ease at which the paste mixes. This multilinear model was shown to have strong predictive capability when used on randomly generated paste compositions.

Additionally, an analytical model was developed that defines a single parameter, idealized as the thickness of water surrounding each particle in the cementitious system. This model, denoted as the "Water Thickness Model," incorporated the absorption capacity of silica fumes discovered during the characterization phase of this study and was shown to correlate strongly with the Herschel-Bulkley rate index. The Water Thickness Model demonstrates how small changes in water content can have a drastic effect on the rheology of low w/c or high silica fume content pastes due to the combined effects of surface area and absorption. The effect of additional water on higher w/c mixtures is significantly less.