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Mechanical, rheological and morphological analysis of cement-based composites with a new LAS-based air entraining agent

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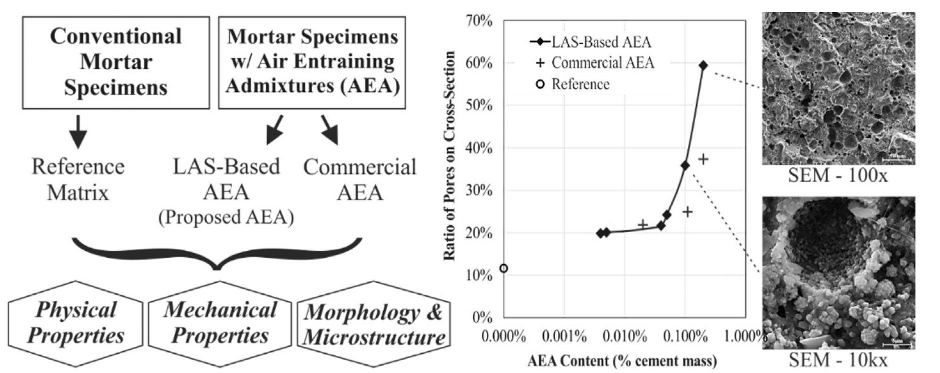
HIGHLIGHTS

- LAS-based washing-up liquids are effective air-entraining agents (AEA).
- Performance and morphology of LAS-based AEA is similar to commercial AEA.
- Excess of AEA compromises significantly the mechanical properties of mortars.
- The pore system of a matrix with AEA is directly related to the AEA content.
- The mechanical/physical properties are directly related to the pore system.

GRAPHICAL ABSTRACT



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ABSTRACT

The air-entraining admixtures (AEA) consist of surfactant compounds, as do washing-up liquids. Washing-up liquids are products widely available, non-toxic and biodegradable. This work proposes the use of an anionic surfactant from washing-up liquids, Linear Alkyl Benzene Sodium Sulfonate (LAS), as AEA. Mechanical, physical and rheological properties of mortars comprising LAS were compared to ones with commercial admixture and one without any. The morphological aspects of the air-entrained matrix were also evaluated, and associated to its properties. Results indicate that the LAS-based AEA is an effective air-entraining agent. Thus, the present work seeks to disclose this new biodegradable high-performance potential AEA, as well as to improve the comprehension of the effects of AEAs in cement-based composites.

1. Introduction

The air entraining admixtures (AEA) introduce small air bubbles (or voids) dispersed throughout the matrix. These homogeneously distributed microbubbles improve the cohesion and workability of cement-based composites, prevent water penetration, and reduce the tendency of segregation and bleeding in fresh concrete [26]. AEAs are usually employed in concrete subjected to freezing and thawing cycles [16], for lightweight concrete and pumped concrete. Surfactants (or surface-active compounds) perform the mechanism of air entrainment [20].

Surfactants reduce the surface tension of substances, due to the balance of forces between its molecules at the interface. In general,

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the molecules of surfactants contain a hydrophobic (or nonpolar) chain and one or more hydrophilic (or polar) groups. One example is the Linear Alkyl Benzene Sodium Sulfonate (LAS) (Fig. 1) – the active component of commonly used washing up liquids. The detergents interact both with fat (nonpolar) and with water (polar), thus promoting the cleaning of the surfaces.

LAS is a family of synthetic detergents, part of most of cleaning products. Some of the benefits of using washing up liquids for this purpose, compared to AEA commercially available, are: wide availability, low cost, tenderness to the skin, and lower environmental impacts since they are biodegradable by government regulation [2]. Furthermore, while commercial admixtures are sold in minimum volumes of 1 L, usually in buckets of 15 L, household washing up liquids are easily adaptable for small concreting.

Therefore, this paper evaluates the potential of using washing up liquids as AEA. The active principle of the proposed admixture is easily accessible to the population, relatively inexpensive, non-irritating to the skin, easy to apply, as well as biodegradable. This work also presents the morphological analysis of the pore system of the new LAS-based AEA. Several mortar matrices were produced, with increasing content of AEA. The morphological characteristics of their pores were presented, evaluated, and associated with physical and mechanical characteristics. Hence, this work aims to improve the comprehension of the physical effects of the addition of AEAs to cement-based materials, as well as the consequences to their properties, and ultimately contribute to the technological development of cement-based composites.

2. Washing up liquids

Washing up liquids are composed by surfactants (or surface-active substances). After a certain critical concentration in water, the surfactant molecules in solution begin to aggregate as micelles, in dynamical systems. Detergents work by inserting the dirt particles of nonpolar nature (such as oil and grease) into the interior of the micelles and keeping them in suspension, facilitating their removal [23]. According to the electrostatic nature of the polar group, surfactants are classified as anionic, cationic, amphoteric and non-ionic.

Detergents may be composed of one or more types of surfactant, aiming to improve their cleaning performance. The LAS is the most widely used surfactant. Its popularity is due both to low production costs, as for its performance [5]. Although the commercial LAS consists of over 20 individual components, including sodium dodecyl-benzene sulfonate (known AEA), the ratio between the various homologues and isomers is relatively constant between different applications. Because of the close consistency of the mixtures, and effects, LAS is usually discussed as a substance [17]. In addition to the surfactants, household detergent may also contain thickeners, sequestering agent, preservatives, hydrotropes, and aesthetic enhancers such as perfumes and dyes [23].

Regarding chemical attack, the predominant surfactant in washing up liquids, LAS, has no sulphates (SO_4^{-2}) that could react with cement hydration products [17]. In its chemical composition, as in many current superplasticizers, the sulphonates (SO_3^{-1}) predominate. The effect of this molecules has been studied by Prince et al. [31], among others, who report a slight delay on the growth rates of the hydrates, but no deleterious consequences on the long term.

Fig. 1. Molecule of LAS.

It is known that anionic surfactants, in the presence of calcium ions, form a insoluble precipitate that may affect their detergency efficiency [35]. The characteristics of this reaction depend on the concentration of salts, the temperature and the presence of micelles in the solution [28]. To avoid this effect, sequestering substances, which interact with the ions responsible for water hardness, mainly Calcium (Ca^{+2}), Magnesium (Mg^{+2}) and Iron (Fe^{+3}) are added to detergents. This mechanism avoids the formation of insoluble salts which may interfere with the detergent function [23].

The concentration of surfactants in the washing up liquid composition is comprised between 5% and 25%; while that of sequestrants, up to 5% [23]. In cement-based composites, the concentration of AEA usually varies between 0.1 g and 10 g per 1000 g of cement. Thus, for every 1000 g of cement, 0.005–2.5 g of surfactants will be present; and 0–0.5 g of sequestrants. Thus, the effect of the formation of insoluble precipitates is negligible in relation to the consequences promoted by the air-entrainment action. On the other hand, sequestrants have the potential to capture free calcium and magnesium ions, which would otherwise contribute to deterioration by hydrating and expanding [26].

In turn, the biodegradability of detergents is bound to its molecular structure. In general, the biodegradable ones contain a linear carbon chain (such as LAS) while non-biodegradable have branches on it [36]. The microorganisms responsible for the bio-degradation break down the linear chains of the surfactant molecules, but do not recognize branched chains.

In general, the chemical admixtures for concrete are toxic substances for humans and for the environment, as their own safety data sheets highlight. These substances can contaminate the soil and waterways during concreting process or cleaning of the mixing equipment. Active ingredients of some AEA, such as Sodium Lauryl Ether Sulphate, are known to be irritating to the skin. In turn, the LAS-based detergent is a product that, in addition to biodegradable, is nontoxic.

3. Mechanism of air entrainment

According to Powers [30], there are two main sources of entrained air in cement-based composites. The first is the infolding of the air into the matrix by vortex action, as in stirring a liquid. In the second phenomenon, also called three-dimensional screen, air bubbles are formed by aggregates cascading upon themselves during mixing. Due to the free surface energy, air voids in the fresh concrete are inherently unstable [20]. The function of the AEA is stabilizing air bubbles or voids, so they do not coalesce, do not collapse, do not emerge to the surface, and the air does not diffuse into larger surrounding bubbles [27]. Thus, the addition of air entrainment agents aims to promote a void system of suitable size and homogeneously distributed pores in the hardened concrete.

The mechanism of action of AEA, shown in Fig. 2, comprises two processes: the interaction in the air-water phase and in the solid-water phase. In the air-water phase, the polar groups are oriented towards the aqueous phase, reducing the surface tension, promoting the formation of bubbles and reducing the tendency of the already formed bubbles to dissolve [24]. Simultaneously, in the solid-water interface, the polar groups adsorb to the cement, maintaining the nonpolar groups oriented to the aqueous phase, converting the cement surface hydrophobic, and thus the air can displace the water and remain linked to the solid particles in the form of bubbles as shown in Fig. 2 [24].