



Review

A review of dispersion of nanoparticles in cementitious matrices: Nanoparticle geometry perspective



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HIGHLIGHTS

- Uniform dispersion of nanoparticles in cementitious matrices is a big challenge and thus requires more attention.
- Geometrical parameters play a key role to achieve proper dispersion.
- Dispersion of 1D nanoparticles is complex due to their bundling and entanglement.
- Dispersion of 2D nanosheets is more difficult due to their higher surface energy.

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ABSTRACT

In recent decades the dispersion of nanoparticles has been extensively investigated for the fabrication of cementitious nanocomposites, owing to their superior performance. In this review, studies related to dispersion of nanoparticles in cementitious matrices are reviewed in the context of the nanoparticle geometry, i.e. zero-, one-, and two-dimensional nanoparticles, leading to better understanding of the advantages and disadvantages of different methods to produce cementitious nanocomposites. The topics covered include different methods and mechanisms of nanoparticle dispersion in cementitious matrices and comparison of their effectiveness in the dispersion of nanoparticles with different geometrical configurations. It can be interpreted from our findings that a combination of methods to disperse and then stabilize would provide a desired state of dispersion. The technique used for each type vary from one nanoparticle to another. This paper provides insight into the application of nanoparticles in cementitious matrices and the efficient procedures for their dispersion.

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Contents

1. Introduction	347
2. Dispersion of nanoparticles in cementitious and aqueous environments	347
3. Dispersion methods	348
4. Dispersion of carbon-based nanoparticles with zero, one, and two dimensions	348
4.1. CNTs	349
4.1.1. Mechanical methods	349
4.1.2. Chemical methods	349
4.2. Graphene, graphene oxide and reduced graphene oxide	350
4.2.1. Mechanical stirring	350
4.2.2. Ultrasonication	351
4.2.3. Functionalization	351
4.2.4. Dispersing agents	352
4.3. Review	352
5. Dispersion of 0D, 1D, and 2D silica-based nanoparticles	352

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5.1.	SiO ₂	352
5.1.1.	Superplasticizers	353
5.1.2.	Ultrasonication	353
5.2.	Halloysites	353
5.3.	Nanoclay	353
5.4.	Review	354
6.	Conclusions	354
	Acknowledgments	354
	References	354

1. Introduction

The use of different nanoparticle types, marvels of modern construction technology, has become more industrial and common since their first appearance in recent decades. As is known, nanoparticles have wide applications in modifying various aspects of concrete and cementitious mixtures, such as mechanical properties, porosity, and frost resistance [1–4]. With these advantages known, the problem regarding nanomodification is dispersion efficacy. Commercial nanoparticles, despite being produced and supplied in sizes below 100 nm, usually appear as large agglomerates with dimensions between 1 and 100 μm [5–7]. Agglomeration reduces the specific surface area of nanoparticles, thereby hindering full exploitation of these precious materials with the aim of improving desired properties of concrete and other cement mixtures. Moreover, the formation and presence of these agglomerates hampers proper dispersion of nanoparticles within cement mixtures, leading to the creation of weak zones in the final product [5]. Therefore, ideal dispersion becomes a goal of high importance and it is vital to investigate different aspects and factors that might have a positive effect, even a minor one. The ideal dispersion can be described as the state in which nanoparticles are completely separated from each other and no clusters or agglomerates exist. Yet it is obvious that this objective is too high to achieve; therefore, as a more pragmatic solution, we may focus on getting as close as possible.

In this article, we have chosen to investigate the influence of geometrical differences between nanoparticles on their final dispersion. The dimensional ratio plays a major part in categorizing nanoparticles as well as their performance in cementitious matrices. For this aim, we classify nanoparticles as zero-dimensional (0D), one-dimensional (1D), and two-dimensional (2D) nanoparticles. A 0D nanoparticle is defined as a particle with all its dimensions subjected to nanoscale. For example, nano-SiO₂, TiO₂, ZnO, and CaCO₃ nanoparticles are categorized as 0D. A 1D nanoparticle can be described as having two dimensions in nanoscale, with the other dimension reaching above nanoscale. Nanotubes such as carbon nanotubes (CNTs) and halloysites belong to this category. A 2D nanoparticle is originally a sheet with its thickness in the nanoscale and its sides spreading beyond the ‘nano’ criterion. This definition covers a considerably large area within nanomaterials, involving nanoclay, graphene, and its derivatives such as graphene oxide (GO).

It is clear that there is a lack of knowledge in the efficient dispersion method of different types of nanoparticles with zero, one and two dimensions in cementitious matrices, while the correct method of mixing and dispersing added nanoparticles into cementitious matrix is crucial for exploiting the potential of nanoparticles for the development of new cementitious nanocomposites. To shed light on this problem, in this review we have gathered and surveyed research regarding the dispersion of 0D, 1D and 2D nanoparticles in cementitious matrices. The main discussion begins with a section elaborating on the dispersion of nanoparticles in cementitious and aqueous environments and the differences between

them, followed by a brief introduction to the dispersion methods. Next, the dispersion of carbon-based nanomaterials within cementitious mixtures is discussed. Subsequently, we investigate three nanoparticles belonging to the family of silica-based nanomaterials, each pertaining to a different dimensional category. The reason for reviewing these silica-based materials was to confirm and complement our investigations of carbonic materials and to apply our conclusions to more extensive types of nanoparticle. Furthermore, as the reviewed materials each have different reactions to water, carbon-based particles being hydrophobic and silica-based particles being hydrophilic, the study also compares the main objective, the geometrical approach, alongside the reaction to water. Finally, the conclusion section of the paper reviews the important findings and suggestions.

2. Dispersion of nanoparticles in cementitious and aqueous environments

Many of the experiments on the dispersion of nanoparticles have been performed in an aqueous environment rather than in a cementitious context, contexts which are significantly different, leaving behind many unanswered questions. In most situations, the quality of dispersion is assumed based on the performance of the composite; improved properties have been mostly translated into a better dispersion. However, there are cases in which the quality of dispersion has been thoroughly assessed in the final matrix using various methods including scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Thanks to Parveen et al. [8], who have provided a review on nanotube and nanofibre dispersion in cementitious composites, and to others who have gathered information with similar focus [9,10], we have some data available, but a large part of it is sparse and unorganized.

The cementitious environment, either paste or mortar, is filled with multiple ions including sodium, potassium, calcium, sulphur and hydroxyl ions that are the main components, with aluminium and silicon in lower concentrations [11]. The aforementioned ions do not maintain a constant concentration, tending to fluctuate during the cement hydration. During the first hours after fabrication of the dispersion, for instance, the concentration of calcium, sulphate and hydroxyl remains almost stable. In contrast, the concentration of calcium and sulphate ions decreases significantly in the time range of 6–24 h after the beginning of the hydration, while the concentration of the hydroxyl, Si, and Al undergoes an increment [12]. Furthermore, depending on the content of incorporated superplasticizer, this addition can also increase the sodium and hydroxyl concentration [11]. In the context of the setting time of cement, a focus on the concentration of each ion, the changing trends in that concentration, and also whether the ions are monovalent or multivalent, all can be effective in finding a proper treatment to achieve the desired dispersion. Apart from Van der Waals attractive forces, the bridging influence of Ca²⁺ ions, resulting from the ionic nature of the cement, is responsible for the agglomeration of nanoparticles when incorporated within ionic systems. For instance, from a close look at nanosilica dispersion behaviour in

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