



## Review

## Clay in cement-based materials: Critical overview of state-of-the-art



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## HIGHLIGHTS

- Critical overview of effects of clay in cement-based materials is provided.
- Dimensional stability of aggregates bearing clay minerals is examined.
- Treatment of clays to mitigate their detrimental effects in concrete is outlined.
- Research on nano-clays in cement-based materials is highlighted.
- How to resolve ambiguity related to clays in cement-based materials is discussed.

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## ABSTRACT

Limitations in guidelines and standards on the amount of clay in sand and coarse aggregate micro-fines and the influence of such micro-fines on fresh and hardened concrete properties is often ambiguous for practitioners and quality control professionals. This is compounded by conflicting related data in the open literature and the inadequacy of some standard test procedures for capturing the real problems associated with the presence of clays in cement-based materials. This paper examines the various types of clay, limitations on clays in aggregates in various standards, and the test methods used to assess the presence of clays in aggregates. A critical overview of literature on the possible effects of clay in cement-based materials is provided, including effects on water demand, workability, mechanical strength, dimensional stability and chemical admixtures dosage. The problem of dimensional stability of aggregates bearing clay minerals is examined. Possible chemical treatment of clays to mitigate their detrimental effects in concrete is also outlined. Finally, emerging research on using nano-clays in cement-based materials is highlighted and recommendations to resolve ambiguity related to the presence of clays in cement-based materials are proposed.

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## 1. Introduction

The Spanish proverb puts it best: “clay and lime conceal much evil”. This author sensed such perception of clay through personal involvement in high-profile international projects (e.g. two of the tallest buildings in the world, world’s deepest and second largest water pumping and treatment plant, one of the world’s largest airports, a world landmark bridge, etc.). Ambiguity arises when standards, technical provisions and subsequently project specifications may be unnecessarily restrictive on the amount of micro-fines allowed in aggregates used in concrete, while locally available sands and/or coarse aggregates cannot meet such limitations at competitive cost. Often futile debate emerges on what exactly is the nature of aggregate micro-fines at hand, is it responsible for any performance issues of the concrete produced, how such an influence can be accurately quantified for various microfine contents, and can the limits on micro-fines be relaxed without short- or long-term performance problems of the concrete. The discussion can be fuelled with conflicting pieces of information sourced from the open literature or through personal communications.

Therefore, this article is an attempt to demystify clay in concrete, critically overview the state-of-the-art on this issue, and provide a self-contained document that can scrutinise existing related data and the various pieces of evidence, examine the available test methods for evaluating the existence and effects of clays in cement-based materials, and essentially save the practitioner the effort of navigating through this often conflicting literature.

## 2. Different clay minerals and their behaviour

Detailed information on clay minerals can be found for instance in Fowden et al. [24]. Clay minerals are very small colloidal-sized and layered crystalline substances that evolved from chemical weathering of certain rock forming minerals. They are hydrous silicates of aluminium and other metallic ions encompassing essentially two fundamental crystal layers, silica (tetrahedral) and alumina (octahedral). It is basically the way in which these layers are stacked together with different bonding and metallic ions in the crystal lattice that differentiates clay minerals such as kaolinite, illite and montmorillonite from one another. Generally, the geological conditions and prevailing environmental effects determine which type of clay is formed.

Clay minerals are commonly found in the fine fraction (sub-75  $\mu\text{m}$  material) of natural aggregates. Such clay particles are typically less than 2  $\mu\text{m}$  in size, but can be 10  $\mu\text{m}$  or longer. The small sized clay materials and their unit cells usually have residual negative charges on their surfaces. When placed in water, the space between clay particles gets occupied by dipolar water molecules. The simultaneous presence of negatively charged clay materials and the dipoles of water create electro-molecular forces, which attract and hold the dipoles of water. Clay particles are perceived as multi-layer formations. The charged surface and the distributed charge around the particles are together termed the diffuse double layer [39].

The space between layers and the free cations present in the interlayer space are the chief reasons for whether a particular clay mineral has a swelling behaviour. The free cations cause the absorption of water dipoles to the interlayer for chemical balance to be achieved. With decreased interlayer space, attractive forces among the layers become powerful and dominate the absorption

of water, so swelling does not occur. Conversely, as the interlayer space becomes more significant, interlayer forces are so that water absorption in the interlayer space can progress until interlayer bonds fail. Hence, the clay particles get separated and rearranged and swelling occurs. The extent and kinetics of the mechanisms described above depend on several other parameters acting individually or in combination. These include the density of the surface charge, the type and valence of the cations, the concentration of the electrolytes and the dielectric constant.

Therefore, swelling depends on the type of clay mineral. Generally, smectite minerals, with montmorillonite being a chief representative of this group, are considered swelling. Such smectites encompass repeat units of silica, gibbsite (aluminium hydroxyl groups) and adjacent silica layers joined by Van der Waals bonds (Fig. 1). The substitution of aluminium for magnesium in the gibbsite layer and for silicon in the silica layer results in a net negative charge in smectites, thus imparting a high cation exchange capacity. In addition to their high cation exchange capacity, the interlayer spacing between silica layers can increase in smectites through water absorption, causing substantial swelling. Conversely, clay minerals such as pyrophyllite, margarite and illite are considered non-swelling. For instance, kaolinites comprise repeat units of silica and gibbsite and lack the inter-layer spacing found between adjacent silica layers in smectites. Hence, their cation exchange capacity is less than that of smectites and is predominantly due to causes other than substitution [68].

Clay swelling is also controlled by the composition of the aqueous solution in contact with the clay. Due to cation substitution, the layered structure of clays is deficient in positive charges. Hence, interlayer cations are required to balance the negative layer charges. The distance between two successive structure units, known as the *d*-spacing, depends on the type of the exchangeable cation, the composition of the solution, and the composition of the clay. The *d*-spacing can be evaluated using X-ray diffraction. Thus, the swelling of clay can be assessed via the increase in the

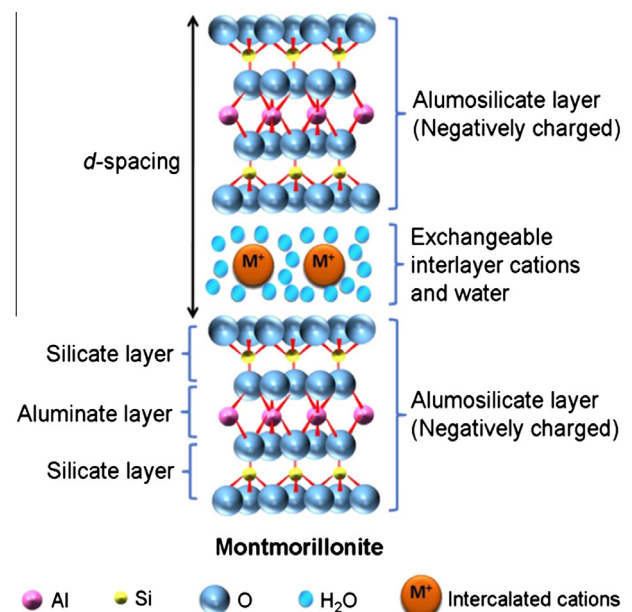


Fig. 1. Schematic illustration of the layered structure present in montmorillonite (courtesy of J. Plank, 2013).

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