

**CEMENT CHEMISTRY AND PHYSICS
FOR CIVIL ENGINEERS**

Cement Chemistry and Physics for Civil Engineers

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Preface

The steadily increasing specialisation in all branches of science is responsible for the fact that scientists working even in allied fields often have difficulty in understanding one another's work. This applies equally to the field of cement and concrete. A cement technologist is not necessarily an expert on concrete and, as is often the case, if the concrete technologist is, with due respect, even less likely to be an expert on cement, then he often has good reasons.

Cement is an artificially produced mixture of minerals of very complicated structure. In the presence of water, these minerals change to an equally complex colloidal product and are thus responsible for the phenomenon of hydraulic hardening. The chemical and physical natures of the hardening of cement are not fully understood even today. By using specialised tools of modern science, such as X-ray analysis, electron microscopy, colloid chemistry and capillary physics, such problems are studied today by cement research workers all over the world, in the hope of finding clues which would interpret the properties of cement more rapidly than is possible by means of the more orthodox, endless series of tests on mortar and concrete. But, this tendency to basic research has been responsible for making the chemistry of cement a closed book to the builder, engineer, reinforced concrete designer, as well as to the concrete technologist and material tester.

The present book should help to bridge this gap. Wherever applicable, an attempt is made to explain the properties of cement rather than to describe them in detail. The reader will be told not only what we know today about cement, but also what we do not yet know. He will learn what to expect of the behaviour of cement in varying conditions. The author assumes no more knowledge of chemistry and physics on the part of the reader than might be retained by him from his schooldays, but he is expected to have some familiarity with the practical characteristics of cement as used in building and some knowledge of the standard methods of its testing.

Even though some simplification is unavoidable and some gaps are left, the author considers it worthwhile to contribute in this way to a better knowledge of hydraulic cements.

Wolfgang Czernin.

TRANSLATION NOTE

The temperature figures in the original German text were rounded, approximate Centigrade figures. Their Fahrenheit equivalents, shown in brackets, give the impression of an extreme accuracy which is not intended in figures that should be treated as approximative.

Certain metric figures, especially those relating to laboratory calculations, have not been given British equivalents because of an increasing tendency to work in metric figures in particular instances.

Introduction

Structural cement belongs to the class of 'hydraulic cements', or cements which harden by reacting with water and give a water-resistant product. Portland cement, aluminous cement and slag cement are typical of this class of hydraulic cements, but not gypsum, which in spite of the fact that it hardens by reacting with water, cannot resist water over longer periods because it is soluble in it. Hardened lime is, however, water-resistant, but because its hardening depends on the absorption of carbon dioxide and not of water, lime is also excluded from the class of hydraulic cements.

The principal basic materials used in the manufacture of hydraulic cements are lime, silica, alumina and ferric oxide. It is with these that we shall deal first. A discussion will then follow on the compounds of the above constituents, which are mainly responsible for the development of hydraulic properties and, finally, methods will be analysed of producing these compounds on a large, industrial scale.

In the second part of the book, the properties of the more important types of cements are examined, with a view to their intelligent use.

CHAPTER ONE

Constituents

LIME

Lime is the classical building material known for many centuries. It is not difficult to imagine how lime mortar could have been invented: a camp fire was lit on a limestone rock; a sudden fall of rain extinguished the fire, and water soaked into the rock underneath, which suddenly disintegrated to a fine powder and formed a white slurry with the rainwater. On returning to the same place some days or weeks later, the slurry was found to have solidified and regained progressively its original stony appearance. The 'discovery' of lime mortar could have been made quite easily, in this or some similar way, thousands of years before the processes involved could be expressed in the form of the following chemical reactions:

1. $\text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2$ (lime burning)
2. $\text{CaO} + \text{H}_2\text{O} \rightarrow \text{Ca}(\text{OH})_2$ (lime slaking)
3. $\text{Ca}(\text{OH})_2 + \text{CO}_2 \rightarrow \text{CaCO}_3 + \text{H}_2\text{O}$ (carbonisation, hardening).

Expressed in words, these equations mean that during heating limestone (chemically: calcium carbonate, CaCO_3) first decomposes into quicklime (calcium oxide, CaO) and carbon dioxide (CO_2). At the temperature of 1000°C (1832°F), i.e. at red heat, this reaction proceeds quickly. As 1 lb of limestone contains about 0.44 lb of carbon dioxide, the quantity of the carbon dioxide gas that has to be expelled is quite considerable. The burnt lime left behind (quicklime) shows little change in its appearance in spite of the considerable loss in weight of 44 per cent. Depending on the impurities originally present in the limestone, its colour may be slightly altered; it may have become slightly lighter in colour or darker, acquired a yellowish tint and shrunk a little, but its porosity has increased appreciably because many vacant spaces have been left behind by the expelled molecules of carbon dioxide. When a lump of quicklime is immersed in water, these fine capillaries absorb water readily, like a sponge, and a violent reaction sets in and results in the formation of calcium hydrate, the process being known technically as slaking of quicklime and chemically as the hydration of calcium oxide.

This chemical hydration represents also a most efficient process of comminution. Because the volume of the hydrated lime produced in this way is larger by some 20 per cent. than that previously occupied by calcium oxide (quicklime), an expansive force is released which bears relentlessly upon each single molecule. (This expansive force will be discussed again in connection with "unsoundness of cement due to free lime".) Thus, within a few minutes, a hard lump of quicklime disintegrates to an impalpable white powder or, depending upon the quantity of the slaking water used, forms a highly dispersed paste known also as "milk of lime".

As we know, the slaking process is accompanied by a considerable evolution of heat. The heat of hydration of calcium oxide is about 280 cal/gm and hence 1 gm of this compound is able, during the process of slaking, to raise by 1°C the temperature of as much as 280 gm water. From this it becomes clear that very high temperatures can develop during lime slaking and that badly stored quicklime may be the cause of fire outbreaks, as has been often found in the past.

The individual particles of hydrated calcium present in slaked quicklime are extremely small: their average size is of the order of 2 microns (0.002 mm). Such finely divided materials are capable of developing considerable cohesive forces during a slow drying process; it is sufficient to mention the example of the high strength shown by a lump of dry, fat clay. 1:1 sand lime mortars have been known to give a compressive strength of some 427 lb/in² (30 kg/cm²) as a result of drying out, a value more than sufficient for ensuring a good bond in brickwork.

It should be remembered, however, that the actual hardening of lime mortar is caused by the carbon dioxide present in air; thus, the same material that was expelled from limestone during its burning penetrates slowly into the pores of the drying mortar and re-converts its lime to a stone-like calcium carbonate. The rate of this process depends on the porosity of the mortar and the quantity of carbon dioxide present in the atmosphere. Under conditions of artificial drying out of buildings by portable ovens, the pore formation is enhanced by a rapid removal of the excess water from the mortar, and a plentiful supply of carbon dioxide is available from the coke combustion.

On the other hand, rich lime mortars placed in the massive structures built in the Middle Ages have remained unchanged to a considerable degree up to the present day because, under such conditions, the penetration of mortar by carbon dioxide from the atmosphere has been extremely difficult.

A hardened lime mortar, as used in building construction, is a material of a relatively low strength: for this reason its elasticity is

quite high and its volume stability during repeated drying and wetting cycles is excellent, as should only be expected from an inorganic building material with a comparatively coarse structure. The constituents of hardened lime mortar can be seen quite clearly under a microscope: especially the small crystals of calcium carbonate and the pores they surround.

A completely different picture is observed if one examines the compounds formed by lime and silica. Here, we pass to the field of colloids, which are responsible for the high strengths of hydraulic cements.

SILICA

Silica (silicon dioxide, SiO_2) is found in most of our natural rock and stones; the forms of silica most often encountered in a more or less pure form are quartz, quartz sand, sandstone, etc. Silica of this kind is extremely stable. It is completely insoluble in water and is not attacked by acids, with the exception of hydrofluoric acid. When heated, silica undergoes a series of changes in its crystalline structure—a process which, in some cases, is accompanied by appreciable volume changes although the chemical composition of silica remains unaltered. If heated to 1900°C (3452°F) silica melts to a glassy mass known as silica glass. The permeability of silica glass to ultra violet radiation is worth noting, a property which is made use of in the making of ultra violet lamps. The volume of quartz glass remains very stable when subjected to changes in temperature. A redhot quartz glass crucible can, for instance, be plunged into cold water without danger of disintegration. This is because the expansion of silica during heating and the corresponding contraction on cooling are extraordinarily small.

In spite of the fact that silica in the form of quartz is a stable, unreactive, solid material, no such characteristics are shown by some of its other modifications. Representative of these other forms of silica are mainly such finely-divided, water-containing varieties as flint, opal and, especially, diatomaceous earth, which consists of weathering residues of extremely small siliceous skeletons of aquatic plants called diatoms. Because of their fine state of sub-division, all these forms of silica are far more reactive than the ordinary quartz. The various reactive forms of silica will be considered in detail in the section dealing with pozzolanic materials.

When heated to a high temperature, even the ordinary quartz variety of silicon becomes chemically reactive. For instance, strong alkalis, e.g. potassium or sodium hydroxide, react with quartz at a

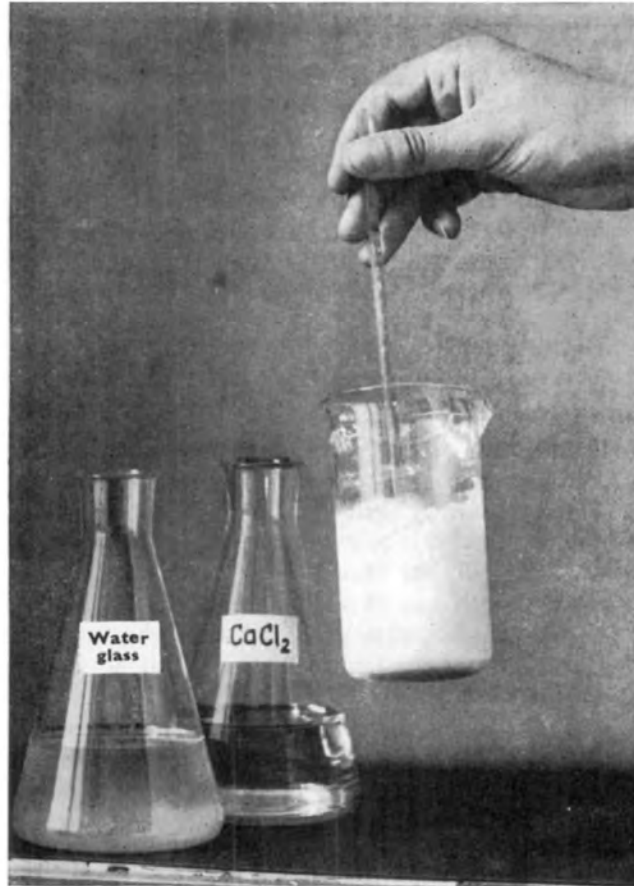


Fig. 1. *Two liquids form a gel-like mass: calcium silicate-hydrate*

high temperature to give a water-soluble product. This process is used in the manufacture of water glass, a viscous, colourless liquid employed by the housewife for preserving eggs. It has also a variety of industrial applications, such as wetting agents, fire-retardants for combustible materials, constituents of mastics, and so on.

As a silica-containing aqueous solution, water glass appears particularly suitable for demonstrating the processes occurring during the reaction of calcium oxide and silica, which is of such a fundamental importance in the process of hydraulic hardening. Since the compounds of calcium oxide and silica belong to a class of substances with a very limited solubility in water, a precipitate must form on mixing two solutions containing lime and silica respectively. In most

precipitation reactions occurring in highly concentrated solutions, the precipitate forms more or less rapidly and settles down at the bottom of the reaction vessel. This is, however, not the case with calcium silicate: when the two solutions are mixed there forms instead a solid material—although the term “solid” may be an overstatement, as we are dealing here with a gel-like mass. Nevertheless, the tensile strength and the cohesiveness of the mass are such that an attempt to withdraw the glass-rod stirrer from the mass results in the whole beaker being lifted together with its content. (Fig. 1.)

This process is known to chemists as gel formation, the term “gel” being used to describe a cohesive substance of individual colloidal particles. The word gel is derived from gelatine, a substance obtained in turn from bone glue by purification. The word “colloid” originates from the Greek “kolla”, which again means glue. Colloids are therefore glue-like substances and, since hydraulic hardening depends on the formation of a “mineral glue”, the colloidal state deserves close examination.

It is customary to define as colloidal such materials whose particle size lies between the dimensions of a molecule and those which are just perceptible under an optical microscope. We are therefore concerned with very small individual particles capable of remaining in a liquid in the state of suspension for an almost unlimited time. The characteristic properties of colloids are due to their extremely large surface area: the more finely sub-divided into smaller particles is a given quantity of a material, the higher the corresponding increase in its surface area.

While, for instance, 1 gm of sand with grains 2 mm. in diameter has a surface area of 10 cm², the total surface area of the same quantity of sand but ground to a particle size of 1 micron in diameter is increased a thousand times. The relation between the specific surface (surface area of 1 gm of the material) and its particle size is shown in Fig. 2, which was designed on the assumption that all the individual particles are spherical in shape. In order to cover the whole range of particle sizes present in an hydraulic mortar consisting of sand with a specific surface of the order of 10 cm²/gm and those found in a gel with a specific surface of 2-3 million cm²/gm, which corresponds to a particle size ranging from 10 cm to 100 Å (1 Å = 1 Ångstrom = 1/10 millimicron and 1 millimicron = 1/1000 micron), it was necessary to use in Fig. 2 a system of logarithmic co-ordinates. In this co-ordinate system, a plot can be made of specific surface areas of particles as large as those of sand grains, cement, slaked lime and also of the minute particles found in colloidal gels. All the individual curves are in the form of straight lines, slightly displaced in relation to each other because of the differences in the density of the

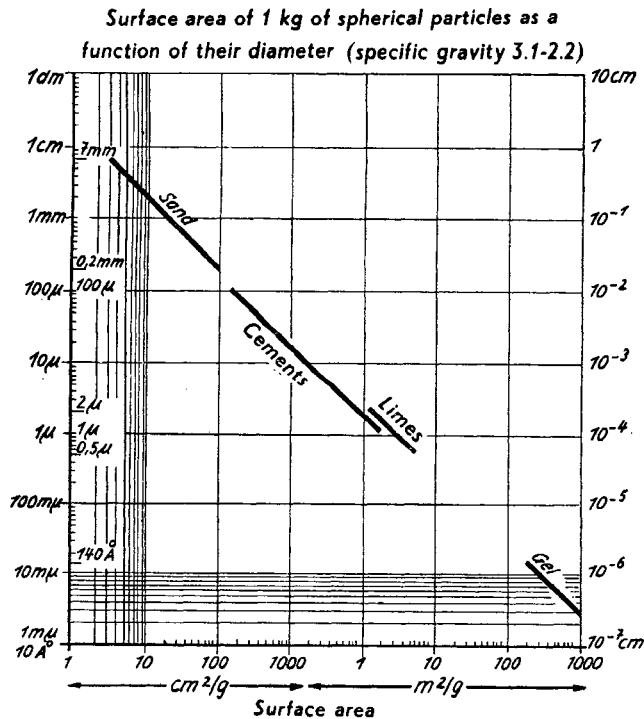


Fig. 2. Grain size and specific surfaces of mortar constituents

different materials. The conventional assumption that all particles are spherical in shape is, in fact, incorrect. Therefore, the graph gives only the theoretical mean values of the various surfaces.

The extremely large surface area of colloids does not, however, explain the stiffening observed in the solution of water glass on its mixing with a solution containing calcium salt. The understanding of this process requires an examination of the surface forces: each solid body owes its stability to the presence of molecular forces of mutual attraction between the individual constituent units. Inside the body of a solid material these forces, acting in all directions, cancel out, but unbalanced attractive forces exist on the surface of solids and attract and hold other materials. If, as in our water glass experiment, a solid colloidal substance is formed in a liquid, the tremendous surface area which it possesses comes into play, with the result that water molecules are drawn to the surface of the particles and held there strongly (adsorbed) thereby reducing very considerably the usual mobility of water. Furthermore, the above effect is accom-

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